Feature Article

Feature Article

Real Time Solid Particle Counting System MEXA-1000SPCS

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The MEXA-1000SPCS measures number concentration of solid particles in engine exhaust gas within a specified particle size range in real-time. The system conforms to the UN/ECE particle concentration measurement standard (Regulation No.83, Rev.3, Amend.2), and consists of a pre-classifier, a volatile particle remover [primary diluter (PND1), a secondary diluter (PND2) and an evaporation tube] and a detector [condensation particle counter (CPC)]. The MEXA-1000SPCS uses two wide range continuous diluters that ensure conditioning of exhaust gas to produce stable solid soot particles by removing the volatile fraction from exhaust. The system takes sample from dilution tunnel however, it is possible to collect sample from engine exhaust manifold with an additional pre-dilution unit.

Introduction

Currently, particulate matter (PM) emitted from automobile engines are regulated gravimetrically as the "particulate mass emission". However, the need for regulations on emitted "particle number" is gaining concern in Europe recently. In the engine emitted particles, a large majority are ultrafine particles of 100 nm or smaller in size. Both solid and volatile particles are included in the exhaust. However the solid particles are stable and such particles are assumed to be infiltrated into the human body through the bronchial tubes and the alveoli, transferred to different organs, exist long time, and their health effects are a cause for concern. Therefore, instead of regulating particulate mass only, regulations on the particle number are gaining attention.

Accordingly, in 2001, Particle Measurement Programme (PMP) was organized under the Working Party on Pollution and Energy (GRPE) of the United Nations Economic Commission for Europe (UN/ECE). A number of countries including Japan are participating in this program. The PMP proposed a particle number counting methods and organized validity tests for the method. Also, EU has already announced limits for particle number emission, scheduled for introduction in 2011.
The MEXA-1000SPCS was developed based on the recommended specifications for particle number counting systems published as the UN/ECE regulation[1]. This report describes the configuration and basic performance of the MEXA-1000SPCS[2], and will provide examples of measuring particle number concentration emitted from vehicles equipped with various after-treatment devices[2, 3].

Outline of the MEXA-1000SPCS

Requirements in UN/ECE Regulation

The volatile compounds in engine exhaust such as soluble organic fraction (SOF) and sulfate are said to be greatly dependent on the dilution conditions. Therefore, the UN/ECE regulation requires that volatile particles shall be removed from the sampled exhaust gas. Only the solid particles should move into the condensation particle counter (CPC), which measures particle number concentration.
Figure 1 shows the schematic of the particle measurement system required in UN/ECE regulation. The pre-classifier provides 50% particle cutoff within 2.5 to 10.0 µm size range. The hot diluter (PND1) dilutes exhaust gas with particle free dilution air. The dilution air is heated above 150 °C. Dilution with high temperature air suppresses the formation of volatile particles in the PND1. The evaporation tube (ET) with

