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Research on Technology for Reduction of Fine Particles and Hazardous Air Pollutants from Engine Exhaust Gas Emissions

(Engine exhaust gas reduction technology group)
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1. Contents of Empirical Research

The purpose of the present research is to investigate and establish methods of measuring hazardous air pollutants (HAPs) and fine particles (FP) distributions, to determine the effects of engine technology and of fuel properties and compositions, and to develop effective reduction technology by combining engine technology and fuel improvements based on these findings.

The following 4 specific items were carried out.

(1) Investigation and establishment of methods for measuring FP in emissions and HAPs

(2) Investigation of the mechanisms of FP and HAPs generation in engines

(3) Investigation of the effects of fuel and engine technology on emissions of FP and HAPs

(4) Investigation and development of FP and HAPs reduction technology by combining fuel and engine technologies

Items (1) to (3) were carried out in 1999.

2. Results of Empirical Research and Analysis Thereof

2.1 Investigation of FP measurement methods

In order to discover the optimum method of measuring FP, various influential factors at the time of measurement were studied. Investigated as influential factors were sampling tube material and length, conditions of diluted air (temperature, humidity), dilution magnification and re-dilution method. There are various methods of measuring particle size. Scanning Mobility Particle Sizer (SMPS) is a method; particles were grown after classification and measured optically from the degree of electrical mobility, which is considered applicable to automobile exhaust particles. Electrical Low Pressure Impactor (ELPI) is other method to measure particle size distribution by measurement of post-classification charge with an impactor.

(1) Effect of sampling tube material and length

The following 6 types of sampling tube material were selected and tubes 3 m and 1.2 m in length were provided: metal, electro-conductive silicone, vinyl chloride, silicone, Teflon, polyethylene. Shown in Figure 2.1-1 is the total particle count of 0.5 μm or below per unit volume of diluted exhaust gas when the sampling tube length is 3 m. Figure 2.1-2 gives the total particle count at the length of 1.2 m as a percentage of the total count at 3 m. In SMPS, the loss in particle count was slight at both lengths with stainless steel, electro-conductive silicone and vinyl chloride, and with silicone, Teflon and polyethylene, a particulate loss of about 50% was observed. In ELPI, there were no large differences by material at 1.2 m but at 3 m, particle loss of stainless steel, electro-conductive silicone and vinyl chloride was slight. According to the effect of length, it was supposed that the results were caused from particle properties because the drop in particle concentration is larger at 25% load than at 95% load.

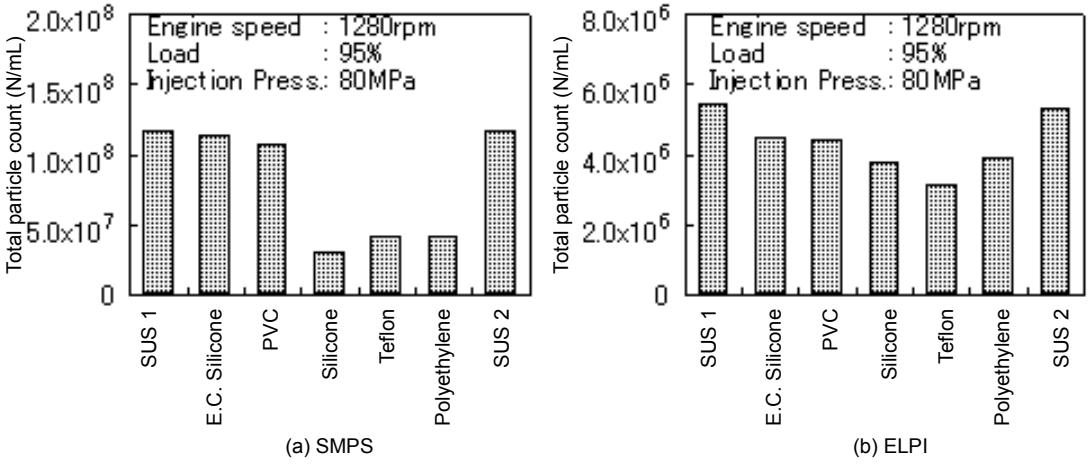

Figure 2.1-1 Effect of differences in sampling tube material on particle size distribution

