

the sulphur content of diesel fuel and its relationship with particulate emissions from diesel engines

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Management Group based on the work carried out by the
Special Task Force on Diesel Fuel Emissions (AE/STF-7)

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ABSTRACT

This report gives the results of a research programme to investigate the influence of diesel fuel sulphur content on diesel engine exhaust particulate emissions.

A representative range of current light-duty vehicles (nine) and heavy-duty engines (four) were tested using European test procedures with four fuels of 0.31, 0.22, 0.12 and 0.055% wt sulphur. The test fuels were produced by progressively desulphurizing the base fuel, avoiding changes in other fuel quality variables which may have influenced the emissions results.

The study found a consistent, but small influence of diesel fuel sulphur on the mass of particulate emissions from the light-duty vehicles: exhaust particulates declined about 7% as fuel sulphur content reduced from 0.3 to 0.05% wt.

For the heavy-duty engines, there was no consistent trend linking reducing particulate levels with reducing fuel sulphur content. This finding differs from the trends found in some US studies. However, those studies used sulphur doped fuels, an artificial situation considered not representative of refining practice in the production of low sulphur fuels.

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SUMMARY

CONCAWE has studied the influence of fuel sulphur content on particulate emissions from a range of light-duty diesel vehicles and a range of heavy-duty diesel engines using European test procedures.

The study used a base fuel at 0.31% wt sulphur content, which was progressively desulphurized to 0.22, 0.12 and 0.055% wt sulphur, avoiding changes in other characteristics of the fuel.

The study found a consistent, but small influence of diesel fuel sulphur content on the mass of particulate emissions from light-duty vehicles. Expressed in terms of the new European test cycle combining the urban (ECE-15) cycle with the extra-urban driving cycle (EUDC), the following percentage reductions in particulate emission were obtained by interpolation, taking fuel at 0.3% wt sulphur as the base case:

	Percentage reduction in particulate emission
0.15% wt sulphur	4.5
0.05% wt sulphur	7

For heavy-duty diesel engines, the study was not able to establish a consistent trend of reducing particulate emissions with reducing fuel sulphur content. This finding contrasts with the literature. However, most publications to date have reported on studies in the United States, where different test procedures and sulphur-doped fuels were employed.

1. INTRODUCTION

The EC directive to limit sulphur content of diesel fuel and gas oil to maximum 0.3% wt, with the option of maximum 0.2% wt in environmentally sensitive areas, came into force on 1 January 1989. Further pressure to change European diesel fuel qualities has arisen because of US plans to move to maximum 0.05% wt sulphur content to meet stringent exhaust emission requirements.

European research work in this area includes an independent heavy-duty diesel emissions study by Ricardo Consulting Engineers Ltd in the UK, sponsored by the Dutch Government Ministry of the Environment (VROM), using fuels supplied by CONCAWE. This study (1) primarily investigated fuel ignition quality and volatility effects upon diesel emissions, although two fuels were included to investigate the influence of diesel fuel sulphur content on particulate emissions.

In order to investigate the sulphur influence in detail, the CONCAWE Automotive Emissions Management Group formed a Special Task Force (AE/STF-7) with the specific purpose of assessing the influence of sulphur content on the amount and nature of particulate emissions from a wide range of diesel vehicles (cars and light vans) and heavy-duty diesel engines.

This report summarizes the CONCAWE findings on the influence of diesel fuel sulphur content on diesel engine particulate emissions. The report does not cover the entirely separate issue of the influence of diesel fuel sulphur content on the performance of oxidation catalysts for diesel vehicles and engines.

The work described was carried out in the laboratories of six CONCAWE member companies. Additional heavy-duty engine studies were carried out by Ricardo in the UK, as an integral part of the programme. Every attempt was made to standardize test and analytical procedures throughout the programme such that a consistent body of data be made available.

Detailed analytical data on the composition of particulates, together with an appraisal of the available analytical procedures, will be given in a separate report.

2. CHOICE OF DIESEL VEHICLES AND ENGINES FOR THE PROGRAMME

The choice of diesel vehicles and heavy-duty engines for the programme was made, within the constraints of availability to CONCAWE member companies, in order to represent a wide range of current European diesel engines, covering all types including naturally aspirated (NA), turbocharged (TC), turbocharged and intercooled (TC/IC), indirect injection (IDI) and direct injection (DI) engines. In addition, one North American engine was included in the programme in view of its increasing applicability to the European market.

The characteristics of the vehicles and engines employed in the programme are:

VEHICLES

VEHICLE No.1	1.6 LITRE	NA IDI	PASSENGER CAR
VEHICLE No.2	1.9 LITRE	NA IDI	PASSENGER CAR
VEHICLE No.3	2.4 LITRE	NA IDI	LIGHT VAN
VEHICLE No.4	2.4 LITRE	TC IDI	PASSENGER CAR
VEHICLE No.5	2.5 LITRE	TC IDI	PASSENGER CAR
VEHICLE No.6	2.5 LITRE	TC IDI	PASSENGER CAR
VEHICLE No.7	2.5 LITRE	NA DI	LIGHT VAN
VEHICLE No.8	1.9 LITRE	TC DI	PASSENGER CAR
VEHICLE No.9	2.0 LITRE	TC DI	PASSENGER CAR

ENGINES

ENGINE No.1	6.0 LITRE	NA DI
ENGINE No.2	6.0 LITRE	TC/IC DI
ENGINE No.3	9.6 LITRE	TC/IC DI
ENGINE No.4	12.7 LITRE	TC/IC DI

Vehicles No.4 and No.6 and Engine No.4 were fitted with electronic control systems optimizing fuel injection timing for given engine operating conditions.

3. PROVISION OF TEST FUELS FOR THE PROGRAMME

A fundamental problem which arises in studies of the influence of changes in fuel characteristics on engine performance lies in the inherent intercorrelation between fuel properties. As a consequence, it is frequently difficult to change one fuel characteristic without altering other properties of the fuel.

In this study, great care was taken, whilst varying diesel fuel sulphur content over the range 0.3% wt down to 0.05% wt, to maintain all other fuel quality parameters constant. The test fuels were prepared in a refinery of a CONCAWE member company, by increasingly severe desulphurization of a light gas oil derived from a Middle East crude. This method of desulphurization was chosen in preference to intermediate blending of feed and low sulphur product, as being representative of actual or potential refining practice. The process conditions and catalyst used in preparation of the test fuels are considered to be typical of modern European refinery practice.

The sulphur content range of 0.3% wt down to 0.05% wt was chosen to cover existing EC legislation and to include the proposed US situation with respect to diesel fuel quality. In all other respects the aim was to produce a series of fuels having typical European qualities, as far as this is possible in a widely varying market.

Analytical data on the four fuels used in the programme are given in Table 1. These data are mean values calculated from individual results obtained in the laboratories of the CONCAWE member companies involved in the programme.