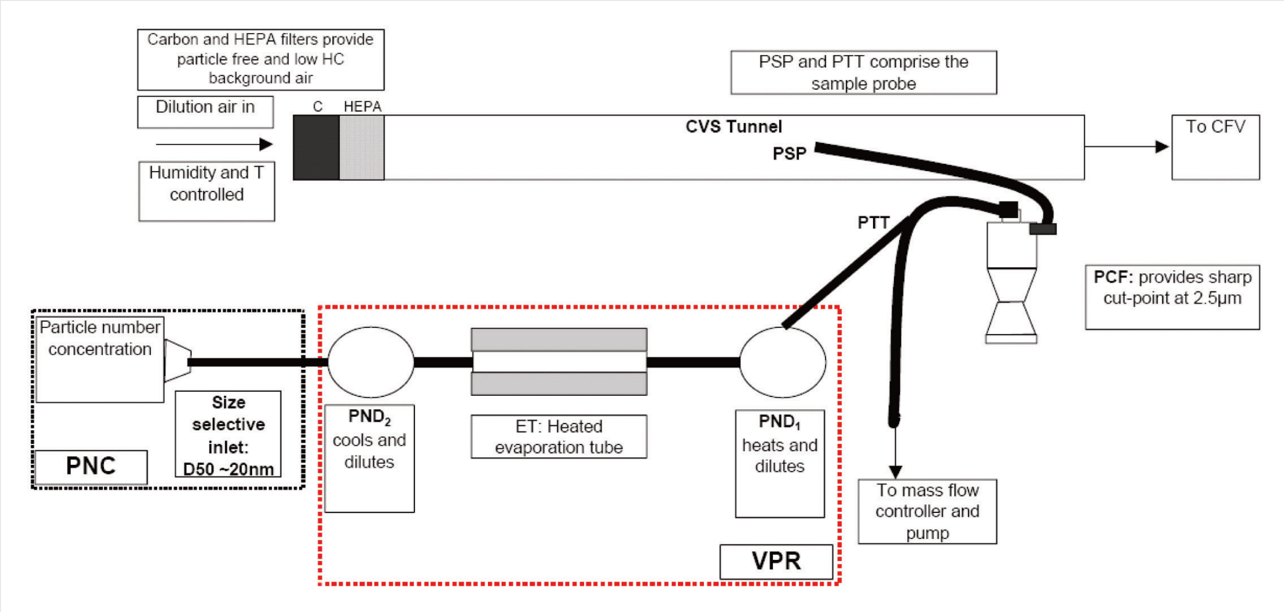

Figure 1 Particle Number Counting System Requirement in UN/ECE

the average temperature between 300 to 400 °C is applied to remove the volatile particles consist of SOF and sulfur compounds by evaporating them to gas phase. By following cold dilution with room temperature air in the cold diluter (PND2), concentrations of volatile compounds at gas phase are decreased to minimum level. As a result, the possibility of re-nucleation and to form new volatile particle is minimized. Finally, the concentration of the solid particle is measured in the condensation particle counter (CPC), where the cutoff size of the CPC is 23 nm by recommendation.

Configuration of MEXA-1000SPCS

Figure 2 shows pictures for the MEXA-1000SPCS. It has been developed based on the UN/ECE regulation. The MEXA-1000SPCS integrates the pre-classifier, PND1, ET, PND2, and CPC in a single cabinet. All the basic performance checks which are recommended in UN/ECE can be carried out from application software. The

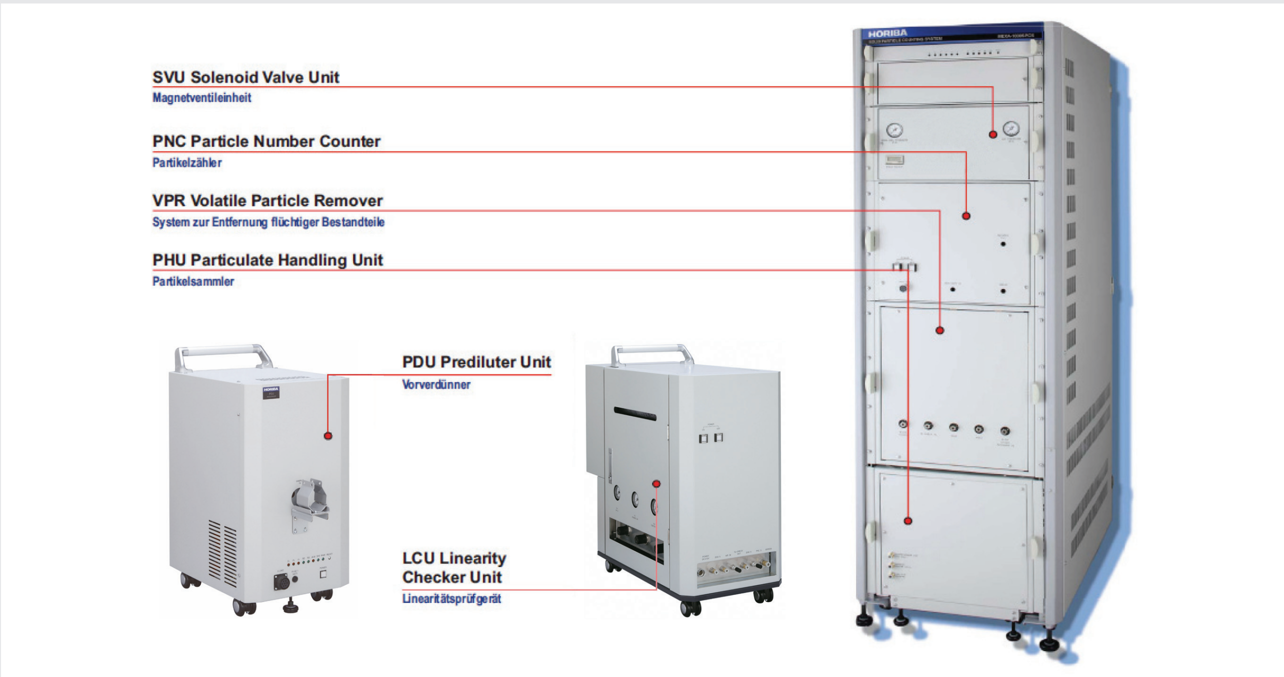

Figure 2 The Solid Particle Number Counting System MEXA-1000SPCS

specifications of MEXA-1000SPCS are summarized in Table 1.

Table 1 Specification of MEXA-1000SPCS

Model	MEXA-1000SPCS
Components and Range	Number concentration of solid particles; 0-10000 up to 50000 particles/cm^3 (after dilution)
Configuration	Main cabinet: Volatile particle remover (VPR), Particle number counter (PNC), Solenoid valve unit, Filter and pump unit Control: PC, LCD, Keyboard, Mouse
Volatile particle remover (VPR)	Pre-classifier (Cyclone): 50% cut point of particles: 2.5 μm to 10 μm Diluter (WRCD): PND1: DF 10 to 700 PND2: DF 10 to 50 Sample flow rate: 10 to 15 L/min
Particle number counter (PNC)	Detector: Condensation particle counting (CPC)

