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# Current trends in measurement and control of particle emissions from engines (perspectives from the 1st Workshop on Particulate Matter Emissions from Engine and Automobile Sources, 2 July 2012, Bielsko-Biala, Poland)

BOSMAL recently hosted the 1st Workshop on Particulate Matter Emissions from Engine and Automobile Sources, entitled Current trends in measurement and control of particle emissions from engines, which featured a series of speciallyselected presentations from experts on emissions of particulate matter from automotive sources, with both industry and academia represented. The workshop's technical programme consisted of one keynote address, five presentations and an expert panel discussion. In common with the emissions symposium hosted by BOSMAL somewhat earlier in the year, the Particulate Matter Workshop formed part of a series of events to commemorate BOSMAL's 40th anniversary. The event built upon and the achievements of BOSMAL's three emissions symposia hosted to date, but altered the format somewhat to cover a concrete subject in great depth. Some of the most important trends mentioned during the symposium included: problems encountered in accurately measuring particle emissions from vehicles, the particle mass and particle number metrics and the relationship between them, particle size profile and surface area and aftertreatment systems for elimination of particles (including for direct injection petrol engines).

Key words: IC engine, particulate matter emissions, nanoparticles, particles mass and number, particles measurement

#### Introduction

Concern over the impact of vehicles on air quality remains high. Emissions of particulate matter are coming under increasing scrutiny as a form of pollution with wide-ranging negative impacts ranging from asthma to climate forcing. While the study of particles has traditionally focused on mass-based quantification of emissions from compression ignition engines, there is now a growing awareness that other engine types (direct injection, port fuel injection) and other metrics and quantification methods (particle number, particle size distribution, particle surface area) are all worthy areas of investigation. In comparison to gaseous emissions, particle emissions still present multiple open questions and large domains of investigation. There are also considerable practical difficulties involved in investigating solid nanoparticle emissions from engines. Despite this, modern aftertreatment systems for reducing emissions of particulate matter have proven effective, although there remains much room for improvement. The introduction of a particle number standard for Diesel vehicles in Europe marked a significant change in the way that particulate emissions are regulated and caused many changes to Diesel engines and aftertreatment systems. The coming introduction of an equivalent limit for direct injection petrol engines is sure to exert similar impacts on that engine type. New aftertreatment systems must sit alongside – and show full compatibility with – existing systems, so that both gaseous and solid emissions can be controlled simultaneously. Concern that laboratory testing may drastically underestimate real-world emissions also extends to emissions of solid particulates. The link between emission of particulate substances and poor air quality is well established scientifically and the situation is of growing interest to politicians, legislators and even the general public.

2011 [3, 4] and 2012 [3, 4], BOSMAL Automotive Research and Development Institute Limited (of Bielsko-Biala, Poland) recently hosted the 1st Workshop on Particulate Matter Emissions from Engine and Automobile Sources, held on 2 July 2012. This workshop was hosted as a result of the successes of the previous emissions-related technical conferences hosted by BOSMAL: the 1st, 2nd and 3rd International Exhaust Emissions Symposia [1 - 6]. Since these previous events [1 - 6] had included excellent presentations on topics related to particulate matter emissions, it was decided to host a specialised event on this subject. The workshop was organised in collaboration with Professor Jan Czerwinski (AFHB, Berne University of Applied Sciences, Biel, Switzerland) and Dr Andreas Mayer (TTM, Switzerland). Symposium delegates, representing a total of fourteen organisations (eleven firms and three universities), hailed from a total of seven countries with both Europe and North America represented, testifying to the strongly collaborative nature of the event. The workshop featured a keynote address from a specially selected expert, Professor David Kittelson from the University of Minnesota, a world-famous pioneer and specialist in research on nanoaerosols from engines. A further five presentations made up the workshop's plenary session and five additional papers were featured as writtenonly submissions and archived in the symposium proceedings. The expert panel discussion also represented a highly important part of the technical programme. On Monday 2 July, workshop organisers Dr Piotr Bielaczyc and Professor Jan Czerwinski commenced proceedings by greeting the delegates and delivering some opening remarks. Dr Bielaczyc welcomed the delegates to BOSMAL and offered a

Following the highly successful 1<sup>st</sup>, 2<sup>nd</sup> and 3<sup>rd</sup> International Exhaust Emissions Symposia hosted in 2010 [1, 2], few remarks on the nature and format of the event, as well as thanking the sponsor of the event (AVL, Austria) and the symposium's media partner (Combustion Engines, published by the Polish Society of Combustion Engines, PTNSS). Professor Czerwinski's opening remarks highlighted the importance of this type of event, given the importance of the topic addressed. Nanoparticles (NP) are generally recognized as particularly toxic due to the easy penetration through the air-blood-barrier in the lungs and even through the olfactory nerves. The current exhaust emission legislation for on-road vehicles has started to limit PN emissions in addition to the particle mass (PM). It is very important for all specialists and laboratories dealing with automotive emissions to work with this new component and with the specific measuring techniques. It is only a question of time until NP emissions will be limited for DI-gasoline engines, followed by all other types of engines. This parameter is roughly 1000 times more sensitive than measuring exhaust gas opacity and it reveals NP originating from the lubricating oil and from different oil and fuel additive packages. The workshop summarized in this paper represented an excellent opportunity for the exchange of knowledge and networking in this new domain. The workshop's organizing committee were delighted to welcome Professor David B. Kittelson from the University of Minnesotta, a worldwide known specialist, as a keynote speaker, together with Professor Jerzy Merkisz from Poznan University of Technology, president of the Polish Scientific Society of Combustion Engines (and the editor-in-chief of this journal).