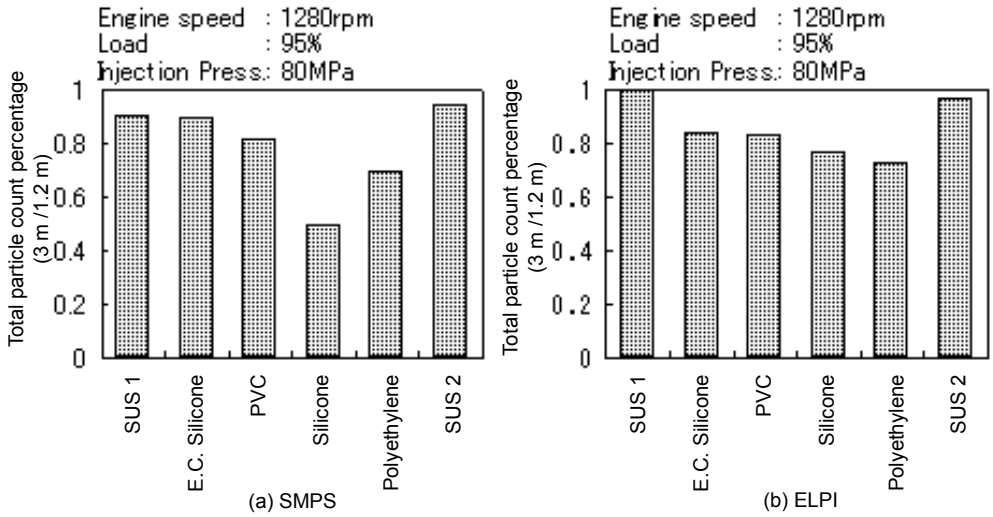

Figure 2.1-2 Effect of sampling tube length with each sampling tube material

(2) Effect of diluted air condition (temperature, humidity)

Figure 2.1-3 shows patterns of particle size distribution as determined by SMPS during low-load operation when diluted air temperature and humidity have been changed.

When temperature and humidity have been changed, the effect is slight at 95% load in SMPS under both conditions. At 25% load, however, when the temperature is high or the humidity is high, an increase in particle number of 30 nm or less can be noted, and this is ascribed to the effect of differences in particulate properties. In ELPI, no clear effect from temperature could be recognized.

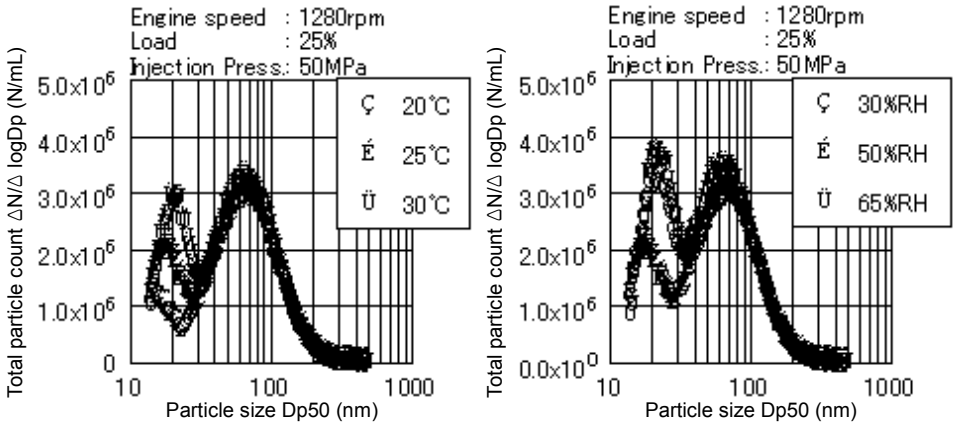

Figure 2.1-3 Effect of diluted air temperature and humidity on particle size distribution (SMPS)

(4) Effect of dilution ratio

The effect of dilution ratio was investigated for cases in which the entire volume of exhaust gas was directed into a dilution tunnel and CVS flow volume was changed and for cases in which CVS flow volume was held constant and the volume of directed exhaust gas was changed. Figure 2.1-4 gives the effect of dilution ratio on the total particle count of diluted exhaust gas at 0.5 μm or less per unit volume by dilution magnification. At high dilution ratio, such things as accuracy in the setting of conditions must also be considered, but in SMPS and in ELPI, no major differences in the distribution patterns of the two conditions could be noted. In SMPS, a downward trend in the number of particles from the total particle count was noted as the dilution ratio becomes higher in the case of 25% load, but in ELPI, the effect of dilution ratio could not be observed.