Wide Range Continuous Diluter

Under the UN/ECE regulation, it is recommended that the PND1 should provide a wide dilution range of up to 1:200. The wide range continuous diluter (WRCD) used in this system has been newly developed for the PND1 and PND2 to achieve the required specification. The schematic of the diluter is presented in Figure 3. The WRCD consists of mass flow controllers (MFC), piezo-valve (PV), pressure transducers, thermocouples, orifice flow meters, and a critical flow orifice. There are no moving parts in contact with the aerosol and therefore the loss of particles is minimized. The flow in the WRCD can be defined by equation 1. The dilution factor (DF) on the WRCD can be defined by equation 2:

$$Q_s = Q_{\text{total}} + Q_{\text{instrument}} - Q_{\text{air}} - Q_{\text{make-up}} \quad \cdots\cdots (1)$$

$$DF = \frac{Q_{air} + Q_s}{Q_s} = 1 + \frac{Q_{air}}{Q_s} \quad \cdots\cdots (2)$$

Where, Q_{air} is the particle free dilution air, and is controlled accurately by a mass flow controller. Q_s is the sample flow, and measured by the orifice flow meter in real-time. By adjusting the make-up air flow ($Q_{make\text{-}up}$), the sample flow (Q_s) can be kept constant under all conditions. This allows for a constant dilution factor to be maintained, even if the total flow (Q_{total}) and instrument flow ($Q_{instrument}$) fluctuate slightly.

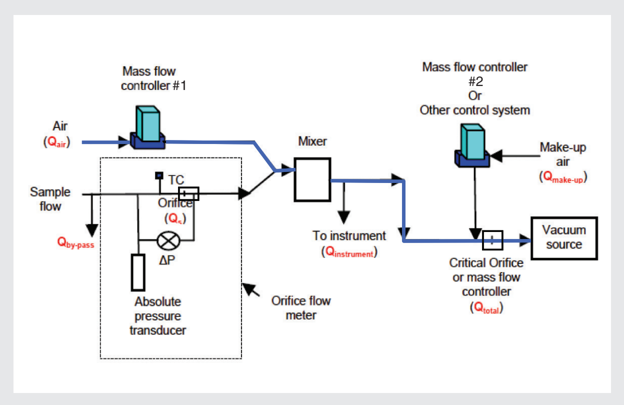

Figure 3 The Schematic of the Wide Range Continuous Diluter

Basic Performances

Particle Penetration

Inertial impaction, particle settling in laminar and turbulence, diffusion, thermophoresis, etc., may induce particle losses in the particle measurement system. Therefore higher particle penetration through diluters is one of the main criteria of the system. Figure 4 shows the schematic for the penetration test with the monodisperse aerosol. The Sodium Chloride (NaCl) aerosol generated by the atomizer flows into the diffusion dryer to remove water vapor or droplets, and then is used as solid particle sample. Figure 5 presents the particle size distributions in log scale measured at upstream and downstream of the VPR using a scanning mobility particle sizer (SMPS). In the graph, "Raw" represents the particle size distribution at the upstream of instrument inlet, and "DR500", "DR750", "DR1000" represent particle size distributions at the CPC inlet under the dilution factors of 500, 750 and 1000.

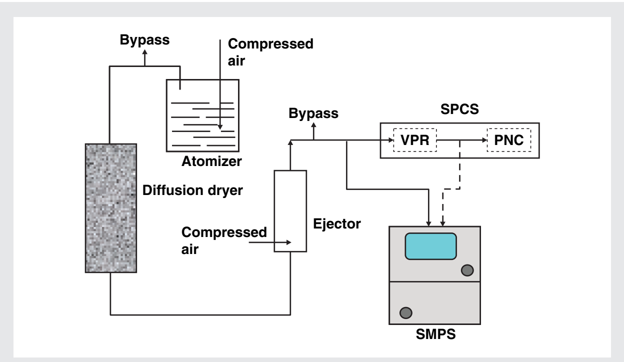

Figure 4 Schematic of Mono-Disperse Particle Generator for Penetration Test

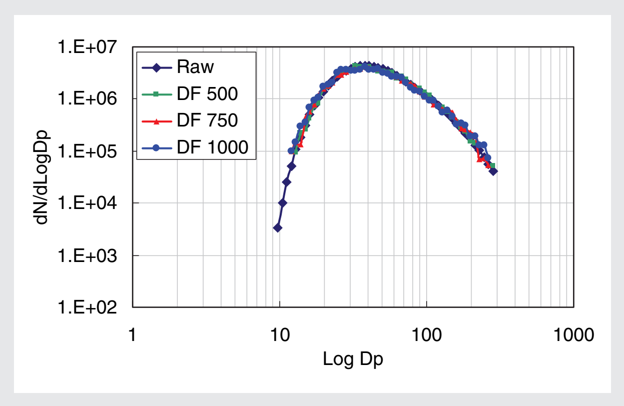

Figure 5 Particle Size Distribution for Different Dilution Factors

Diluted size distributions are corrected back to raw concentrations by multiplying the SPCS overall dilution factors. Furthermore, to evaluate penetration rates including particle losses due to thermophoresis, the temperatures of each unit were adjusted to normal measurement conditions. The particle penetration rates for all particle sizes are represented in Figure 6. The particle penetration rate decreases as the dilution factor (DF Total) increases due to increased losses by diffusion. In spite of that, more than 95% particles penetration is obtained for a total dilution factor of 1000.