## Presentation abstracts and selected key slides

*NB: the authors listed here are presenting authors only. See the Workshop's Technical Programme (Fig. 7) for the full listings of all co-authors of each presentation, where present.* 

#### Keynote address: Professor David B. Kittelson, University of Minnesota (USA) – Issues associated with solid particle measurement

Regarding Diesel applications, the use of a DPF dramatically reduces particle emissions; DPFs are generally extremely efficient for most size ranges. What little particles remains in the exhaust gas is mainly composed of volatiles - but such particles are so hard to measure accurately and reproducibly that it was decided to deliberately exclude them from the measurement. A number of mass- and number-based standards have been introduced for automotive particulate matter emissions. In every case, the number-based standards are much stricter. The current 23 nm cut-off might appear to be quite arbitrary (Figure 1); aircraft may soon be subject to emissions limits for particles as far down at 10 nm, since a large proportion of such particles are < 23 nm in diameter. Particles of size around 23 nm are effectively removed by the filter, regardless of its loading. Even the removal of volatile particles is in fact extremely difficult – certain types are very resistant to temperature and other parameters used to remove such material. A catalytic stripper system may be more effective than a volatile particles remover (VPR),

although the  $\Delta T$  of the system does cause a small sample loss. However, this sample loss is quantifiable and a correction factor can be applied. Over the years, perfection of the design of the catalytic stripper system has helped to reduce losses and make them more constant and correctable. Testing with a synthetic mixture of HC and sulphates revealed that the system is extremely effective at eliminating volatiles. Investigations into the root cause of particle formation have revealed that metal-based additives in the oil and fuel are the cause of the generation of large numbers of tiny particles. Metal oxides appear to play a central role and transmission electron microscope with an electron probe can reveal the elemental compositions of tiny particles (though not without considerable practical difficulties). The size distribution of such particles is not always continuous; distributions can be bi-modal, with a low band over the size range 11 - 23 nm low, but with large numbers of particles at lower diameters (Fig. 1). The fact that different instruments and different setups can return different results is a real concern and has been a major factor in US reluctance to turn to a number-based standard. Agreement between different systems depends on system operating conditions, temperatures, dilution settings, etc. Any observed dependence on the dilution ratio suggests particle formation downstream of the tailpipe or incomplete/inadequate removal. It is important to remember that solid residue to can be produced from nothing but volatile material – i.e. gas+gas $\rightarrow$ solid reactions can (and do) occur. Currently, relatively strict particle size cut-off points are specified. Changing the cut-off point by as little as a few nm can change the results by an order of magnitude, as demonstrated graphically in Figure 1.

Engine out, light-load, low soot conditions: Most of the number emissions are solid with Dp < 23 nm



Fig. 1. Size distribution of solid particulates from a Diesel engine presented in comparison to the PMP 23 nm CPC cutoff point

#### **Plenary session**

# Manfred Linke, AVL (Austria) – Measurement of volatile and non-volatile particles

Ultrafine particles are now subject to a level of concern that was once shown regarding gaseous emissions. Measures to reduce PM (mass) emissions may have inadvertently increased number emissions, so investigation of number emissions widespread. For Diesel applications, DOCs alone may increase volatile particle emissions but this phenomenon depends on the engine operating conditions and the fuel sulphur content. Nucleation mode particles are desirable to measure, since PN emissions of this particle type can be very high, particularly at high speed, but there are multiple practical barriers to accurate measurement of this particle type (Fig. 2). For this reason European legislation introduced the measurement of the non-volatile particles. The roundrobin evaluation performed as part of the development of the legislative procedure for quantification of PN confirmed that the new non-volatile particle method is robust method for regulation. The challenges presented by light-duty and heavy-duty applications are related but subtly different. The upcoming particle number limit for GDI engines will be hard to fulfil - but the phase-in period will allow time for research to be performed which should at least begin to overcome these problems. Most of the mass collected on the filter during an emissions test of DPF equipped vehicles is in the form of volatiles, which depend very strongly on the sampling conditions; measuring only the black carbon fraction gives a better correlation between PM and PN at these low emissions levels. Other key topics for quantification of particle number remain unresolved (analyser linearity, calibration procedures, drift, etc). A move towards including particles of smaller diameter in the measurement is likely, and fortunately this could be achieved with relatively little difficulty. In future, calibration will be performed at lower particle sizes and corrections will have to be made for particles losses in the volatile particle remover (VPR).