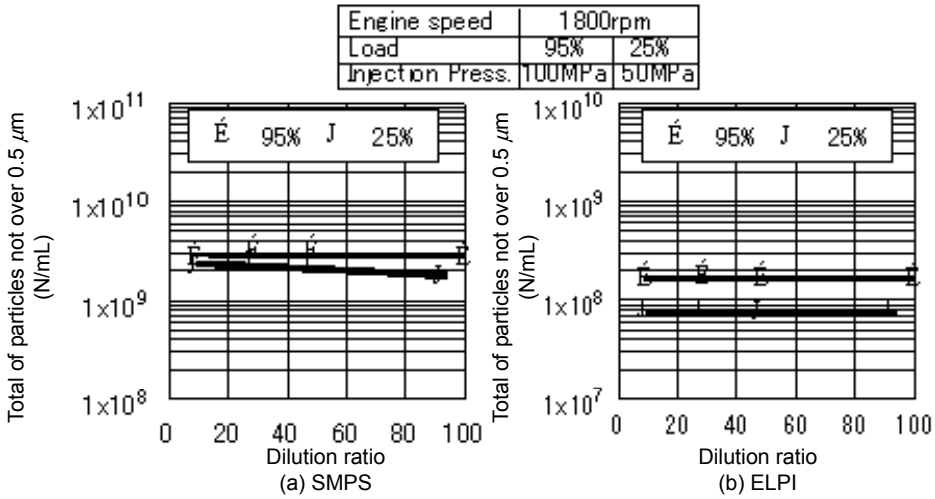

Figure 2.1-4 Effect of dilution ratio on total particle count

(5) Effect of re-dilution method

Taking the condition in which the entire volume of exhaust gas has been directed into a dilution tunnel as dilution 1, the diluted exhaust gas was re-diluted using an ejector type diluter. The dilution ratio of this diluter was fixed at 10, and by using two of the diluter units in series, a dilution ratio of 100 could be easily obtained. Dilution percentage and total particle count percentage are shown in Figure 2.1-5. From total particulate count, a linear relationship of slope 1 can be obtained for roughly both variables, and measurement with re-dilution method is considered possible.

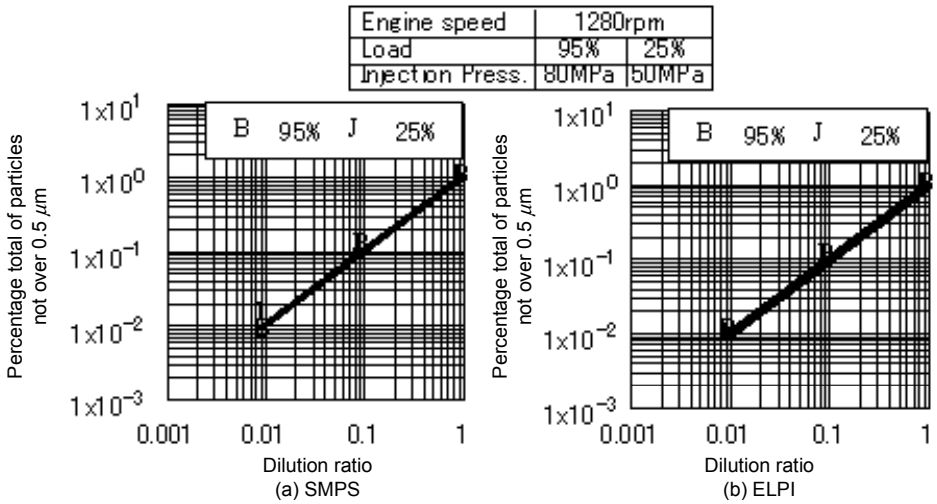

Figure 2.1-5 Dilution ratio versus total particle count percentage

(6) Effect of ELPI temperature

Figure 2.1-6 presents the results of a comparison of particle size distribution in diluted exhaust gas when the ELPI main unit is at room temperature (about 25°C) and when it has been heated to about 120°C. Under both conditions, a drop, albeit slight, in particle count of 0.1 μm or below could be noted from heating, and the extent of the drop was large at low load. This is ascribed to volatilization of organic ingredient particles due to heating of part of the effecter.