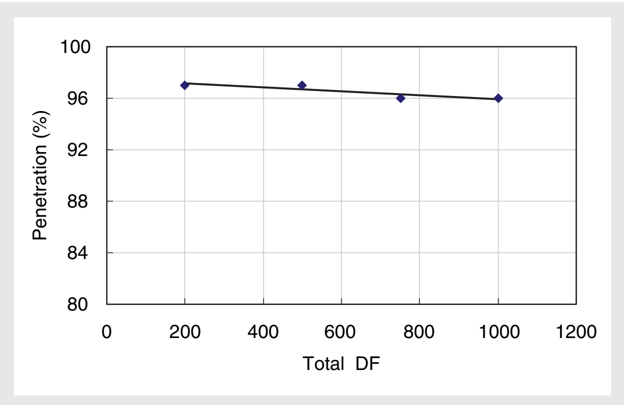

Figure 6 Particle Penetration for All Particles at Different Dilution Factors

Linearity of the CPC

The MEXA-1000SPCS has the function of daily linearity check of the CPC using particle generator. Figure 7 shows an example of the linearity check of the CPC. The X-axis is the reference concentration which is obtained by dividing the raw concentration from particle generator by dilution factor of the diluter. The Y-axis is the average concentration measured by the CPC. Figure 7 shows that the CPC has good linearity with a correlation co-efficient and a co-efficient of determination of approximately equal to unity.

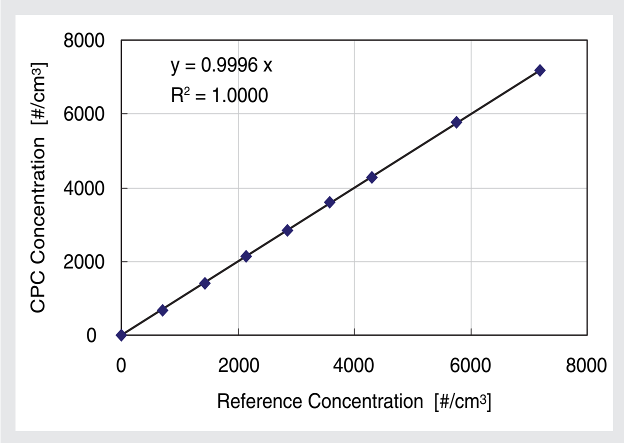

Figure 7 Daily Linearity Check for the CPC

Removal Efficiency on the Evaporation Unit

An evaporation unit has been integrated in the MEXA-1000SPCS to remove soluble organic fraction (SOF) and sulfur compounds from the aerosol. As a result, the aerosol with solid particles only moves into the CPC. The UN/ECE regulation recommends that removal efficiency on the evaporation unit should be higher than 99.0% for 30 nm tetracontane (C40) particles.

Figure 8 shows the schematic of the removal efficiency test on the evaporation unit. A C40 aerosol generator generates C40 particles by heating the pure tetracontane to the sublimation point. Then C40 particles move into a DMA where single size particles are classified.

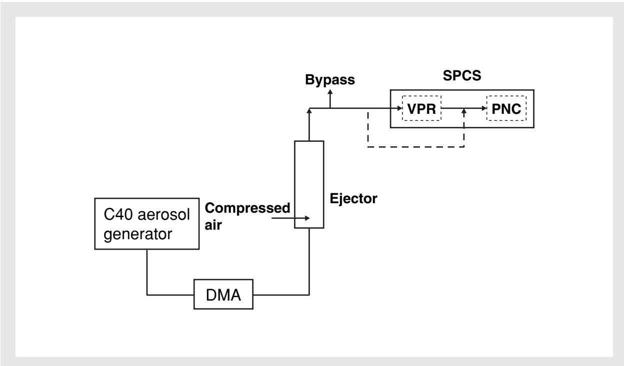

Figure 8 Schematic of Mono-Disperse C40 Aerosol Generator

Figure 9 shows minimum temperatures of the evaporation unit to remove 30, 50, and 100 nm C40 particles from sample gas. Constant concentration monodisperse aerosols of about 10^4 particles/cm^3 were used in the test. The dilution factor in the PND2 was maintained at 24.5. It is observed that the minimum temperature for C40 removal is higher for larger particles. The evaporation unit temperature is controlled at 320 °C, and is high enough to achieve the removal efficiency of 99.9 % or more for C40 particles of 30, 50, and 100 nm size.