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Fig. 2. Nucleation mode particle size emission profiles for two vehicles at constant speed

#### Dr Amanda Lea-Langton, University of Leeds (UK) – Effects of Oxygenated Biofuels on Particulate Emissions from Diesel Combustion

Ambitious targets for improving fuel life-cycle GHG in the EU mean interest in biofuels has increased rapidly in recent years. Biodiesel is of particular interest and various types and blends are currently under investigation. Given the multiple physicochemical differences between fossil Diesel and biodiesel and mindful of the impact of particulate matter emissions, research on particulate substances emissions from biodiesel blends is an increasingly important research topic. A series of experiments were performed using a 6-cylinder engine operating at two fixed points. Thermogravimetric analysis was used in addition to gravimetric PM analysis; an ELPI particle sizer was used to measure the particle size distribution. Significant differences were observed in the number and mass distributions for three different fuels, both upstream and downstream of the DOC. The effectiveness of the DOC in removing particulate matters varied strongly with the engine operating point and between the three fuels. Emission of polycylic aromatic hydrocarbons (PAH) is of great concern, and it is of interest that biodiesel contains no PAH - in contrast to standard Diesel, which contains up to 11% PAH. However, PAH of pyrolitic origin is still a concern regarding biodiesel. Danger to human health is proportional to molecular mass (and therefore the number of rings). Concentrations of unburned fuel products are lower at higher loads (and therefore higher engine temperatures). Biodiesel and rapeseed oil showed lower PAH emissions than standard Diesel. Prolonged usage of rapeseed oil causes substantial deposits to build up, which cause PM emissions to gradually increase, although certain detergents can eliminate this problem. Oxygen enrichment of the intake air can improve matters significantly, by promoting more complete oxidation of hydrocarbon species, but such an approach is better suited to non-mobile engines (power generation, etc), for reasons of practicality. (It should also be noted that increasing the oxygen level was found to increase NO emissions). In order the further investigate pyrolysis (the first step in the combustion reaction) of the three fuels, a micro-pyrolisis reactor was used in conjunction with a gas chromatography/mass spectrometry system.

# Similarity of products at 900°C



Similar range of PAH observed. Diesel fuels high relative response for naphthalene and phenanthrene indicate an additional formation route.

This is probably due to dealkylation reactions of alkyl-PAH present in the original fuel sample.

Lower abundance of aromatics for biodiesel suggests that this is due to the oxygen in the fuel.

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Fig. 3. Gas chromatography/mass spectrometry analysis of pyrolysis products of three fuel types following treatment at 900 °C

As the temperature was increased from 700 - 900 °C, the chemical speciation changed from that of the original fuel to include a large number of PAH species. At 900 °C,

almost the only hydrocarbons left are aromatics, some of which are very large indeed. However, the aromatics profiles for biodiesel, diesel and hexadecane showed differences. Generally, the species present are exactly the same, but concentrations of the larger PAH molecules are significantly lower for biodiesel, as shown in Figure 3.

This implies that the chemical makeup of biodiesel (and particularly its oxygen content) makes the fuel less predisposed to PAH formation.

## Dr Piotr Bielaczyc and Joseph Woodburn, BOSMAL (Poland) – Particle mass and number emissions from a range of European light-duty vehicles

BOSMAL has 40 years' experience in dealing with automotive emissions and almost 15 years' experience in dealing with emissions of particulate matter from engines and vehicles. Currently, BOSMAL performs a wide range of testing activities on engines, vehicles, fuels and aftertreatment systems and an increasingly important aspect of such research is quantification of particulate matter emissions. To that end, BOSMAL has facilities for performing gravimetric analysis of such emissions, as well as a fully PMP-compliant system for measuring solid particle emissions from vehicles. Gravimetric and number based emissions results are powerful tools for investigating engines, fuels, aftertreatment systems, particularly (but not exclusively) for Diesel engines and fuels and direct injection petrol engines. PM and PN results can be used to compare fuels, both in terms of legislative emissions limits, as well as meritocratically, whereby a direct comparison is made between different fuels or fuel blends. Interest in biofuels is increasing and since such fuels generally differ chemically and physically from fossil fuels, PM and PN measurements form an important part of the armoury of tests that can be used to assess the environmental performance of biofuels and biofuel blends. It is tempting to compare PM and PN, and thereby derive characteristics such as particle mass and even mean diameter. However, closer examination of the sampling conditions and measurement techniques reveals that the two methods measure quite different things and therefore PM and PN cannot be directly correlated (Fig. 4). However, with these caveats in mind, the two metrics can be compared. Where PM is sufficiently high (for some Diesel engines and direct injection petrol engines), a linear trend may be observed. However, this trend does not apply in all cases. Modern Diesel engines featuring DPF systems are easily able to meet the PM limit; the margin by which their PN emissions lie under the limit varies by orders of magnitude. Research performed by BOSMAL has shown that while mass emissions from modern direct injection engines are reasonably low (generally below 5 mg/km, at least over the NEDC), while PN emissions are very high. Thus, the upcoming PN limit for this engine type will force the use of some form of aftertreatment system (either a GPF or a continuouslyregenerating POC). Emissions limits are unlikely to remain static and further reductions in the limits are anticipated in the coming decades. Additionally, the specified methodologies for measuring PM and PN may well change. Furthermore, particle emissions limits may eventually be introduced for all engine types, regardless of injection strategy or fuel type. The result of these trends is that research facilities (and indeed expertise) for measurement of gaseous and solid pollutants are vital for R&D activities on virtually all engine types.