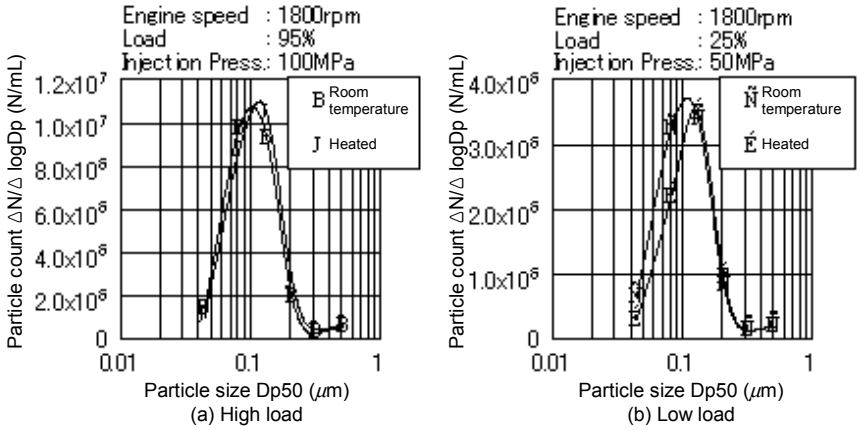

Figure 2.1-6 Effect of ELPI unit temperature on particle size distribution

2.2 Investigation of generation mechanism of FP and HAPs in engines

In order to determine the exhaust behavior of FP and HAPs in response to changes in engine operating conditions, the effect of operating conditions was studied using a single-cylinder engine. The principle specifications of the engine sample are given in Table 2.2-1. A common rail type fuel injector was equipped. As test conditions, fuel injection pressure was varied under four conditions: low speed and high load, low speed and low load, high speed and high load, high speed and low load. The fuel injection timing was fixed for each load. By adjusting the common rail pressure, fuel injection pressure was set. At high load, λ was set at 1.4 on the case of 95% load, and at low load, λ was set at 4.0 on the case of 25% load. JIS No. 2 grade diesel fuel was used as the sample fuel.

In measurements of particle size, using SMPS, samples were taken from a dilution tunnel into which the entire volume of engine exhausts had been directed. For measurements of methane, benzene and nitrous oxide, diluted air and diluted exhaust gas in the dilution tunnel were collected into a tetra bag and analyzed by gas chromatography. For measurement of aldehydes, diluted air and diluted exhaust gas in the dilution tunnel were collected into a cartridge and analyzed by liquid chromatography after removing the solvent.

Table 2.2-1 Test engine specifications

Model	Water-cooled, 4-cycle, single cylinder direct injection
Bore × stroke	ϕ108 mm × 115 mm
Displacement	1053 cm^3
Compression ratio	18.0
Intake and exhaust valve count	4
Fuel injection	Common rail
Injection nozzle hole diameter	ϕ0.18
Injection nozzle hole number	5

(1) Effect of engine operating conditions on particle size

Figure 2.2-1 presents the effect of each operating condition on particle size when the fuel injection pressure has been changed. Particulate matter (PM) emission per unit fuel consumption under the same conditions is shown in Figure 2.2-2.

When injection pressure has been increased, the particle count per unit fuel consumption declines under all operating conditions, and the PM exhaust per unit fuel consumption is reduced. As for particle distribution, when the injection pressure is elevated, particle size becomes small under all engine conditions. This is ascribed to an improvement in combustion due to various factors such as atomization of fuel injection resulting from an increase in injection pressure, a rise in the injection rate, an increase in the fuel injection volume during the ignition delay period, and an increase in the pre-mixed combustion percentage.

When engine speed has been changed, there are no major differences in the particle size that becomes the peak value of particle distribution.

When load has been changed, the peak value of the particle distribution at low load becomes smaller by about 30 nm than at high load. Here, at low load, two peaks are measured, and with the small particle size (about 30 nm), the particle count increases due to an increase in injection pressure and a drop in engine speed. It is suspected that this is due to condensation of fuel not yet burnt, but this must be investigated in future.