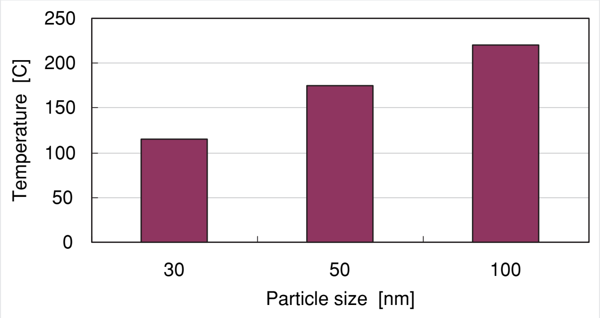

Figure 9 Minimum Temperature for Complete Removal of C40 Particles

Accuracy of Dilution Factor

Figure 10 presents the percentage difference between the SPCS reference dilution factor and the gas based measured dilution factors. Propane (C_3H_8) was used as the span gas in the measurement. The dilution factor was estimated based on the measurement of diluted gas concentration by the HC analyzer (FID). In this case, background corrections are necessary as the concentration of diluted propane approaches to atmospheric levels during high dilution. Based on the measurement result from the HC analyzer, the dilution factor is defined in equation[3].

$$DF = \frac{C_{in} - C_{air}}{C_{out} - C_{air}} \qquad (3)$$

Where, DF is the dilution factor; C_{in} is the concentration of the span gas in ppm; C_{out} is the concentration measured with the HC analyzer in ppm; and C_{air} is the HC background concentration in dilution air. In general, the measured dilution factors are within ±2% of the reference dilution factor in the range of 200 to 1000. The regulation requires that the difference between the gas based measured dilution factor and the reference target dilution factor should be within ±10%.

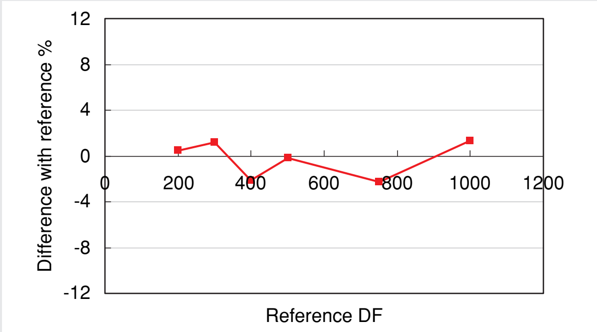

Figure 10 Percentage Difference between the Reference Dilution Factor and Measured Dilution Factors.

Comparisons with PMP's Systems

The PMP has arranged an Inter-Laboratory Correlation Exercise (ILCE) in order to test the validity of the method recommended. A DPF-equipped modern diesel vehicle called Golden Vehicle (GV) was used as the reference vehicle.

A reference particle counting instruments called Golden Particle Measurement System (GPMS) was used to compare with the other systems close to the recommended specifications. The exercise was conducted in different laboratories world wide. In the National Traffic Safety and Environment Laboratory (NTSEL) Japan, tests were conducted with the MEXA-1000SPCS along with the GPMS[2]. Table 2 outlines the specifications of the vehicles used in the experiment.

Table 2 Specification of Test Vehicles

Vehicle	Swept Volume	Engine type	After-treatment system
GV	2.0 L	Common-rail direct injection diesel (w/ turbocharger)	SiC DPF
JV-1	3.0 L	Direct injection gasoline	Three-way catalyst + de-NOx catalyst
JV-2	2.0 L	Common-rail direct injection diesel (w/ turbocharger)	DPF + Oxidation catalyst

Figure 11 shows the particle concentration measurement results of the MEXA-1000SPCS. The upper side graph shows the instantaneous emission concentration of solid particles during NEDC testing, and the bottom side graph is a close up of the upper graph from cold start to first 400 seconds. In the graph seven measurement results show excellent repeatability. Since particle emission increases immediately after DPF regeneration, tests were conducted after two or more dummy runs after the DPF was regenerated.

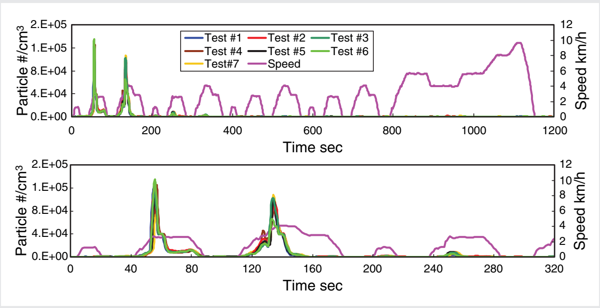

Figure 11 Solid Particle Emission from Golden Vehicle under NEDC Test Cycle

Figure 12 shows the comparison of particle number emission measured by the GPMS and MEXA-1000SPCS. Both systems showed nearly identical measurement results. The PM mass emission measured by conventional gravimetric method was approximately 0.37 mg/km for the GV.

Figure 13 shows the repeatability of the tests in terms of coefficient of variance (COV). The repeatability of MEXA-1000SPCS is better than that of the GPMS. It is thought that the poor repeatability of JV-2 measurement than other two vehicles is due to high DPF regeneration frequency in JV-2.