How comparable are PM and PN?



Fig. 4. Simplified scheme of the route taken by PM and PN samples, showing the dependency of the relationship between PM and PN on the sampling point and sample treatment process

## Dr Paul Zelenka, VERT Association (Austria) – Benefit/cost analysis when using emission control devices for IC engines

Various ATS can provide benefits to society, such as improved air quality, avoided climate change, etc - but at what financial cost, and who is to pay for these benefits? Swiss law contains a stipulation that the cost of such systems must be in reasonable proportion to the benefit. In order to compare apples to apples (i.e.  $\in$  to  $\in$ ), a price has to be put on the benefits, as well as the cost. Obtaining a value for cost is relatively straightforward - and the result is already in the required unit (e.g. €). Different assessments of the costs of PM exposure have generated different 'prices'. The assumptions and methodology used to perform such an assessment influence the value ascribed to the benefits - and therefore the cost-benefit quotient. Various factors can be considered, not least of which include deaths from particulate pollution and reduced life expectancy, but also lost work days, etc. These vary from country to country and should be carefully selected. The cost efficiency is strongly dependent on the uncontrolled emissions from the engine and therefore to the age of the vehicle/engine, as well as the engine type (Fig. 5). For particulate matter emissions, the situation is further complicated somewhat by the use of two metrics – particle mass and particle number. Health effects of these two metrics have been studied, but the value of the health benefit is not the same for PM and PN. Using a PM10 metric may not adequate capture the health risk of the pollutant. Research has indicated that purely carbon-based PM species are among the least mutagenic and that particles based on zinc and copper

oxides may be far more mutagenic. It was once thought that Diesel PM limits could be met without the use of a filter, but in the end public pressure forced manufacturers to start to fit filters and the introduction of the PN limit eventually made this completely unavoidable. The same situation could occur with regard to vehicles featuring direct injection petrol engines. The GPF will likely be integrated into the TWC, rather than installed as a completely separate system. Analyses reveal that heavy-duty retrofit options are considerably more expensive than light-duty retrofit options. The global warming potential of carbon black is some 1600 times higher than that of  $CO_2$  (kg for kg) and this should be taken into account when analysing the global warming potential of vehicular exhaust gas. (However, when the residence time is factored in, the ratio falls to around 1:2000). Measures that reduce an engine's particulate matter emissions could have a larger positive impact on the global warming potential of the exhaust gas than measures that reduce the CO<sub>2</sub> emissions from the same engine.

# Health Benefit of Diesel LDV versus Gasoline based on <u>soot particle mass</u> PM

	Diesel+FFF	Gasoline+FFF
PM-Emission (Euro 3 or in use)	50 mg/km	10 mg/km
Mileage per anno	10'000 km pa	10'000 km pa
Average Performance [kW]	10	10
PM Emission [kg/year]	0.5	0.1
Overall vehicle life [year]	10	10
Emission [kg/vehicle life]	5	1
Filter type	wall flow	wall flow
Filter efficiency [%]	99.9	99.9
Health Cost [€/kg soot]	1'200	1'200
Total prevented soot [kg/life]	5	1.0
Health Benefit [€]	6'000	1'200

Fig. 5. An example of the benefit quantification methodology for use in an emission control system benefit/cost analysis

#### Professor Jan Czerwinski, AFHB (Switzerland) – VERTdePN: A Swiss quality control for exhaust aftertreatment systems (DPF+SCR)

Given the level of concern over particulate emissions and the number of manufacturers offering Particulate matter ATS solutions, a robust quality control procedure is required in order to ensure that these systems lead to actual reductions in emissions. Combining a DPF with an SCR system makes sense for a number of technical reasons, but can complicate matters somewhat regarding emissions and so the VERTdePN procedure aims to perform quality control checks of both parts of the system. An SCR system requires the input of ammonia; the ratio of ammonia supplied to the amount required for stoichiometric reduction of NO<sub>2</sub> is termed  $\alpha$ . The occurrence of ammonia slip is strongly dependent on the value of  $\alpha$ . A test procedure is defined for quality control processes of filtration devices, involving both engine test bed measurements and field (on-road) durability testing [7, 8]. For some laboratory investigations, measurements are performed at three sampling positions: upstream of the particle trap, between the trap and the SCR and downstream of the SCR system (including any NH, slip catalyst), as shown in Figure 6. Low load cycles can lead to limited SCR system efficiency and testing has indicated that the test cycle profile and the resulting exhaust gas temperatures have a strong effect on the performance of the system. The same metric is used to quantify the efficacy of pollutant removal, regardless of whether the test cycle involves transient or steady-state operation. There are some indications in the literature that an SCR system can reduce particle concentrations. While this effect has indeed been observed, it should be recalled that a TWC or DOC can also reduce particle number concentrations to a similar extent. This reduction depends on the engine operating point and can often be low-to-negligible. In short, 'conventional' SCR, DOC and TWC systems cannot be relied on to significantly reduce particle mass or number. On the other hand, a DPF, which fulfils the VERT criteria has excellent particle count filtration efficiencies (up to 99.9%) both at stationary and at dynamic operation. The international network project VERTdePN (de-activation, decontamination, disposal of particles and NO<sub>2</sub>) has established quality verification procedures and standards for SCR-, or combined DPF+SCR-systems for retrofit applications. On-vehicle SCR testing is of high importance and different testing procedures (including a simplified low-cost check) are under development.