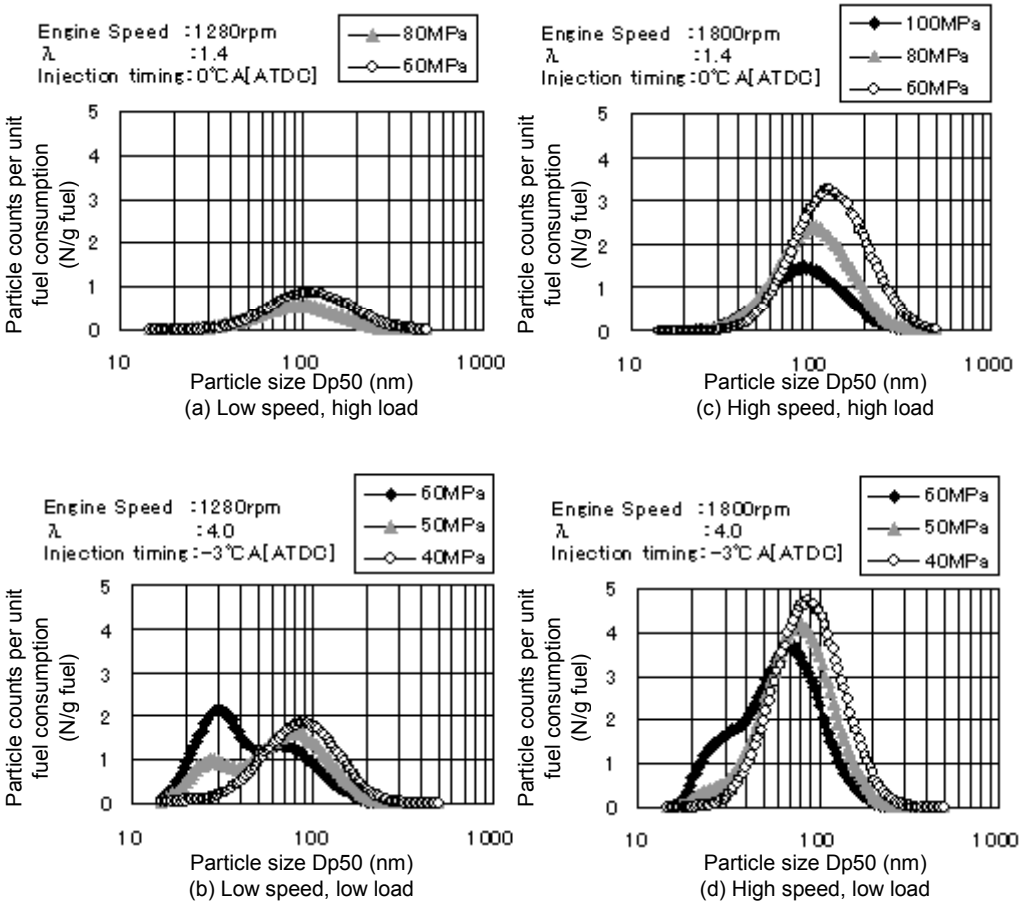

Figure 2.2-1 Effect of fuel injection pressure on particle size distribution

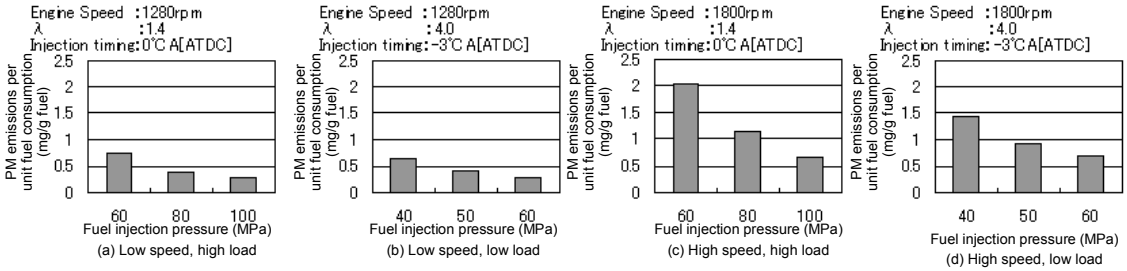

Figure 2.2-2 PM emissions per unit fuel consumption

(2) Effect of changes engine operating conditions on HAPs emissions

Figure 2.2-3 shows the effect on emissions of methane, benzene, formaldehyde, acetaldehyde and dinitrogen oxide when fuel injection pressure has been changed under each operating condition.

Methane and benzene emission rates declined or remained the same when injection pressure was high, but no clear trend could be noted in relation to speed or load.

Formaldehyde and acetaldehyde emission rates exhibited the same trend as THC emission rate at high speed and the same as CO at low speed.

When injection pressure was high, dinitrogen oxide emissions rate declines at low speed, but at high speed it increases.