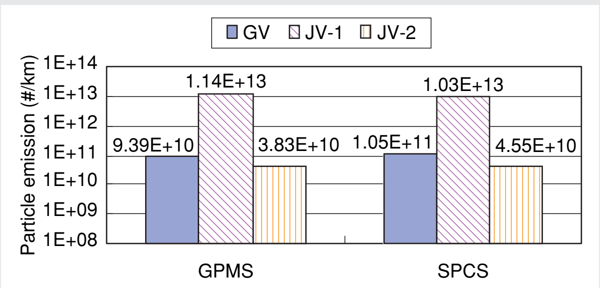

Figure 12 Differences in the Number of Solid Particles Emitted for Each Vehicle

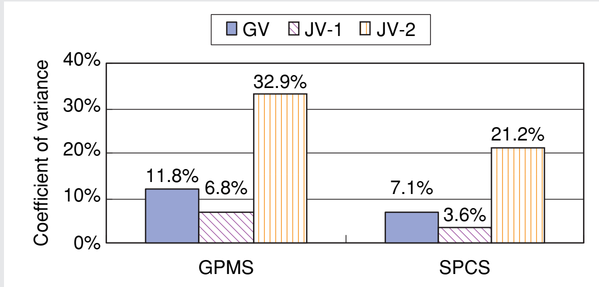

Figure 13 Repeatability of Particle Number Counting Systems (GPMS, SPCS)

Particle Number Emission from Various Vehicles

MEXA-1000SPCS has been used to investigate the soot emission behavior from different vehicles with different after-treatment technologies[3]. Table 3 shows the specification of the test vehicles. Three DI diesel vehicles with catalyzed DPF named as DDPF-1, DDPF-2, and DDPF-3 were tested. One DI diesel vehicle without DPF was also considered to observe the emission behavior. In addition, one DI gasoline vehicle was also tested. Tests were performed in various chassis dynamometer test cells. All the vehicles were preconditioned well before each test in order to have comparable data. For test under cold start condition the vehicles were soaked at least five hours after preconditioning. Tests are limited to sampling from tunnel rather than direct sampling.

Particle Emission from Vehicles with and without DPF

First of all the comparison of emission behavior from vehicles with and without DPF has been attempted. Two DI diesel vehicles i.e. non-DPF and DDPF-1 (as shown in Table 3) have been considered in this purpose. The

Table 3 Specifications of Test Vehicles

Parameter	Vehicles				
	Non-DPF	DDPF-1	DDPF-2	DDPF-3	DIG
Engine	DI Diesel	DI Diesel	DI Diesel	DI Diesel	DI Gasoline
Injection System	Common Rail	Common Rail	Common Rail	Common Rail	DI-EFI
Swept Volume	2.2 L	2.0 L	2.2 L	2.0 L	3.0 L
Max Power kW/rpm	105/4000	Unknown	105/4000	86/3500	188/6200
Max Torque N-m/rpm	340/2000	Unknown	340/2000	178/2000	314/3600
Transmission	6-Manual	6-Manual	6-Manual	5-Manual	Automatic
Vehicle Weight	1637	1590	1637	1430	1855
Fuel Economy km/L	14.5	Unknown	14.5	15.4	11.8
After Treatment	Unknown	DPF (SiC)+ FBC	DPF + DOC	DPF + DOC	TWC + NRC
Intake	TCIC	TCIC	TCIC	TCIC	NA
Emission Level	Unknown	EURO-IV	EURO-IV	JP (2000)	JP (2000)

NEDC driving cycle under hot start condition was used for each vehicle in the same test facility under the same dilution condition. Figure 14 represents the results obtained from test done in this case. Bottom side graph of Figure 14 shows the "particle emission rate" for non-DPF and DDPF-2 vehicles. The particle emission rate is defined as the ratio of the real-time cumulative particle number emission to the total particle number emission during the driving cycle.

It shows that the particle emission rate for DDPF-2 vehicle increases almost linearly through out the driving cycle under hot start condition. Under this condition a slight portion of fine particles those are difficult to be trapped on the filter surface are assumed to pass through the DPF due to intermittent pressure fluctuation in the exhaust pipe[4]. On the other hand the particle emission rate from the non-DPF vehicle remains low in the ECE part of the driving cycle while it increases very sharply in the EUDC part where the vehicle is accelerated sharply. For non-DPF vehicle the particle emission rate is directly correlated with the engine operating condition. At high load and high speed condition, many particles are emitted and the particle emission rate increases.