Fig. 6. Measurement setup for measuring gaseous and solid emissions from an engine equipped with a DPF+SCR system, in order to evaluate the effectiveness of the ATS

#### **Expert panel discussion**

This discussion took the following format: pre-prepared questions contributed by Professor Merkisz and his team (Poznan University of Technology, Poland) were read to the audience in general, and the various experts present expressed their views and considered opinions in response to the questions. These responses in turn generated further comments and questions, so that each initial question fanned out into a broader discussion. The discussion was moderated by Dr Piotr Bielaczyc and the following experts participated: Professor David Kittelson, Professor Jan Czerwinski, Dr Amanda Lea-Langton (University of Leeds, UK), Dr Paul Zelenka (VERT Association, Austria), Douglas Trombley (GM Powertrain, USA), Manfred Linke (AVL) and Joseph Woodburn. The questions posed are reproduced below, together with a synopsis of the answers.

**Question 1:** Under which conditions should particulate mass and number be compared? Do the different ways of measurement exclude the possibility of such a comparison? On one hand, the gravimetric method requires the measurement of the mass of the particles deposited on the filter (all carbon, including liquid hydrocarbons and ash) and on the other hand, the particle number count is determined exclusively by the measurement of the carbon fraction of particulate matter. In each of these cases the density of particulate matter is different. What are the recommendations as regards the above actions? (The data published on this point vary widely).

There are multiple differences between PM and PN, considering their structures, visibility, measuring techniques and health effects. Apparatus which recalculates PM from PN measurements cannot deliver exact an PM value, but only a reference value according to certain models. The experts and the audience in general were generally of the opinion that PM & PN should be considered to be two different parameters which are not comparable. (One could even go so far as to say that PM and PN are "completely different worlds".) It was pointed out that a sample and helpful way to underline this difference is to use different symbols for the two metrics: PM & PN (mass & number). Since particulate matter emissions from vehicles are a flavor of the moment with politicians, politically is it advisable to separate PM and PN in order to avoid conflation and unnecessary complexity. The real problem is that the gravimetric particulate matter measures mass collected on a filter and for very low emission engines much of this mass may be semi-volatile artifact, not true suspended paerticles. On the other hand, when it works correctly the PMP PN measurement measures solid accumulation mode particles, true suspended particles. Even if the filter measurement technique had no artifact and measured true suspended PM, the relationship between mass and number would be influenced by the presence of volatile material, particle density, and particle size.

**Question 2:** In light of the current EU actions introducing limits for particulate matter for vehicles fitted with direct injection spark ignition engines, what is the enforcement of the admissible minimum values going to be like for both particulte mass and number under actual operating conditions? (This question pertains to the possibility of validating such emissions by both the OBD system and during periodical technical inspections).

Due to the health effects the PN values of GDI engines will be limited at the same level as Diesel engines (in the long term). The possibilities of validating such emissions via the OBD and during periodical technical inspections are under development. When a DPF or GPF is fitted to a vehicle, PN emissions are so low that an opacimeter is ineffective at measuring such emissions A CPC+VPR system could be used, but the cost of such a system is very high. Fortunately, various cheaper alternatives are available and systems of this type are currently under consideration. Regarding onboard measurements, the OBD system has the capability to monitor emissions, but on-board quantification of particle number is much less challenging than measuring particle mass on-board. The U.S. is at present not considering limiting particle number for any engine type (except for aircraft). However, the proposed PM limits of 3 mg/mile and ultimately 1 mg/mile (i.e. 1.8 mg/km and 0.6 mg/km) will challenge GDI engines, even without a number standard in place. Such moves will make GPFs a necessity for direct injection SI engines of the future. While the idea of passenger cars featuring GPFs may still appear far-fetched and somewhat unlikely, the DPF was once in a similar situation, but has now become commonplace. Acceptance of the GPF concept by the industry and the car-buying public will take time. However, given the low opinion of Diesel engines held by many in the USA (including CARB), GDI concepts are an attractive option, notwithstanding the need to develop and implement new aftertreatment technologies.

**Question 3:** Is giving up of the measurement of the PM (mass) (maintaining the measurement of the PN (number)) considered in favor of the size distribution of its diameters i.e. instead of the mere counting of all the particles, counting of the PN along with a relevant measurement of the particulate diameter would take place.