The sulphur contents of the fuels cover the required range and are evenly distributed throughout the range. Because the fuels were prepared by desulphurization of a light gas oil feedstock, and not by a blending technique, their distillation ranges are relatively narrow. The light gas oil feedstock was prepared from paraffinic crude oil (Middle East source) and consequently has a cetane number above the range predicted by CONCAWE for future European diesel fuels (2). However, the cetane number (and aromatic content determined by IP high performance liquid chromatography techniques) are remarkably consistent for this set of fuels and it is considered that a comparison of their emissions performance is valid in the European context.

4. PROVISION OF LUBRICATING OIL FOR THE PROGRAMME

In order to eliminate any influence of lubricating oil quality on the amount or nature of particulates generated in this programme, a commercial lubricating oil was chosen which satisfied the short-term test requirements of each engine employed in the programme. This oil, which was of SAE 15W/40 quality meeting API SF and DB 227.1 requirements, was used throughout the test programme. Inspection data on the unused oil are:

REFERENCE GRADE	L88/1801 SAE 15W/40, API SF, DB 227.1	
Pour Point	°C	-24
Sulphur Content	% wt	0.92
KV @ 40°C	mm ² /sec	106
KV @ 100°C	mm ² /sec	14.3
KV @ 150°C	mm ² /sec	5.70
Viscosity Index		138
NOACK (DIN 51581) 1 hour, 250°C	% wt	12.0
Phosphorus	% wt	0.115
Calcium	% wt	0.206
Magnesium	% wt	0.093
Zinc	% wt	0.106
Hydrocarbon Distribution	C ₂₃ to greater than C ₄₂	
IBP	°C	288
FBP	°C	537

5. VEHICLE AND ENGINE TEST PROCEDURES

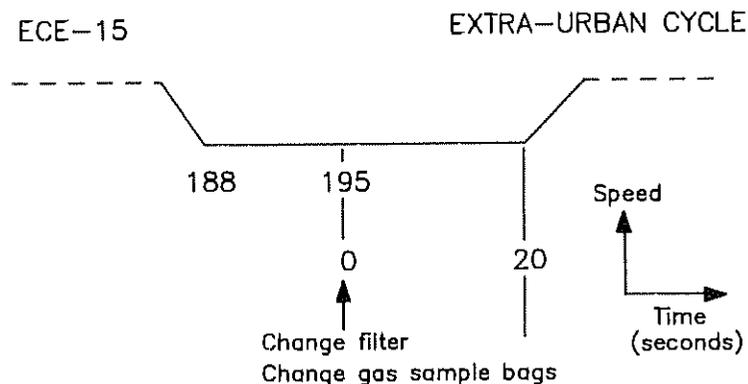
The procedures used for this European programme are those either adopted by, or proposed for, current and pending EC legislation covering emissions from diesel vehicles.

Thus for the vehicle tests the ECE-15 test cycle was used followed by the EUDC (extra urban driving cycle). All test procedures were carried out in duplicate, using a random order of fuel testing in each laboratory. Vehicle tests were carried out in five CONCAWE member companies' laboratories, the sixth laboratory concentrating on heavy-duty engine testing.

Details of the vehicle test procedures used are as follows:-

1. Change lubricating oil to standard STF-7 quality requirement (Section 4).
2. Carry out a lubricating oil and injector nozzle pre-conditioning programme using CEC reference fuel RF-03-A-84, with a total driving distance of 1000 km, duration two days (500 km/day), 33% running on freeway at 130 km/h, or about 165 km/day, 33% on road at average 60 km/h (3).
3. Pre-condition the vehicle under test using three EUDC high speed cycles, followed by an eight hour soak period.
4. Cold start, followed by the current ECE-15 procedure, measuring gaseous and particulate emissions.
5. Immediately change the particulate filter, and proceed with the high speed cycle (4), again measuring gaseous and particulate emissions. The changeover from ECE-15 to EUDC is shown schematically in Fig. 1.

Fig. 1 Schematic Diagram for Changeover from ECE-15 to EUDC driving cycle



For the diesel engine tests it was necessary to make assumptions concerning test procedures, based on the ECE-R49 13-mode procedure for gaseous emissions from heavy-duty engines. These assumptions were necessary since European legislation on heavy-duty particulate emissions had not been formulated when drawing up this programme. The situation was however complicated by the requirement to collect particulate over the 13-mode R49 cycle for subsequent chemical analysis.

The proposed European diesel particulate measurement technique (5) specifies that "for particulate matter one sample over the complete test cycle shall be taken", and that "the particulate sample flow must be adjusted at each mode to take account of the modal weighting factor and to give proportionate sampling".

Two approaches were used in this programme, depending upon the particulate collection system in use. In one laboratory, thirteen separate filter papers were used, one for each mode of the R49 procedure, and combined for analysis. Weighting factors for each individual mode were taken into account by varying the sampling time in each mode.

In the second laboratory a single filter paper was used according to the procedure developed by Berlin University (5).

Tests in the third laboratory demonstrated the equivalence of the 13 paper and single paper techniques. The actual procedure used is as follows:-

1. Drain, flush and re-fill with lubricating oil to standard STF-7 quality requirement (Section 4) at the start of each pair of duplicate tests.
2. Carry out R49 type 13-mode tests measuring gaseous and particulate emissions, either by changing particulate filters at the end of each mode or by using a single filter throughout the 13-mode procedure.

6. CHEMICAL ANALYSIS OF DIESEL PARTICULATES

Diesel particulate samples were generated over ECE-15/EUDC and modified ECE R49 cycles as discussed in the previous Section. Glass fibre filter papers (Whatman GF/A) were preferred for particulate collection to facilitate determination of particulate carbon content. Particulate-loaded filter papers were stored before analysis in covered Petri dishes, kept in a refrigerator below 0°C.

Particulate samples were analyzed for total carbon, sulphate, and hydrocarbons in the fuel and lubricating oil boiling ranges. In addition to the analyses given above, selected samples were analysed for total and individual polycyclic aromatic hydrocarbons. The data obtained in these analyses will be discussed in a further report.

Fuel sulphur to sulphate conversion is however discussed here, and the method used for sulphate determination is as follows.

(In accordance with other literature references, this report refers to the popular term "sulphate" although the contribution of sulphur to particulates is predominately in the form of sulphuric acid).

6.1 Total Sulphate (6)

A weighed "pie" sector or slice of each filter paper was taken for this analysis since experiments had shown that the distribution of sulphate across the radius of the filter decreases slightly from perimeter to centre. This is considered possibly to arise from a temperature gradient across the filter. This sector of filter paper was extracted with 20:80 iso-propanol:water, and the sulphate in the extract determined by ion chromatography using a Dionex 2000i ion chromatograph. The procedure was also applied to a sector cut from an unused filter and the "blank" value subtracted from the measured value before reporting.