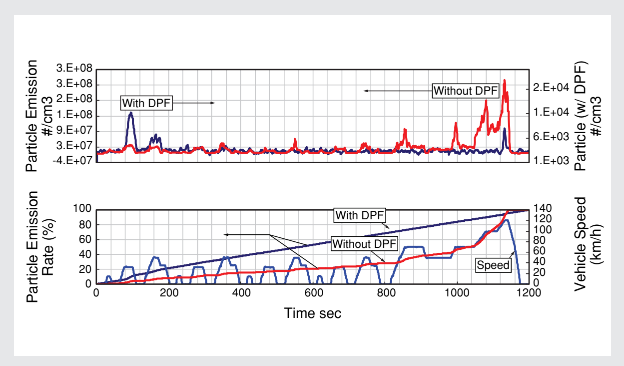

Figure 14 Particle Emission from DI Diesel Engine with and without DPF

Particle Emission from Vehicles Fitted with Catalyzed DPF

Emission behavior of solid particles from three vehicles; DDPF-1, DDPF-2 and DDPF-3, has been investigated. NEDC driving cycle under cold start condition was used for each vehicle but in different facilities, and the results are represented in Figure 15. Only first half (600 seconds) of the driving cycle is plotted in this figure rather than whole cycle because of no significant difference in the later part. Three graphs at the upper part of figure 15 represent the real-time solid particle emission from three vehicles. It shows that the DDPF-1 vehicle emits higher number of solid particles during the first 200 seconds of the driving cycle as compared with the other two vehicles. On the other hand, the DDPF-2 and DDPF-3 vehicles emit lower number of particle during the first 200 seconds compared with the DDPF-1 vehicle, but they emit certain degree of emission continuously thereafter. As a result the numbers of emitted particles were nearly identical for the overall test cycles. The accumulative particle emissions calculated from the real-time emissions are shown in the bottom graph of Figure 15. The DDPF-1 vehicle emitted roughly 80% of its total number in the first 200 seconds. This shows a great difference from the DDPF-2 and DDPF-3 vehicles, which emitted around 30 to 50% of the respective total number in the first 200 seconds.

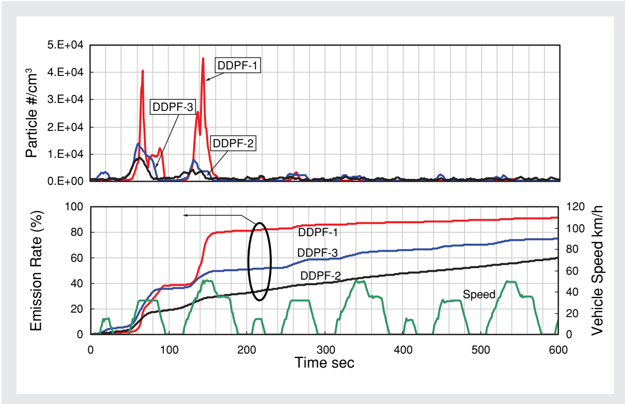

Figure 15 Emission Behavior of Solid Particles from Three Vehicles Fitted with DPF

Regeneration and Particle Emission

Particle emission behavior during regeneration and after the regeneration has been investigated by using the DDPF-2 vehicle. The vehicle was driven under NEDC driving cycle in hot condition. One test was done before regeneration and then regeneration was initiated externally. Just after regeneration four successive tests were also performed under hot start condition. The results are shown in Figure 16. Graph at the top of figure shows the real-time particle emission behavior during regeneration of the DPF.

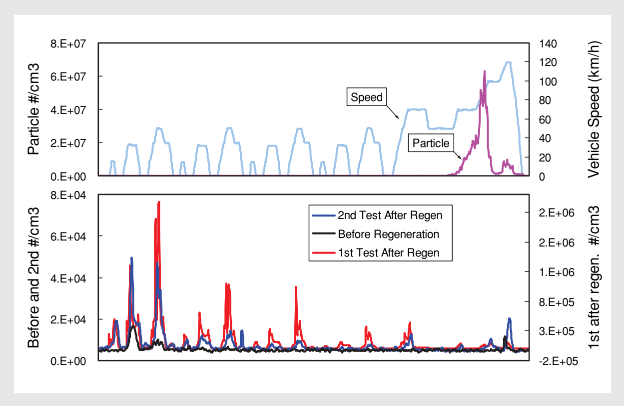

Figure 16 Particle Emission Behavior during Regeneration and after the Regeneration

It shows that huge number of particles is emitted during regeneration in the EUDC part of the driving cycle. Graph at the middle of the figure shows the real-time particle emission behavior for first test just after regeneration. In the first test immediately after the regeneration huge number of particle emission was observed.

The particle concentration is about 2 orders higher than normal DPF condition. Graph at the bottom of the figure shows the particle emission for two different tests, i.e. second test after regeneration and before regeneration. The particle concentration decreases significantly in the second test after regeneration, but it did not return to the level before regeneration. Average particle concentration measured by the SPCS and estimated filtration efficiency are summarized in Figure 17. The filtration efficiency was estimated by assuming the non-DPF emission as the maximum. It shows that from the fourth test after regeneration excellent repeatability in number counting can be achieved. Filtration efficiency reaches to maximum after fourth test and the DPF itself assumed to be stable. Upon regeneration, filter becomes clean and filtration performances decreases. It is reported that PM is first collected in the pores of the filter wall[5]. After a transition phase, the PM builds up as a discrete layer, or soot cake, on the inlet channel walls, therefore, filtration efficiency increases.

During regeneration particle number emission is only about 20% of the non-DPF particle number emission. However this emission is about 200 times of the particle number emission in case of a stable DPF operation with sufficient loaded soot.

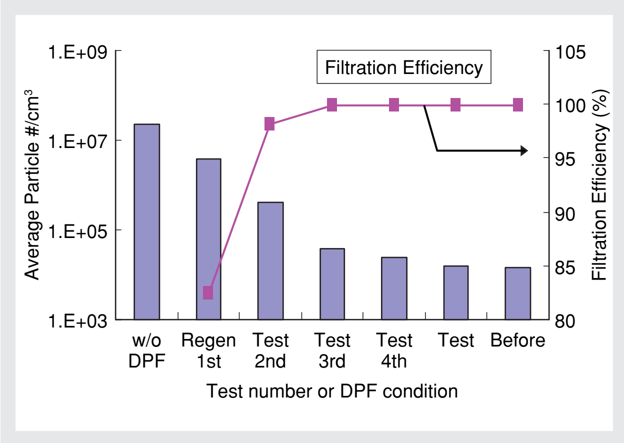

Figure 17 Filtration Efficiency of DPF at Different Situation

Particle Emission from a DI Gasoline Vehicle

Comparison of a DPF fitted diesel vehicle with a DI gasoline vehicle has been attempted in this case. The DDPF-1 and DIG vehicles were used in this purpose. Both vehicles were driven under NEDC driving cycle in cold start condition in the same test facility. Results are summarized in Figure 18.