Giving up the gravimetric PM-measurement for homologation of new engines is possible in the long term. The measurement of PN in certain size range (SMPS) (currently set at 23-300 nm) is sufficient to guarantee the required low emission level, but SMPS measurements require a steady aerosol. Measurement of the size distribution with 'fast sizing' instruments like a DMS500 or an EEPS can be done during transient testing and would give good results, but these instrument are expensive and too complex to be used in routine testing. The current PMP PN measurement technique (CPC) of total number larger than a certain size (23 nm) PN, with no regard to the PN size distribution, enables dynamic measurements to be performed and is recognized for legal purposes (GRPE PMP) Another possibility is to use instruments that measure the active surface area (like the TSI NSAM or the Matter Aerosol miniDiSC). Indeed, many health specialists suggest that surface area might be a better indicator of health hazard than either particle number or mass, since surface area correlates most closely with biological response. These instruments are fast-operating and could, at least in principle, be used instead of a CPC in a PMP-type system However, concern has been expressed over

what such instruments actually measure – the real surface area of the particle or the surface area of the 'envelope' in which the particle is contained. Such considerations may appear trivial, but the scale (resolution) at which the particle surface area of is measured can change the result by orders of magnitude. The situation in the USA is somewhat different. Authorities there have been reluctant to adopt a number-based standard due to concerns over accuracy and repeatability. The Clean Air Act specifies that mass-based metrics must be used to quantify particle emissions; changing this to include any kind of number-based standard would involve negotiating significant legal hurdles.

**Question 4:** What is the opinion of the panel on the comparison of the laboratory measurements and measurements from actual operating conditions (i.e. on-road) when analyzing particulate matter emissions (nanoparticles in particular)? Which type of measurements are (or will be) a priority in planned exhaust emission legislation? What are the arguments for the introduction to the legislation of on-road exhaust emission tests? Do these steps imply that the actions aiming at introducing diagnostic systems on vehicles and their on-going control are insufficient or unsuccessful?

Laboratory measurements of solid nanoparticles (PN) during dynamic legislative tests are representative of the real world operation. The arguments for introducing PN in the legislation are – first and foremost – the health effects. The measuring systems and procedures are already regulated in UN ECE Regulation No. 83. Current diagnostics and control systems are sufficient for the present situation, but they are to be adapted to the new requirements in the sense of technical development. The question becomes more complex if it is decided that volatile nanoparticles should also be measured. The formation of such nanoparticles is extremely sensitive to sampling and dilution conditions and any method would need to be validated against real-world ambient dilution experiments. An alternative approach to this problem could be to measure the precursors to volatile nanoparticle formation like sulfuric acid, heavy hydrocarbons, organic acids, etc, and then calculate a 'volatile particle formation potential' index, or some other similar metric. Planned changes to introduce PEMS testing in the EU will apply to various types of emissions, including PM and PN.

## Conclusions

Internal combustion engines have been, are and will be the main solution for transportation for the foreseeable future [5]. However, the ICE concept has certain key existential challenges to overcome; one of the greatest of which is dealing with particulate emissions. The particular challenges can depend somewhat on the engine type and fuel, but other challenges apply in all cases. Technology related to particle emissions and their control current moves too fast for legislation to keep up. In the end, it appears likely that particle emissions will be limited for all vehicles featuring ICE, regardless of combustion strategy or fuel type. As was mentioned multiple times during the workshop, correlating PM and PN is at best difficult and error-prone and at worst misleading and unscientific. Therefore, it should be expected that both mass- and number-based metrics will continue to be used in the EU; whether other markets (e.g. the USA, Japan) introduce a PN limit remains to be seen. The current PN measurement method is relatively young and possibly unrefined and the current PM method is very close to (or possibly even below) the limit of detection for accurate, repeatable measurements. For these reasons, changes to the test methodology are likely at some point in the future. The next few years will be a relatively rocky road for direct injection petrol engines, at least for the European market, but ongoing R&D work will likely overcome the particulate matter related problems for this engine type. The impact of the cost of such systems on sales remains to be seen (see [9] for a recent discussion of the cost of various vehicular ATS technologies, including GPFs), although the impact of improved ecological credentials should also be taken into consideration.

While operation of passenger cars and other road vehicles is in fact only one aspect of a range of human activities that degrades air quality through emission of nanoparticles, the automotive industry is now subject to controls and these controls will likely become far stricter in the future. Increasing interest is being shown in air quality issues by politicians, lawmakers and even the general public; particulate matter has made the transition from an 'outsider' (a pollutant of relatively little importance to engineers, politicians or the general public) to a key concern regarding nothing less than the fitness for purpose of the very air we breathe. As such, the subject area addressed by the workshop is a vital research direction, with dimensions and implications that extend far beyond the emissions testing laboratory.

The success of the workshop echoed previous automotive emissions events BOSMAL has hosted [1-6]. The workshop and its social programme were well-received and considerable interest was expressed in attending future events of the same type. BOSMAL continues to perform research on this subject; various publications are forthcoming, to be featured both in this journal and elsewhere.

The proceedings from the 1st Workshop on Particulate Matter Emissions from Engine and Automobile Sources have been archived on a CDROM entitled '*Particulate Matter Emissions from Engine and Automobile Sources*', ISBN: 978-83-931383-3-3. This CD-ROM is attached to this issue of Combustion Engines/*Silniki Spalinowe*.

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# Programme

9:00 - Workshop opening

1. 09.15-10.00 - Keynote lecture: <u>Prof. David B. Kittelson</u> – University of Minnesota, USA, Jacob Swanson – University of Cambridge, UK, Heejung Jung – University of California, USA. Issues associated with solid particle measurement

#### **Plenary session**

Chairs: Professor Jan Czerwinski, Switzerland, Dr. Piotr Bielaczyc, Poland.