7. EMISSIONS DATA - CARS/LIGHT COMMERCIAL VEHICLES

Few problems were reported by the five CONCAWE members involved in applying the ECE-15/EUDC procedures to a wide range of diesel vehicles (see Section 2). The gaseous and particulate emissions data obtained from duplicate runs are shown in the form of mean values for each vehicle/fuel combination in Tables 2 to 9.

The first four Tables (2 to 5) give separate data (in g/test) for ECE-15 and EUDC cycles. There is a general decrease in CO and HC on changing from the ECE-15 to the high speed EUDC cycle, an increase in NO_x (apart from vehicle number 3), whilst the change in particulate level is less regular.

In the second set of Tables (6 to 9) the data are expressed in terms of total emissions (in g/km) over the combined ECE-15 and EUDC cycles, in order to conform to expected changes in European emissions legislation. The "equivalent distance" of the combined cycles has been taken to be 11 km, and this factor used to calculate emissions in g/km.

For all the data generated, there is a wide range of emissions levels covering the different engines and fuel injection systems under test. The ranges found for individual emissions are set out below:

APPROXIMATE RANGES OF EMISSIONS VALUES FOR LIGHT DUTY DIESEL ENGINES/INJECTION SYSTEMS

	ECE-15 CYCLE g/test	EUDC g/test	COMBINED CYCLES g/km
CO	3.5-9.0	1.9-7.0	0.5-1.4
HC	0.8-4.8	0.5-3.8	0.1-0.8
NO _x	2.2-14.0	3.1-22.0	0.5-3.3
PARTICULATE	0.3-2.4	0.5-4.0	0.08-0.6

Using the combined cycle data, the percentage changes across the range of vehicles are as follows:-

CO	180%
HC	700%
NO _x	560%
PARTICULATE	650%

The final column in Tables 2 to 9 shows the influence of diesel fuel sulphur content on the level of each individual emission. The percentage change has been calculated by summing the individual emission values for all nine vehicles tested, and relating this sum to that obtained for the high sulphur (0.3% S) fuel as base or reference fuel. In this way, the influence of fuel sulphur content may be simply and directly expressed for a range of vehicles typical of the diesel market.

For ease of reference the percentage changes obtained on the combined cycle are repeated below:

INFLUENCE OF DIESEL FUEL SULPHUR CONTENT ON OVERALL EMISSION VALUES (DIESEL VEHICLES), EXPRESSED AS PERCENTAGE CHANGE

FUEL SULPHUR CONTENT % wt	0.31	0.22	0.12	0.055
CO	0	+ 1.1	- 0.6	- 0.9
HC	0	- 1.8	+ 2.1	- 5.9
NO _x	0	+ 1.7	+ 4.3	- 1.6
PARTICULATE	0	- 2.9	- 4.3	- 7.4

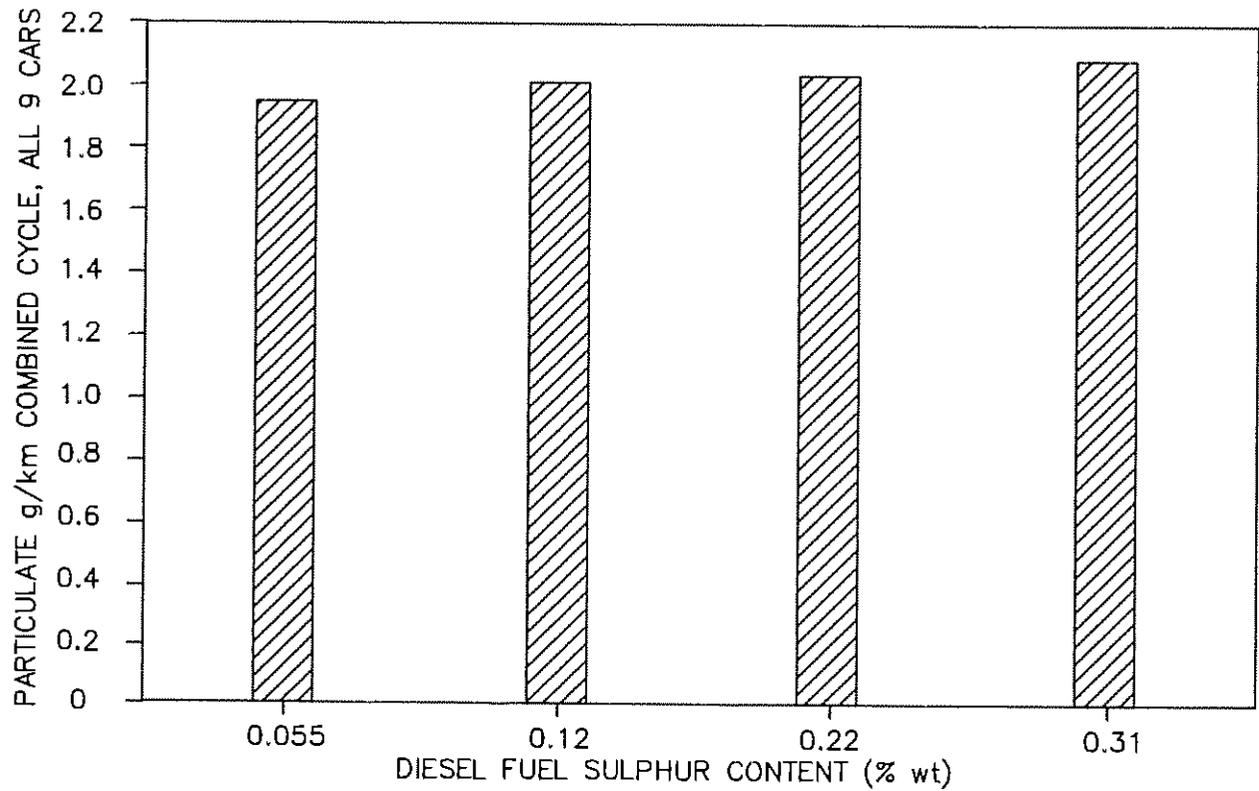
+ Increase in level compared with reference fuel

- Decrease in level compared with reference fuel

Whilst the statistical significance of these percentage changes is discussed in Section 9, they should also be considered in comparison with the engine to engine changes set out earlier in this Section. Variations in particulate levels due to changes between different vehicles are of the order of 100 times greater than the influence of sulphur level.

The data are set out in the form of a bar chart in Fig. 2

Fig. 2 Influence of Fuel Sulphur Content on Particulates from Light-duty Diesel Engines



8. EMISSIONS DATA HEAVY-DUTY DIESEL ENGINES

Prior to carrying out this programme, two of the three laboratories concerned had only limited experience in particulate testing and analysis from heavy-duty diesel engines.

The characteristics of the engines which were available for this programme were discussed in Section 2. The gaseous and particulate emission results obtained under ECE R49 conditions are shown in the form of mean values for each engine in Tables 10 to 13.