Two graphs at the upper part of the figure show the real-time particle emission from DDPF-1 and DIG vehicles. Concentration of particles emitting from DDPF-1 vehicle is very high in the early part of the driving cycle while the DIG vehicle emits consistently higher concentration of particles through out the driving cycle. The DDPF-1 vehicle emits significantly lower total number of particles in comparison with the DIG vehicle. Particle emission rate calculated from this real-time emission is presented in the bottom side graph of Figure 18.

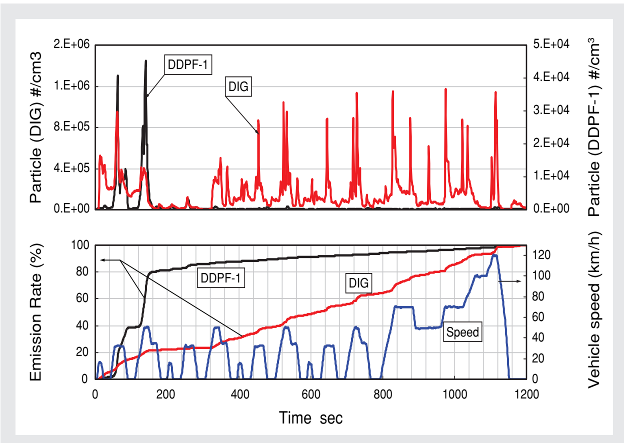

Figure 18 Comparison of the Real-time Particle Emission from DDPF-1 and DIG Vehicles

It shows that DDPF-1 vehicle exhibits higher particle emission rate during the first 200 seconds of the driving cycle. However the DIG vehicle shows relatively constant emission rate. Therefore DDPF-1 vehicle should be considered for first 200 seconds in order to improve its particle emission quality. On the other hand, DIG vehicle should be considered for overall driving cycle for further improvement.

Particle Emission from Different Vehicles

Figure 19 shows the overall comparison of particle emission in number per kilometer from all the test vehicles used in this study. DDPF-1, DDPF-2 and DDPF-3 vehicles show almost similar particle number emission and the value is lower than 10^{11} particles/km. The DIG vehicles emit higher number of particles. The Non-DPF vehicle emits the highest number of particles as expected.

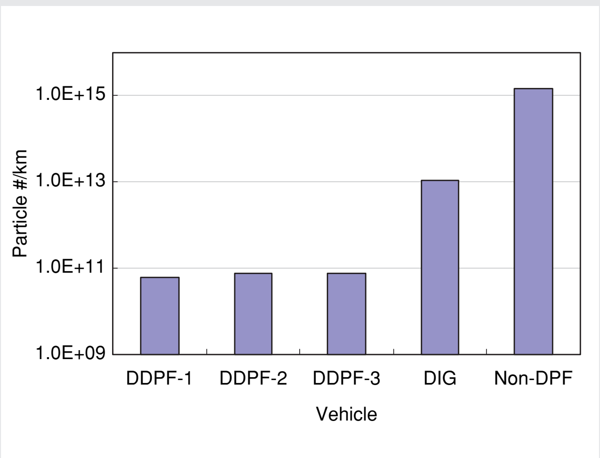

Figure 19 Overall Comparison of Particle Emission in Number / Kilometer from Different Test Vehicles

Conclusion

The MEXA-1000SPCS is developed under a completely different concept from the conventional gravimetric PM measurements methods. All the basic components such as pre-classifier, VPR (PND1, ET and PND2) and CPC comply with the specifications in the UN/ECE regulation. The overall performance also satisfies the criteria. Therefore, it is expected that the system will become a highly effective tool to respond to the forthcoming particle regulations.

The MEXA-1000SPCS can primarily be used for sampling from CVS tunnel. However direct sampling from tail pipe is also possible when coupled with a dedicated pre-diluter. It is assumed that the MEXA-1000SPCS can contribute to low concentration particle measurement for research and development of engine and after-treatment system.

Abbreviations

CPC : Condensation Particle Counter
DF : Dilution Factor
DMA : Differential Mobility Analyzer
DPF : Diesel Particulate Filter
ET : Evaporation Tube
GRPE : Groupe des Rapporteurs pour Pollution et Energie
ILCE : Inter-Laboratory Correlation Exercise
NEDC : New European Driving Cycle
PMP : Particle Measurement Program
PNC : Particle Number Counter
PND : Particle Number Diluter
SOF : Soluble Organic Fraction
SMPS : Scanning Mobility Particle Sizer
VPR : Volatile Particle Remover
WRCD : Wide Range Continuous Diluter

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