2.10.00-10.30 - Manfred Linke, Dr. Alexander Bergmann, Dr. Barouch Giechaskiel, Dr. Michael Arndt AVL, Austria. Measurement of volatile and non-volatile particles

3.10.30-11.00 - <u>Amanda Lea-Langton</u>, Gordon Andrews, Hu Li - University of Leeds, England Effects of Oxygenated Biofuels on Particulate Emissions from Diesel Combustion

11.00-11.30 - Coffee Break

4.11.30-12.00 - Dr. Piotr Bielaczyc, Dr. Piotr Pajdowski, Dr. Andrzej Szczotka, Joseph Woodburn BOSMAL, Poland Particle mass and number emissions from a range of European light-duty vehicles

5.12.00-12.30 - Dr. Andreas C.R. Mayer – TTM, Switzerland, <u>Dr. Paul Zelenka</u> – VERT Association, Austria, Prof. Jan Czerwinski – AFHB, Switzerland.

Benefit/Cost-Analysis when using Emission Control Devices for IC Engines

6.12.30-13.00 - <u>Prof. Jan Czerwinski</u> – AFHB, Switzerland. VERTdePN A Swiss Quality Control for Exhaust Aftertreatment Systems (DPF + SCR)

13.00-14.00 - Expert Panel Discussion

14.00-15.00 - Lunch

15.00 - Visit to BOSMAL's testing laboratories

16.30 - Workshop Summary & Closing

17.00 - Guest transfer to Krakow

#### Written papers (no oral presentation):

Dr Andreas C.R. Mayer – TTM, Switzerland. Worldwide Experience with DPF-retrofit

8. Wolfgang Thiel – Technical University Munich, Germany. Measuring PM or PN in Automotive Exhaust Emissions – A Big Challenge

9. Joseph Woodburn – BOSMAL, Poland A Graphical Literature Survey of Studies Evaluating the Effect of Ethanol in Petrol on Particulate Matter Emissions from Light-Duty SI Engines

10. Prof. Jerzy Merkisz, Dr. Jacek Pielecha - Poznan University of Technology, Poland On-board Particle Mass and Number Emissions Measurement from Light Duty Diesel Vehicles

11. Piotr Bielaczyc, Jerzy Merkisz, Piotr Pajdowski, Joseph Woodburn A Comparison of the Results Generated by two Commercially Available PMP-compliant Particle Number Counting Systems

Fig. 7. The technical programme of the 1st Workshop on Particulate Matter Emissions from Engine and Automobile sources

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#### Abbreviations and definitions

ATS	Aftertreatment	system
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- CI Compression Ignition
- CNG Compressed Natural Gas
- GTL Gas-to-liquid
- CO Carbon monoxide
- Carbon Dioxide CO,
- DOC Diesel Oxidation Catalyst
- Diesel Particulate Filter DPF
- EGR Exhaust gas recirculation EPA
- Environmental protection agency
- EU European Union
- FAME Fatty-acid methyl ester GDI Gasoline Direct Injection
- GHG Greenhouse gas GPF
- Gasoline particulate filter HC
- Hydrocarbons
- HCCI Homogenous charge compression ignition ICCT International Council on Clean Transportation

- Lean NOx Trap LNT
- NEDC New European Driving Cycle
- NH. Ammonia
- NO Oxides of nitrogen
- NP Nanoparticles
- Polycyclic aromatic hydrocarbons PAH
- PGM Platinum group metals
- PM Particulate mass
- PMP **UN-ECE** Particulate Matter Programme
- PN Particle Number
- POC Particulate Oxidation Catalyst
- SCR Selective catalytic reduction
- SI Spark ignition
- UN-ECE United Nations Economic Commission for Europe
- VERT Verification of Emission Reduction Technologies (See: www.vert-certification.eu, www.vert-dpf.eu)
- Volatile particles remover VPR
- WLTC World Harmonised Light Duty Vehicle Test Cycle
- WLTP World Harmonised Light Duty Vehicle Test Procedure

### Analiza trendów rozwojowych dotyczących pomiarów i ograniczania emisji cząstek stałych z silników spalinowych

Słowa kluczowe: silnik spalinowy, emisja cząstek stałych, nanocząstki, masa i liczba cząstek, pomiar cząstek

Zmniejszenie emisji związków szkodliwych i toksycznych spalin silnikowych, jak również zmniejszenie globalnej, antropogenicznej emisji CO, są głównymi wyzwaniami dla przemysłu motoryzacyjnego, spowodowanymi czynnikami politycznymi, ekonomicznymi i technicznymi. Coraz większe znaczenie ma również ograniczanie emisji cząstek stałych (PM) obecnych w spalinach nie tylko silników o zapłonie samoczynnym (ZS), ale także o zapłonie iskrowym (ZI), szczególnie wyposażonych w układ bezpośredniego wtrysku paliwa. W ostatnim czasie zwraca się szczególną uwagę na ograniczanie emisji cząstek o małych średnicach – nanocząstek, przez wprowadzenie limitów emisji dotyczących masy emitowanych cząstek, a także ich liczby, a w przyszłości także ich powierzchni całkowitej.