As for the light-duty vehicles, there is considerable variation of emission level between the different engines under test. The ranges found for individual emissions are set out below. Also given are the current limits for gaseous emissions as set out in EC Directive 88/77.

APPROXIMATE RANGES OF EMISSIONS VALUES FOR HEAVY-DUTY DIESEL ENGINES/INJECTION SYSTEMS

g/kWh	ECE R49 EMISSIONS	LIMIT ACCORDING TO EC DIRECTIVE 88/77
CO	1.2 - 5.9	11.2
HC	0.06 - 1.3	2.4
NO _x	6.8 - 13.9	14.4
PARTICULATE	0.18 - 0.68	-

The percentage changes across the range of engines are as follows:

CO	390%
HC	2000%
NO _x	100%
PARTICULATE	280%

The final column in Tables 10 to 13 shows the influence of diesel fuel sulphur content on the level of each individual emission. The percentage change has been calculated by the same method as for the light-duty vehicles, giving a direct expression for a range of engines typical of the diesel market.

For ease of reference the percentage changes obtained on the ECE R49 cycle are repeated below.

INFLUENCE OF DIESEL FUEL SULPHUR CONTENT ON OVERALL EMISSION VALUES (DIESEL ENGINES), EXPRESSED AS PERCENTAGE CHANGE

FUEL SULPHUR CONTENT % wt	0.31	0.22	0.12	0.055
CO	0	+11.0	-0.4	-5.4
HC	0	-2.2	-0.9	+6.7
NO _x	0	-8.1	+1.9	-3.9
PARTICULATE	0	+9.0	+2.3	-0.1

- + Increase in level compared with reference fuel.
- Decrease in level compared with reference fuel.

It will be seen that there is no overall trend in relation to diesel fuel sulphur content, and the percentage changes should be considered in relation to the engine to engine changes given earlier in this section.

Only one engine (engine No. 2, Table 13) showed a linear trend of increasing particulate emission with increasing fuel sulphur content. For this engine alone, the reduction in particulate emission which was obtained on changing from 0.3% wt sulphur fuel to 0.05% wt sulphur fuel was about 10%. It must be stressed however that the other three engines tested did not show any trend in particulate emission with fuel sulphur content.

STATISTICAL SIGNIFICANCE OF EMISSIONS DATA

A statistical analysis of the data generated on the light-duty vehicles was carried out in order to establish whether the differences between gaseous and particulate emissions for the 0.3% wt sulphur fuel and the other fuels are significant once the effect of the car type has been removed.

Each vehicle gives a different mean value for the emission species, but the variability of the replicates is assumed to be the same. The tests of differences were applied to the emission species for each cycle type and the variability of these means was calculated by taking into account the mean car effect. The comparisons involved an F-test to check whether the variances of the data sets being compared have the same variance, and subsequently a t-test to check whether the mean difference is significant.

The statistical data for the combined ECE-15/EUDC cycles are given in Table 14, in terms of F-ratio probability, the difference in emission between fuels, the t-ratio and the t-ratio probability, with significant differences shown by an asterisk.

The probabilities associated with the F-ratio and the t-ratio are the probabilities of having this difference if the data sets are from the same underlying distribution. When the probabilities are small (5% or less) it can be assumed that the underlying distributions are different, and that the observed differences are significant.

It can be seen that there is a consistent and significant effect across the range of fuels studied for the particulate emission case. For the gaseous emissions there is no consistent and significant fuel quality influence.

A similar statistical exercise was carried out on the data generated from the heavy-duty vehicles over the ECE R49 cycle, Table 15.

Although several of the observed differences were found to be statistically significant, no consistent trends with fuel sulphur content were observed.

10. ANALYSIS OF PARTICULATES - SULPHATE CONTENT

Previous studies (8,9) have highlighted the contribution of bound sulphate to diesel particulate emission as collected under the conditions of standard test procedures. As described in Section 6, a single sulphate determination was carried out for each vehicle and engine test in this programme. Tables 16 and 17 show the total sulphate measured on the filter paper as a percentage of the total particulate, for each vehicle and engine and for each fuel. There is considerable variation between vehicles, arising from the large differences in particulate emission rates. There is also an underlying trend towards an increase in retained sulphate (as a percentage of total particulate) as fuel sulphur content increases. Within the dataset there is however considerable variation, possibly arising from the non-linearity of sulphate retention across the filter paper, see Section 6.1. The cause is unlikely to be the sulphate determination itself, which is known to be highly accurate.

For the heavy-duty engines tested under R49 conditions, Table 19, there is again an underlying trend associated with sulphur content.

An alternative way of presenting the sulphate data is to consider the sulphate trapped on the filter paper as a percentage of the fuel sulphur consumed by the vehicle or engine during the test procedure. This is simply calculated from the known fuel consumption. Tables 18 and 19 show this approach for the light-duty vehicles and the heavy-duty engines, respectively. Using this method of presentation, there is a definite trend to greater trapping efficiency of sulphate as fuel sulphur content decreases.

It should be emphasised that the sulphate trapping rates are low: the bulk of the sulphur present in the fuel is oxidised to gaseous products which are not retained on the filter under the conditions of the R49 procedure. (Sulphate in this programme refers only to sulphate: any contribution from associated water has not been included in the calculations).

11. COMPARISON OF CONCAWE STF-7 RESULTS WITH PUBLISHED DATA

As a result of a search of the published literature CONCAWE believes that there are very few available data relating particulate emission levels for light-duty diesel vehicles with diesel fuel sulphur content.

The French motor industry organization UTAC has published (7) emission data obtained on three diesel passenger cars, using the fuels employed in the VROM heavy-duty programme. Annex 5 of the UTAC report gives data on particulate emissions comparing VROM fuels H and E, respectively 0.29 and 0.07% wt sulphur (fuel H was prepared from fuel E by doping with benzothiophene). Over this range of sulphur content UTAC was unable to demonstrate any significant change in level of particulate emission when measured to the requirements of European legislation. This compares with the 7% reduction found in the CONCAWE study over a similar range of fuel sulphur content and a wider range of vehicles.

For heavy-duty diesel engines there are considerably more published data available, primarily reflecting the US interest in this area. European data are however also available from a recent programme in which the fuels were supplied by CONCAWE. In this programme, commissioned by the Dutch Environment Ministry (VROM) (1) the fuels H and E referred to above were tested on a wide range of heavy-duty engines, seven in total. The ECE R49 particulate emissions data have been abstracted from VROM documentation supplied to CONCAWE and are shown in Table 20. It will be seen that a mean reduction of particulate emission of 12.4% was achieved on reducing fuel sulphur content from 0.29 to 0.07 (achieved as stated above by doping the lower sulphur fuel). (Engine V.7 in the VROM programme was the same as engine 4 in the CONCAWE programme).