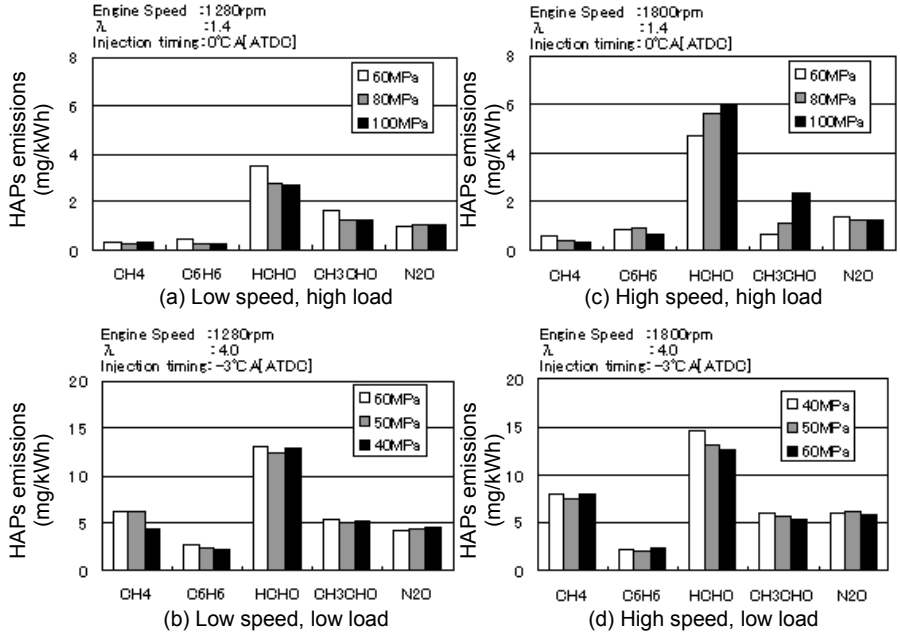

Figure 2.2-3 Effect of fuel injection pressure on HAPs emissions

2.3 Investigation of the effect of fuel and engine technologies on FP and HAPs emissions

2.3.1 Effect of fuel properties and components on FP and HAPs emissions

(1) Test method

A direct injection diesel engine adapted Japanese 1997 emission regulations (short-term regulations) was used as the test engine, and a D13 mode step test was performed.

Using heated piping, CO, THC, NOx and CO_2 were introduced directly into an exhaust gas analyzer. Using a flow divider type dilution tunnel, PM was collected in a filter and its weight was measured. In PM analysis, SOF was quantified by Soxhlet extraction and the remained was treated as soot. In ELPI also, measurements were taken after dilution. Samples of aldehyde were taken from the dilution tunnel through heated piping and collected into a cartridge. Upon completion of testing, the cartridge was extracted with acetonitrile, and aldehyde was identified by HPLC.

Typical properties of the test fuel are given in Table 2.3-1. Here test fuel was used in which distillation, component and cetane number were changed. For DF-1 to DF-4, cetane number was varied while distillation was kept constant. For DF-5 and DF-6, in contrast to DF-1 to DF-4, distillation was made heavy in nature and cetane number was elevated. In contrast to these, in DF-7 and DF-8, the aromatic contents were reduced.

Table 2.3-1 Test fuel specifications

			DF-1	DF-2	DF-3	DF-4	DF-5	DF-6	DF-7	DF-8
Density (15°C)		kg/m^3	835.0	835.0	846.0	846.0	834.0	834.0	807.0	807.0
Viscosity (@30)		$mm^2/sec.$	3.355	3.355	3.480	3.480	3.024	3.024	1.852	1.852
Distillation	IBP	°C	174.0	174.0	176.0	176.0	175.0	175.0	148.0	148.0
	10%	°C	203.0	203.0	207.0	207.0	215.0	215.0	170.0	170.0
	50%	°C	266.0	266.0	267.0	267.0	284.0	284.0	214.0	214.0
	90%	°C	348.0	348.0	352.0	352.0	339.0	339.0	300.0	300.0
	EP	°C	379.0	379.0	381.0	381.0	364.0	364.0	342.0	342.0
Cetane index			54.5	54.5	50.0	50.0	56.3	56.3	48.7	48.7
Cetane number			56.1	59.2	48.6	51.7	61.2	63.5	51.2	54.3
HPLC	Saturates	mass %	71.9	71.9	72.3	72.3	75.0	75.0	83.8	83.8
	Olefines	mass %	1.0	1.0	0.8	0.8	0.4	0.4	0.4	0.4
	Total Aromatics	mass %	27.1	27.1	26.9	26.9	24.6	24.6	15.8	15.8
	1-Aromatics	mass %	19.7	19.7	19.7	19.7	21.3	21.3	14.1	14.1
	2-Aromatics	mass %	6.1	6.1	6.0	6.0	2.9	2.9	1.7	1.7
	3-Aromatics	mass %	1.3	1.3	1.2	1.2	0.4	0.4	0.0	0.0
Sulfur concentration		mass %	0.05	0.05	0.05	0.05	0.03	0.03	0.04	0.04

(2) Test results

Figure 2.3-1 presents the results of D13 mode step test, compiled by relationship to fuel properties and aldehyde emission. In these results, no effect on aldehyde emission by distillation or component could be observed. On the other hand, when cetane number was elevated, a relatively linear pattern of decline in aldehyde was noted. From these results, it can be suggested that increase of the cetane number is effective in reducing aldehydes by fuel.