Instytut Badań i Rozwoju Motoryzacji BOSMAL sp. z o.o. w Bielsku-Białej był organizatorem, przy współpracy z prof. Janem Czerwińskim z Laboratorium Silników Spalinowych (AFBH) Uniwersytetu Nauk Stosowanych w Biel i dr. Andreasem Mayerem – TTM ze Szwajcarii, oraz gospodarzem pierwszego międzynarodowego spotkania specjalistów zajmujących się ograniczaniem emisji cząstek stałych w gazach spalinowych pojazdów samochodowych – 1st International Workshop on Particulate Matter Emissions from Engine and Automobile Sources, które odbyło się 2 lipca 2012 r. w Bielsku-Bialej. Spotkanie to było również jedną z kilku uroczystości związanych z czterdziestoleciem Instytutu BOSMAL, który jest sukcesorem OBR SM BOSMAL w Bielsku-Białej. W pierwszym spotkaniu specjalistów PM uczestniczyli przedstawiciele 14 firm z przemysłu motoryzacyjnego i paliwowego oraz instytutów badawczych i uczelni technicznych z 7 krajów. W czasie tego spotkania był zaprezentowany referat programowy wygłoszony przez znanego światowego eksperta ds. emisji nanocząstek prof. Davida B. Kittelsona z Uniwersytetu Minnesota – USA oraz pięć innych referatów zaprezentowanych przez znanych specjalistów w tym zakresie: Manfereda Linke z AVL – Austria, dr Amandę Lea-Langton z Leeds University – Anglia, dr. Paula Zelenkę z VERT Association – Szwajcaria i prof. Jana Czerwińskiego z AFBH – Szwajcaria, a z ramienia BOSMAL referat dotyczący doświadczeń tej firmy w badaniach emisji cząstek stałych zaprezentowali dr. Piotr Bielaczyc i Joseph Woodburn.

Bardzo istotna podczas tego spotkania była dyskusja panelowa, podczas której zaproszeni eksperci oraz przedstawiciele uczestniczących firm odpowiadali na pytania dotyczące podstawowych zagadnień związanych z ograniczaniem emisji cząstek stałych, przygotowane przez zespół prof. Jerzego Merkisza z Politechniki Poznańskiej, zagadnień związanych z porównaniem emisji liczby i masy cząstek stałych i metod ich pomiaru, z emisją cząstek stałych z nowoczesnych silników z bezpośrednim wtryskiem paliwa do komory spalania, możliwością pomiaru liczby cząstek o określonych średnicach, porównaniem laboratoryjnych metod pomiaru emisji cząstek z metodą pomiaru ich rzeczywistej emisji w czasie ruch pojazdu na drodze, a także prowadzili dyskusję na temat dalszych kierunków rozwoju metod pomiarowych i ograniczania emisji PM z różnych typów silników i pojazdów.

Ponieważ silniki spalinowe będą przez jeszcze wiele lat podstawowym źródłem napędu różnych pojazdów i maszyn roboczych, więc ograniczanie emisji związków szkodliwych i toksycznych, do których zaliczana jest również emisja cząstek stałych, a szczególnie nanocząstek, pozostaje jednym z najważniejszych problemów do rozwiązania dla konstruktorów tych silników i pojazdów, nie tylko wyposażonych w silniki z zapłonem samoczynnym, ale także z zapłonem iskrowym, z układami bezpośredniego wtrysku benzyny do komory spalania silnika (GDI). Emisja cząstek stałych jest ograniczana przepisami prawnymi dotyczącymi maksymalnej masy emitowanych cząstek zebranych na filtrach pomiarowych podczas specjalnych cykli badawczych i dla niektórych typów silników również liczby emitowanych nanocząstek (PN). Wkrótce będzie ograniczona dla wszystkich typów silników spalinowych w ich różnych zastosowaniach. Korelacja masy (PM) i liczby (PN) cząstek stałych jest bardzo trudna. W przepisach Unii Europejskiej obie te wartości są obecnie limitowane dla silników samochodowych. Dla nowoczesnych pojazdów z silnikami ZS, wyposażonych w filtry cząstek stałych (DPF), które w dużym stopniu ograniczają emisje cząstek, właśnie pomiar ich liczby staje się podstawowym pomiarem do określenia poziomu emisji cząstek stałych silnika w odniesieniu do obowiązujących limitów. Limit maksymalnej dopuszczalnej emisji PN będzie także wyprowadzony w przepisach Euro 6-1 dla pojazdów z silnikami GDI. Trwa obecnie dyskusja nad wprowadzeniem limitowania liczby cząstek PN również w przepisach USA i Japonii.

Wszystkie wygłoszone referaty oraz pięć innych prezentacji przygotowanych tylko w formie pisemnej zostały opublikowane w materiałach konferencyjnych pod tytułem: *Particulate Matter Emissions from Engine and Automobile Sources*, ISBN 978-83-931383-3-3, wydanych na płycie CD, dołączonej do tego numeru Combustion Engines/*Silników Spalinowych*.

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