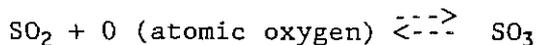
Turning to the US data, there are a number of papers which provide data for purposes of comparison. The first of these is by Wall and Hoekman (8), which deals primarily with steady state emissions data obtained on one engine. Wall and Hoekman used a regression technique to separate the influence of fuel variables over a wide range of fuels. Using their Fig. 12, it is possible to estimate a reduction of about 27% in particulate emission (expressed in g/kWh) when reducing fuel sulphur content from 0.3 to 0.05% wt, the range considered in this CONCAWE programme.

Barry et al (9) also found an influence of fuel sulphur content on particulate emissions. However, their work was carried out over the range 0.25 to 1.0% wt fuel sulphur content, and is thus outside the range of interest in Europe.

In view of the lack of correlation with fuel sulphur content found in this CONCAWE programme for heavy-duty engines, it is interesting to note the comment by Wall, Shimpi and Yu (10) that at low sulphur levels, other effects (such as tunnel background particulate and sulphur/sulphate measurement errors) can severely distort the observed sulphur effect on directly emitted particulate. Using Fig. 3 from (10), and converting to a g/kWh basis it is possible to estimate a reduction of about 14% in particulate emission over the range of 0.3 to 0.05% wt, using sulphur-doped fuels and US transient cycle testing conditions.

Reference (10) gives a statistical treatment of the calculation procedure used to determine the amount of trapped sulphate on the filter paper as a percentage of fuel sulphur consumed. It is postulated that at low fuel sulphur levels dramatic increases in percentage trapped rate can arise from measurement variability. This may be the reason for the increase in calculated trapped sulphate percentage shown in Table 18 for light-duty vehicles.

There are two possible explanations which may account for the observed behaviour. The first is that under the conditions of test, the filter paper has a fixed capacity to absorb sulphate as sulphuric acid vapour. Once the paper (or particulate) becomes saturated with sulphate further absorption becomes impossible and no increase in mass due to sulphate will be recorded. The second is that in the absence of a catalyst SO_3 is formed from SO_2 and atomic oxygen according to the following equilibrium reaction



At a given reaction temperature the absolute amount of SO_3 formed (eg. in moles) depends on the available moles of SO_2 and atomic oxygen. The amount of SO_2 is linearly related to the sulphur content of the fuel. However, the amount of atomic oxygen formed by dissociation of molecular oxygen is constant at a given temperature and independent of the sulphur content of the fuel. Thus the number of moles of SO_3 formed per mole of SO_2 will decrease with increasing SO_2 content in the exhaust gas in view of an increasing lack of atomic oxygen.

A more recent programme is that of Baranescu (11). Using data from Tables 7/8/9 (11) and again converting to a g/kWh basis it is possible to estimate a reduction of about 20% in particulate emission over the range 0.29 to 0.05% wt sulphur. As in previous work, sulphur doping was used with US transient emissions testing.

The final programme which provides comparative data is that carried out by the Southwest Research Institute on behalf of the Coordinating Research Council (12). Data were generated on three heavy-duty engines using doped sulphur fuels. Using data from Tables 28, 36 and 45 (12) it is possible, after conversion to a g/kWh basis, to estimate a reduction of about 12% in particulate emission over the range 0.29 to 0.05% wt sulphur.

Thus programmes carried out in the USA, using sulphur doped fuels and US transient test procedures, have found a consistent influence of fuel sulphur content on particulate emissions. This amounts to a reduction of about 12 to 27% (depending on the programme) in the amount of particulate emitted when reducing fuel sulphur content from 0.30 to 0.05% wt. This consistent behaviour has not been found in this CONCAWE programme, apart from one engine out of four tested which showed about 10% reduction in particulate emission over this range. The reasons for this difference are not immediately obvious.

One fundamental difference which should be recognised however is that the fuels used in the CONCAWE programme were prepared by desulphurization of a high sulphur basestock, which would seem to be a preferable technique to the approach of doping a low sulphur basestock which has been widely adopted in the USA. Doping represents an entirely artificial situation, whereas the approach adopted by CONCAWE represents refining practice in the production of low sulphur distillate fuels.

12.

INFLUENCE OF A REDUCTION IN DIESEL FUEL SULPHUR CONTENT ON PARTICULATE EMISSION FROM EUROPEAN DIESEL VEHICLES

The data generated in this CONCAWE programme suggest that the following reductions in particulate emission may be estimated to arise from a reduction in fuel sulphur content over the range 0.30 to 0.05% wt. These data are derived by calculating mean values from Table 9 and interpolating for the intermediate sulphur level.

For light-duty vehicles :-

		Particulate mass mean of 9 vehicles g/km
Base case	0.30% wt sulphur	0.236
Intermediate case	0.15% wt sulphur	0.227
Low sulphur case	0.05% wt sulphur	0.220

For heavy-duty vehicles (using data generated on one engine only, engine No. 2 Table 13, and considering this to be a maximum influence of fuel sulphur content):-

		Particulate mass g/kWh
Base case	0.30% wt sulphur	0.490
Intermediate case	0.15% wt sulphur	0.461
Low sulphur case	0.05% wt sulphur	0.441

These emission values are considered to be typical for a modern turbo-charged and intercooled engine, and are intermediate between particulate emissions from older naturally aspirated engines and newer low emission engines designed to meet US specification requirements. In the absence of further data, they can thus be assumed to be representative of the European heavy-duty vehicle parc.

13.

CONCLUSIONS

Using the new European test cycle combining the urban (ECE-15) cycle with the extra-urban driving cycle (EUDC) this study has found a consistent influence of diesel fuel sulphur content on particulate emission from a range of light-duty diesel vehicles. These vehicles included indirect and direct injection engine types, passenger cars and light commercial vehicles. The mean percentage reduction in particulate emission obtained over the range of fuel sulphur content 0.3 % wt to 0.05 % wt was 7.4%.

Using the European ECE R49 13-mode steady state test procedure, this study found no overall consistent influence of diesel fuel sulphur content on particulate emission from a range of heavy-duty diesel engines. These engines, which were all of the direct injection type, included naturally aspirated and turbocharged engines with intercooling. Only one engine showed a consistent influence of fuel sulphur content, giving a percentage reduction of 10.3% in particulate emission, over the fuel sulphur content range 0.3 % wt to 0.05 % wt.

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15.