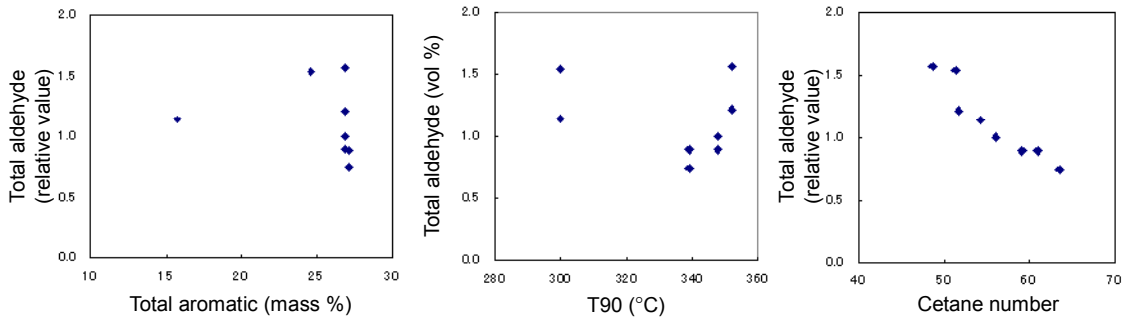

Figure 2.3-1 Fuel properties versus aldehyde emissions

2.3.2 Effect of engine technology on FP and HAPs emissions

(1) Test method

A small diesel engine adapted Japanese 1999 emission regulations (long-term regulations) was used as the test engine, and tests were the 10.15 mode simulation that method used at JCAP.

Table 2.3-2 Operating conditions of 10.15 mode simulation method

Test condition	Speed rpm	Torque Nm	Weight %
Idle			32.3
40 km/h R/L	1300	25	18.9
70 km/h R/L	1800	39	13.4
Typical acceleration point	1700	88	12.6
Maximum acceleration point	2100	108	0.7

Table 2.3-3 Exhaust gas test results

(1) CO g/h

Test conditions		
1	Idle	0.83
2	40 km/h R/L	1.09
3	70 km/h R/L	0.39
4	Typical acceleration point	0.07
5	Maximum load	0.13

(2) THC g/h

Test conditions		
1	Idle	0.11
2	40 km/h R/L	0.29
3	70 km/h R/L	0.50
4	Typical acceleration point	0.14
5	Maximum load	0.13

(3) NOx g/h

Test conditions		
1	Idle	0.09
2	40 km/h R/L	0.75
3	70 km/h R/L	3.23
4	Typical acceleration point	3.62
5	Maximum load	5.55

(4) PM g/h

Test conditions		
1	Idle	0.19
2	40 km/h R/L	0.23
3	70 km/h R/L	1.45
4	Typical acceleration point	2.03
5	Maximum load	1.24

(5) SOF g/h

Test conditions		
1	Idle	0.08
2	40 km/h R/L	0.13
3	70 km/h R/L	0.33
4	Typical acceleration point	0.43
5	Maximum load	0.40

(6) Soot g/h

Test conditions		
1	Idle	0.10
2	40 km/h R/L	0.10
3	70 km/h R/L	1.12
4	Typical acceleration point	1.60
5	Maximum load	0.84