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Table 1

ANALYTICAL DATA ON TEST FUELS USED IN THE PROGRAMME
(MEAN VALUES)

Fuel No.		1	2	3	4
Sulphur content	% wt	0.055	0.12	0.22	0.31
Density @ 15°C	kg/m ³	832.7	833.0	834.2	835.1
KV @ 20°C	mm ² /sec	5.41	5.44	5.57	5.58
KV @ 40°C	mm ² /sec	3.44	3.44	3.46	3.47
Flash Point	°C	101	102	103	102
Cloud Point	°C	-4	-4	-4	-4
CFPP	°C	-6	-6	-6	-6
Water Content	ppm	53	58	62	76
Copper corrosion	-	1A	1A	1A	1A
Carbon Residue	% wt	0.01	0.01	0.01	0.01
<u>Aromatics</u>					
FIA	% vol	25.0	25.0	24.0	25.4
HPLC (a)	% wt	24.4	24.6	24.4	24.9
HPLC (b)					
Mono-aromatics	% vol	22.0	21.3	21.7	19.8
Di-aromatics	% vol	4.6	4.1	4.5	4.9
Tri-aromatics	% vol	1.2	0.8	1.4	1.5
Total aromatics	% vol	27.4	26.2	27.6	26.2
Cetane Number		58.2	58.1	58.4	58.9
Calculated Cetane Index (IP380)		60.2	60.0	59.6	59.3
<u>Distillation Data</u>					
IBP	°C	222	222	222	223
10% vol	°C	251	250	250	250
20% vol	°C	259	258	259	259
30% vol	°C	266	266	267	266
40% vol	°C	274	273	274	274
50% vol	°C	281	281	282	283
60% vol	°C	290	290	291	291
70% vol	°C	300	300	302	302
80% vol	°C	312	313	314	315
90% vol	°C	329	329	330	330
95% vol	°C	342	341	342	343
FBP	°C	354	353	354	353
Recovery	% vol	98.3	98.6	98.4	98.4
Loss	% vol	0.3	0.2	0.1	0.3
Residue	% vol	1.3	1.2	1.4	1.3

(a) IP 368/84

(b) IP 391/90

TABLE 2

LIGHT-DUTY DIESEL VEHICLES - MEAN CARBON MONOXIDE DATA (g/TEST)
A. ECE 15 CYCLE

Fuel	Sulphur Content wt %	Vehicle No. 1	Vehicle No. 2	Vehicle No. 3	Vehicle No. 4	Vehicle No. 5	Vehicle No. 6	Vehicle No. 7	Vehicle No. 8	Vehicle No. 9	% Change*
1	0.055	3.63	3.48	8.53	5.67	5.96	5.42	9.04	6.66	8.12	-0.7
2	0.12	3.91	3.55	7.66	5.77	6.11	5.37	8.99	7.11	8.05	-0.7
3	0.22	3.60	3.55	8.49	5.31	6.06	5.61	9.31	6.76	8.45	+0.4
4	0.31	3.76	3.51	8.55	5.51	6.26	5.35	8.99	6.74	8.24	0

B. EUDC "HIGH SPEED" CYCLE

1	0.055	1.99	1.93	3.45	1.82	2.24	2.62	6.81	2.45	3.17	-1.3
2	0.12	2.01	1.78	3.24	2.08	2.39	2.69	6.63	2.62	3.24	-0.5
3	0.22	2.05	2.04	3.33	1.89	2.33	2.79	7.10	2.51	3.48	+2.6
4	0.31	1.98	1.92	3.33	1.89	2.33	2.73	6.70	2.67	3.27	0

* Expressed as change in mass emitted as percentage of total from all cars, related to fuel 4 (0.31% S) as base/reference fuel

TABLE 3
 LIGHT-DUTY DIESEL VEHICLES - MEAN HYDROCARBON DATA (g/TEST)
 A. ECE 15 CYCLE

Fuel	Sulphur Content wt %	Vehicle No. 1	Vehicle No. 2	Vehicle No. 3	Vehicle No. 4	Vehicle No. 5	Vehicle No. 6	Vehicle No. 7	Vehicle No. 8	Vehicle No. 9	% Change*
1	0.055	1.25	1.50	1.20	1.41	2.09	0.78	4.63	2.20	4.16	-4.4
2	0.12	1.24	1.67	1.95	1.51	2.19	0.82	4.75	2.31	4.40	+3.6
3	0.22	1.15	1.70	1.01	1.37	2.27	0.82	4.91	2.40	4.31	-0.8
4	0.31	1.25	1.57	1.40	1.62	2.04	0.80	4.71	2.46	4.26	0

B. EUDC "HIGH SPEED" CYCLE

10	0.055	0.62	0.79	0.47	0.50	0.79	0.44	3.71	1.36	2.43	-8.3
2	0.12	0.59	0.83	0.76	0.62	0.76	0.48	3.80	1.38	2.83	-0.5
3	0.22	0.60	0.86	0.36	0.58	0.95	0.53	3.87	1.42	2.54	-3.3
4	0.31	0.61	0.84	0.56	0.67	0.72	0.46	3.91	1.52	2.82	0

* Expressed as change in mass emitted as percentage of total from all cars, related to fuel 4 (0.31% S) as base/reference fuel

TABLE 4

LIGHT-DUTY DIESEL VEHICLES - MEAN NITROGEN OXIDES DATA (g/TEST)
A. ECE 15 CYCLE

Fuel Sulphur Content wt %	Vehicle No. 1	Vehicle No. 2	Vehicle No. 3	Vehicle No. 4	Vehicle No. 5	Vehicle No. 6	Vehicle No. 7	Vehicle No. 8	Vehicle No. 9	% Change*
1 0.055	3.38	2.37	8.41	3.43	3.59	2.26	6.87	13.9	6.01	-2.4
2 0.12	3.54	2.47	11.49	3.82	3.61	2.22	6.73	13.7	6.21	+4.6
3 0.22	3.28	2.54	8.85	3.61	3.56	2.25	6.84	14.6	6.07	+0.3
4 0.31	3.24	2.42	9.37	3.25	3.66	2.20	6.89	14.4	6.00	0

B. EUDC "HIGH SPEED" CYCLE

1 0.055	3.89	3.06	6.45	3.92	4.17	3.13	6.90	21.6	7.64	-1.0
2 0.12	4.06	2.91	7.56	4.46	4.27	3.08	7.48	22.2	7.79	+4.0
3 0.22	3.85	3.43	6.73	3.93	4.19	3.13	7.27	22.8	7.77	+2.8
4 0.31	3.82	3.01	6.87	3.71	4.23	3.01	6.85	22.3	7.56	0

* Expressed as change in mass emitted as percentage of total from all cars, related to fuel 4 (0.31% S) as base/reference fuel

TABLE 5
 LIGHT-DUTY DIESEL VEHICLES - MEAN PARTICULATE DATA (g/TEST)
 A. ECE 15 CYCLE

Fuel	Sulphur Content wt %	Vehicle No. 1	Vehicle No. 2	Vehicle No. 3	Vehicle No. 4	Vehicle No. 5	Vehicle No. 6	Vehicle No. 7	Vehicle No. 8	Vehicle No. 9	% Change*
1	0.055	0.286	1.17	1.87	0.810	0.635	0.698	2.25	0.974	0.813	-7.7
2	0.12	0.350	1.26	1.80	0.870	0.652	0.733	2.53	1.15	0.910	-0.4
3	0.22	0.342	1.25	1.84	0.777	0.650	0.784	2.44	1.06	0.838	-3.1
4	0.31	0.344	1.23	1.89	0.978	0.624	0.689	2.49	1.19	0.862	0

B. EUDC "HIGH SPEED" CYCLE

1	0.055	0.486	0.727	1.23	0.712	0.580	0.860	3.92	2.54	1.14	-7.1
2	0.12	0.547	0.758	0.76	0.886	0.578	0.841	4.00	2.56	1.23	-7.4
3	0.22	0.576	0.787	1.16	0.844	0.630	0.929	4.02	2.84	0.973	-2.8
4	0.31	0.567	0.762	1.20	0.826	0.588	0.966	4.21	2.83	1.18	0

* Expressed as change in mass emitted as percentage of total from all cars, related to fuel 4 (0.31% S) as base/reference fuel

TABLE 6

LIGHT-DUTY DIESEL VEHICLES - MEAN CARBON MONOXIDE DATA (g/km)
COMBINED ECE 15/EUDC CYCLE

Fuel Sulphur Content wt %	Vehicle No. 1	Vehicle No. 2	Vehicle No. 3	Vehicle No. 4	Vehicle No. 5	Vehicle No. 6	Vehicle No. 7	Vehicle No. 8	Vehicle No. 9	% Change*
1	0.055	0.51	1.09	0.68	0.75	0.73	1.44	0.83	1.03	-0.9
2	0.12	0.54	0.99	0.71	0.77	0.73	1.42	0.88	1.03	-0.6
3	0.22	0.51	1.07	0.65	0.76	0.76	1.49	0.84	1.08	+1.1
4	0.31	0.52	1.08	0.67	0.78	0.73	1.43	0.86	1.05	0

* Expressed as change in mass emitted as percentage of total from all cars, related to fuel 4 (0.31% S) as base/reference fuel

TABLE 7
 LIGHT-DUTY DIESEL VEHICLES - MEAN HYDROCARBON DATA (g/km)
 COMBINED ECE 15/EUDC CYCLE

Fuel	Sulphur Content wt %	Vehicle No. 1	Vehicle No. 2	Vehicle No. 3	Vehicle No. 4	Vehicle No. 5	Vehicle No. 6	Vehicle No. 7	Vehicle No. 8	Vehicle No. 9	% Change*
1	0.055	0.17	0.21	0.15	0.17	0.26	0.11	0.76	0.32	0.60	-5.9
2	0.12	0.17	0.23	0.25	0.19	0.27	0.12	0.78	0.34	0.66	+2.1
3	0.22	0.16	0.23	0.12	0.18	0.29	0.12	0.80	0.35	0.62	-1.8
4	0.31	0.17	0.22	0.18	0.21	0.25	0.11	0.78	0.36	0.64	0

* Expressed as change in mass emitted as percentage of total from all cars, related to fuel 4 (0.31% S) as base/reference fuel

TABLE 8

LIGHT-DUTY DIESEL VEHICLES - MEAN NITROGEN OXIDES DATA (g/km)
COMBINED ECE 15/EUDC CYCLE

Fuel	Sulphur Content wt %	Vehicle No. 1	Vehicle No. 2	Vehicle No. 3	Vehicle No. 4	Vehicle No. 5	Vehicle No. 6	Vehicle No. 7	Vehicle No. 8	Vehicle No. 9	% Change*
1	0.055	0.66	0.49	1.35	0.67	0.71	0.49	1.25	3.23	1.24	-1.6
2	0.12	0.69	0.49	1.73	0.75	0.72	0.48	1.29	3.26	1.27	+4.3
3	0.22	0.65	0.54	1.42	0.69	0.70	0.49	1.28	3.40	1.26	+1.7
4	0.31	0.64	0.49	1.48	0.63	0.72	0.47	1.25	3.34	1.23	0

* Expressed as change in mass emitted as percentage of total from all cars, related to fuel 4 (0.31% S) as base/reference fuel

TABLE 9
 LIGHT-DUTY DIESEL VEHICLES - MEAN PARTICULATE DATA (g/km)
 COMBINED ECE 15/EUDC CYCLE

Fuel Sulphur Content wt %	Vehicle No. 1	Vehicle No. 2	Vehicle No. 3	Vehicle No. 4	Vehicle No. 5	Vehicle No. 6	Vehicle No. 7	Vehicle No. 8	Vehicle No. 9	% Change*
1 0.055	0.070	0.172	0.282	0.138	0.110	0.142	0.561	0.319	0.178	-7.4
2 0.12	0.082	0.183	0.233	0.160	0.112	0.143	0.594	0.337	0.195	-4.3
3 0.22	0.083	0.185	0.273	0.147	0.116	0.156	0.587	0.355	0.165	-2.9
4 0.31	0.083	0.181	0.281	0.164	0.110	0.150	0.609	0.365	0.186	0

* Expressed as change in mass emitted as percentage of total from all cars, related to fuel 4 (0.31% S) as base/reference fuel

TABLE 10

HEAVY-DUTY DIESEL ENGINES - MEAN CARBON MONOXIDE DATA (g/kWh)
ECE 49 13 - MODE

Fuel	Sulphur Content wt %	Engine No. 1	Engine No. 2	Engine No. 3	Engine No. 4	% Change *
1	0.055	4.65	1.75	1.24	2.85 +	- 5.4
2	0.12	5.21	1.73	1.28	2.83 +	- 0.4
3	0.22	5.86	1.98	1.42	3.05 +	+11.0
4	0.31	4.88	1.69	1.37	3.15	0

* Expressed as change in mass emitted as percentage of total from all engines, related to fuel 4 (0.31%S) as base/reference fuel

+ Single result only

TABLE 11

HEAVY-DUTY DIESEL ENGINES - MEAN HYDROCARBON DATA (g/kWh)
ECE 49 13 - MODE

Fuel	Sulphur Content wt %	Engine No. 1	Engine No. 2	Engine No. 3	Engine No. 4	% Change *
1	0.055	1.29	0.24	0.32	0.068 +	+6.7
2	0.12	1.24	0.20	0.28	0.062 +	-0.9
3	0.22	1.18	0.22	0.28	0.079 +	-2.2
4	0.31	1.25	0.18	0.29	0.078	0

* Expressed as change in mass emitted as percentage of total from all engines, related to fuel 4 (0.31%S) as base/reference fuel