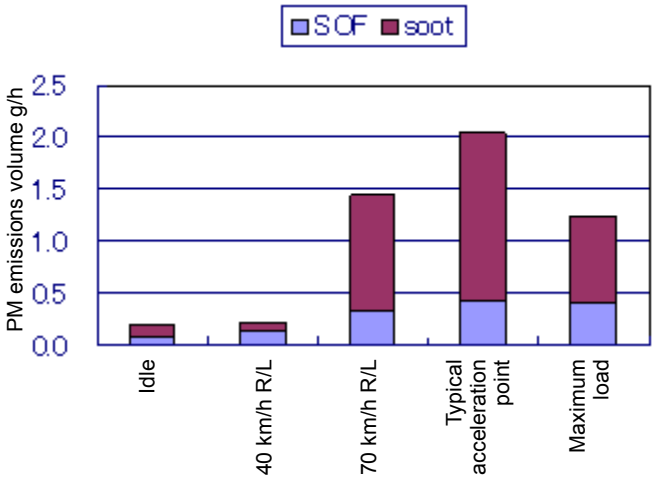

Figure 2.3-2 SOF and soot percentages

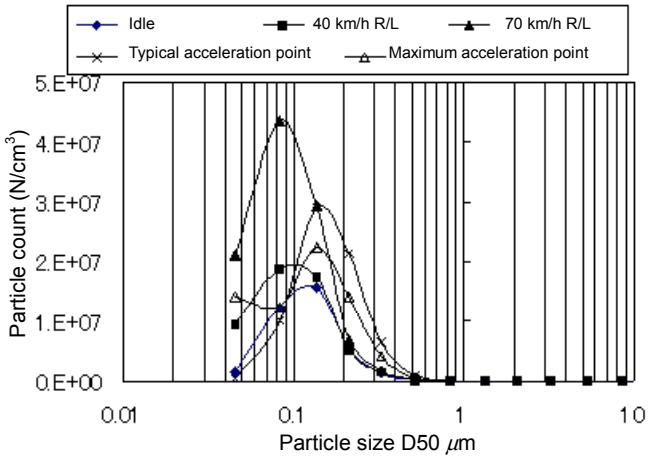

Figure 2.3-3 Particle size distribution under each operating condition

Exhaust gas measurement results are given in Table 2.3-3. In comparison to idle condition, PM emission increased at 40 km/h R/L. PM ingredient percentages under each operating condition are given in Figure 2.3-2. In this way, the increase in PM centered on the SOF component and soot remained unchanged. On the other hand, at the typical acceleration point, 70 km/h R/L, PM emissions increased further. SOF emissions volume also increased, but the increase in the soot emissions is especially large and the percentage of soot in PM is extensive.

Measurements of particle size distribution by ELPI are given in Figure 2.3-3. From these measurements it appears that under high load (70 km/h R/L, typical acceleration point), as compared to low load (idle, 40 km/h R/L), PM of small particle size spreads more widely. Nevertheless, as mentioned previously, under operating conditions with these high loads, the percentage of soot becomes high in comparison to SOF, and for this reason, it is believed that the question of whether differences in particle size distribution stem from the percentages of SOF and soot must be resolved.

3. Results of Empirical Research

From the aforementioned test results, valuable technological information could be obtained on the generative factors and the generative behavior of PM and HAPs emitted from engines, as well as on technologies for reducing the same. An outline of the essentials is presented below.

3.1 Investigation and establishment of method for measuring FP in exhausts

The following results were obtained from an evaluation of each measurement method and of incidental conditions at the time of measurement, etc.

(1) Stainless steel, electro-conductive silicone and vinyl chloride were suitable in the sampling tube at the time of FP measurement.

(2) As conditions of FP measurement, effects from diluted air temperature and humidity were observed under low-load operation in SMPS, and in ELPI, the effect of distillation duration was noted.

3.2 Investigation of the generation mechanism of FP and HAPs emissions in engines

From an investigation of the relationships between each operational control factor, FP and HAPs, using a single-cylinder engine, the following results were obtained.

(1) When fuel injection pressure was elevated, particle size became smaller. At low loads, a peak near 30 nm, believed to be due to condensation of unburned fuel, was observed. And at this particle size, the number of emitted particles increased when the injection pressure was set high.

(2) The emission characteristics of HAPs when fuel injection pressure was elevated followed different trends depending on engine speed and load, and they were not clear.

3.3 Effect of fuel and engine technologies on emissions of FP and HAPs emissions

(1) A correlation was observed between aldehyde emissions and cetane number. It was suggested that elevating the cetane number is effective in reducing aldehydes.

(2) In the distribution of particle sizes, the trend was toward large particle size under operating conditions with abundant SOF and toward smaller particle size under operating conditions with abundant soot.

4. Future work

In view of the results obtained this fiscal year, it is believed that further investigations are required for the following items.

(1) Establishment of a method for analyzing ingredients of medium and high boiling points among HAPs and application of the method to engine exhaust gas emission measurements.

(2) Detailed analysis of particle properties of each particle size to understand how to exhaust fine particles.

(3) Comprehension of the emission actions of FP and HAPs under supercharging or EGR conditions to investigate and realize generation mechanisms.

(4) Investigation of the effect of fuel properties on FP emissions.