+ Single result only

TABLE 12
 HEAVY-DUTY DIESEL ENGINES - MEAN NITROGEN OXIDES DATA (g/kWh)
 ECE 49 13 - MODE

Fuel	Sulphur Content wt %	Engine No. 1	Engine No. 2	Engine No. 3	Engine No. 4	% Change *
1	0.055	13.70	10.20	7.33	7.85 +	-3.9
2	0.12	13.52	10.93	8.71	8.29 +	+1.9
3	0.22	12.67	10.12	6.78	7.78 +	-8.1
4	0.31	13.89	10.34	8.11	8.32	0

* Expressed as change in mass emitted as percentage of total from all engines, related to fuel 4 (0.31%) as base/reference fuel

+ Single result only

TABLE 13
 HEAVY-DUTY DIESEL ENGINES - MEAN PARTICULATE DATA (g/kWh)
 ECE 49 13 - MODE

Fuel	Sulphur Content wt %	Engine No. 1	Engine No. 2	Engine No. 3	Engine No. 4	% Change *
1	0.055	0.557	0.442	0.275	0.232 +	-0.1
2	0.12	0.571	0.455	0.335	0.181 +	+2.3
3	0.22	0.674	0.473	0.295	0.201 +	+9.0
4	0.31	0.508	0.493	0.269	0.238	0

* Expressed as change in mass emitted as percentage of total from all engines, related to fuel 4 (0.31%) as base/reference fuel

+ Single result only

TABLE 14

STATISTICAL DATA, LIGHT-DUTY VEHICLES, COMBINED CYCLE

COMPARISON OF FUELS	F-RATIO PROBABILITY %	DIFFERENCE	T-RATIO	T-RATIO PROBABILITY %
CO	4 vs 3	0.012	1.293	>20
	4 vs 2	0.006	0.649	50
	4 vs 1	0.008	0.807	>20
HC	4 vs 3	0.015	1.102	>20
	4 vs 2	-0.066	-1.373	>10
	4 vs 1	0.019	2.702	2 *
NO _x	4 vs 3	0.067	3.069	1 *
	4 vs 2	-0.048	-2.936	1 *
	4 vs 1	0.018	1.115	>20
PARTICULATE	4 vs 3	0.027	6.791	0.1 *
	4 vs 2	0.010	3.055	1 *
	4 vs 1	0.018	5.142	0.1 *

Statistically significant differences are shown with an asterisk

TABLE 15

STATISTICAL DATA, HEAVY-DUTY ENGINES, ECE 49 CYCLE

COMPARISON OF FUELS	F-RATIO PROBABILITY %	DIFFERENCE	T-RATIO	T-RATIO PROBABILITY %
CO	4 vs 3	-0.469	-4.479	2 *
	4 vs 2	-0.138	-1.859	5 *
	4 vs 1	0.025	0.427	>50
HC	4 vs 3	-0.060	-4.644	0.2 *
	4 vs 2	-0.066	-5.563	0.1 *
	4 vs 1	-0.105	-7.191	0.1 *
NO _x	4 vs 3	0.374	1.174	>20
	4 vs 2	-0.723	-1.730	>10
	4 vs 1	-0.109	-0.338	>50
PARTICULATE	4 vs 3	-0.076	-5.415	0.1 *
	4 vs 2	-0.050	-3.539	1 *
	4 vs 1	-0.032	-2.207	>5

Statistically significant differences are shown with an asterisk

TABLE 16

LIGHT-DUTY DIESEL VEHICLES - SULPHATE ON PARTICULATE & wt
COMBINED ECE15/EUDC CYCLE

Fuel	Sulphur Content wt %	Vehicle No. 1	Vehicle No. 2	Vehicle No. 3	Vehicle No. 4	Vehicle No. 5	Vehicle No. 6	Vehicle No. 7	Vehicle No. 8	Vehicle No. 9	Mean Value
1	0.055	9.27	1.31	1.82	2.93	3.24	5.13	0.99	*	2.78	3.43
2	0.12	5.24	1.78	1.52	2.30	4.63	5.55	0.73	*	2.96	3.09
3	0.22	11.94	1.97	1.43	4.86	7.05	7.05	0.94	*	3.23	4.81
4	0.31	9.80	3.59	3.31	5.11	6.94	6.71	1.94	*	4.30	5.21

* Not available

TABLE 17
 HEAVY-DUTY DIESEL ENGINES, SULPHATE ON PARTICULATE, % wt
 ECE 49 CYCLE

Fuel	Sulphur Content wt %	Engine No. 1	Engine No. 2	Engine No. 3	Engine No. 4	Mean Value
1	0.055	1.79	1.92	2.81	*	2.17
2	0.12	1.55	2.80	7.06	*	3.80
3	0.22	1.59	4.30	6.32	*	4.07
4	0.31	1.71	6.73	7.52	*	5.32

* Not available

TABLE 18

LIGHT-DUTY DIESEL VEHICLES - TRAPPED SULPHATE AS PERCENTAGE OF SULPHUR CONSUMED,
COMBINED ECE15/EUDC CYCLE

Fuel	Sulphur Content wt %	Vehicle No. 1	Vehicle No. 2	Vehicle No. 3	Vehicle No. 4	Vehicle No. 5	Vehicle No. 6	Vehicle No. 7	Vehicle No. 8	Vehicle No. 9	Mean Value
1	0.055	21.0	6.2	8.4	10.2	9.1	16.7	11.1	*	17.2	12.5
2	0.12	7.3	4.5	3.5	4.6	6.5	8.8	4.2	*	10.1	6.2
3	0.22	8.1	2.7	1.9	4.6	6.0	6.4	2.9	*	5.5	4.8
4	0.31	5.1	3.4	3.1	4.0	3.8	4.8	4.5	*	5.4	4.3

* Not available

TABLE 19
HEAVY-DUTY DIESEL ENGINES; TRAPPED SULPHATE AS PERCENTAGE OF SULPHUR CONSUMED,
ECE 49 CYCLE

Fuel	Sulphur Content wt %	Engine No. 1	Engine No. 2	Engine No. 3	Engine No. 4	Mean Value
1	0.055	6.05	6.18	4.82	*	5.68
2	0.12	2.38	4.65	8.00	*	5.01
3	0.22	1.97	4.01	2.96	*	2.98
4	0.31	1.13	4.66	2.66	*	2.82

* Not available

TABLE 20

VROM PROGRAMME
 HEAVY-DUTY DIESEL ENGINES - PARTICULATE DATA
 ECE 49 13-MODE (g/kWh)

Fuel	Sulphur Content wt %	Engine V.1	Engine V.2	Engine V.3	Engine V.4	Engine V.5	Engine V.6	Engine V.7	% Change *
E	0.07	0.326	0.484	0.374	0.288	0.251	0.553	0.153	-12.4
H	0.29	0.368	0.637	0.470	0.320	0.279	0.514	0.184	0

(Engines have been coded V.1 to V.7 to distinguish them from engines 1 to 4 used in the CONCAWE programme)

* Expressed as change in mass emitted as percentage of total from all engines, related to Fuel H (0.29%S) as base/reference fuel