

Experimental assessment of particulate measurement instrumentation

SP Bell
CJ Dickens
EA Feest
AP Smith
AM Tope

January 1999

Experimental assessment of particulate measurement instrumentation

SP Bell
CJ Dickens
EA Feest
AP Smith
AM Tope

January 1999

Title	Experimental assessment of particulate measurement instrumentation
Customer	CEC DGXI
Customer reference	B4-3040/97/000500/MAR/D3
Confidentiality, copyright and reproduction	<p>Restricted - Commercial</p> <p>This document has been prepared by AEA Technology plc in connection with a contract to supply goods and/or services and is submitted only on the basis of strict confidentiality. The contents must not be disclosed to third parties other than in accordance with the terms of the contract.</p>
File reference	mainrep3.doc
Report number	AEAT-4978
Report status	Issue 1

AEA Technology
Allan Smith
551 Harwell Laboratory
Didcot
Oxfordshire
OX11 0RA
Telephone +44 (0)1235 434115
Facsimile +44 (0)1235 432404

AEA Technology is the trading name of AEA Technology plc
AEA Technology is certificated to BS EN ISO9001:(1994)

	Name	Signature	Date
Author	SP Bell CJ Dickens EA Feest AP Smith AM Tope		

Reviewed by	EA Feest		
--------------------	----------	--	--

Approved by	AR McCabe		
--------------------	-----------	--	--

Executive Summary

Recent investigations into the impact of air pollution on human health have identified the smaller sized particulates which are respirable to be of particular concern in relation to their effect on human health. However, the measuring procedure for particulate emissions, as defined in current legislation, does not explicitly aim at the collection of particulates within a defined size range.

As more information from particulate studies has emerged, the possibility that vehicle particulate emissions will need to be regulated with regard to both particle size and number, in addition to particle mass, has become evident. DGXI of the CEC have therefore commissioned this study to investigate how such measurements might be made with respect to future type approval tests.

This work comprised two phases:

Phase 1: Literature review to help formulate the recommendations for measuring techniques to be investigated in Phase 2.

Phase 2: Experimental assessment of instrument performance.

The main objective was to provide proposals for changes in the current measuring procedure for particulate emissions, if appropriate.

The evidence from the Phase 1 literature review suggests that there is a link between particulate exposure and health, and that the ultra-fine component of ambient particulate may be most significant. It seems likely that the next few years will see an increase in the quantity and quality of health effects data gathered. Review activities suggest, therefore, that there is unlikely to be a basis for new particle metric standards before 2002 with implementation by 2005. However, significant health-related and congestion costs have been attributed to road transport in urban areas and public and political opinion may require more rapid action on ultrafine particulate abatement than can be justified on potential cost savings alone. In this scenario, the use of markers for particles, which usefully discriminate automotive sources, could be accelerated, particularly for urban locations. In view of the uncertainty over causal links it is not known whether number, mass or both are the most appropriate particulate markers.

The Phase 1 review led to the recommendation that particle emissions be classified as <300nm and >300nm for this study. 300nm was chosen due to its significance in lung deposition; particles above 300nm tend to be deposited in the upper airways by impaction, whereas particles below 300nm tend to reach the alveoli region of the lung where they are deposited due to diffusion. Separating particles about 300nm therefore roughly separates particles by their deposition site in the lungs. It was recognised that future health data may require this cut-off point to be changed. The Scanning Mobility Particle Sizer (SMPS) was

recommended as a baseline technique with which to compare the simpler condensation nucleus counters (CNCs) for measuring particle numbers.

Phase 2 consisted of an experimental programme to assess particulate measurement techniques and methods recommended from Phase 1. Regulatory emissions, temperatures, and smoke were also measured.

Initial work (Phase 2a) focused on a latest technology diesel DI VW Golf. Sampling methodology, driving conditions and reproducibility were investigated in detail. The particle measurements were made with a wide range of dilutions and from raw exhaust. High temperature sampling was also investigated. For each of these sampling schemes, hot and cold start steady state cycles, hot and cold start NEDC (New European Drive Cycle) and FAS (Free Acceleration Smoke) tests were performed. Multiple runs gave a good indication of the reproducibility of the measurements.

It was found that sampling method had an insignificant effect on the particle measurements, so the use of standard CVS conditions was recommended. It was difficult to make clear recommendations regarding the different drive cycles by studying just one vehicle, so it was recommended that most of the cycles be studied for the next stage of the work (Phase 2b). The CNC data agreed well with the SMPS measurements for most conditions. However, at high speed (~120kph) the CNC gave many more particles than SMPS. These appear to be very small particles below the range of the SMPS (<7nm). There are no significant numbers of particles above 300nm, so it was recommended that the CNC be used without a size cut-point. The use of an Andersen impactor showed that there is significant mass of particles both above and below 300nm. However, the impactor was unable to sample sufficient volume to provide reliable mass data from diluted samples and only gave reproducible results when sampling from the raw exhaust.

Phase 2b consisted of applying the recommended techniques from Phase 2a to 8 diesel vehicles, old and new, covering DI and IDI technologies. The CNC and SMPS data were again in good agreement for all vehicles except under high speed conditions. Interestingly, there were two vehicles which did not show the high CNC data at high speed. Reproducibility was good and the CNC appears to be a valid method for determining particle number emissions. Further, there should not be any significant problems in its use for alternative technologies such as gasoline, LPG, etc. The Andersen impactor gave poor results due to the small mass samples collected. Improvements in impactor design and sampling volumes are necessary to make this a practicable technique for diesels. There was no obvious reason for changing the test cycle from the cold start NEDC. Other cycles related well to each other apart from the FAS tests which do not appear to correlate with more realistic driving conditions. Older technology vehicles tended to give higher emissions of smoke, CO and THC, but particulate mass from the NEDC did not seem to relate to technology. Particle numbers

were very similar for the NEDC for all vehicles studied. In other words, improvements in particulate mass emissions do not necessarily lead to particle number reductions.

Based on the work in this report the following preliminary recommendations are made as a route for incorporating size-related particulate measurements into type approval protocols.

1. Until the health position is clarified further, measure particulate in terms of mass and number.
2. As the number of particles above 300nm was found to be insignificant, total particle flux is a convenient indicator of ultra-fine (e.g. <300nm) particle flux.
3. A CNC with diluter is an appropriate technique to measure total particle number flux.
4. It is appropriate for diesel vehicles to split the particulate mass at 300nm.
5. To achieve a split in mass range an impactor can be used, though improved design is necessary.
6. Standard CVS sampling conditions can be used.
7. There is no reason to change from using the standard cold start NEDC.

However, the restricted number of experiments in this work means that the following experiments are needed to clarify certain points before any implementation of the above recommendations:

1. A more comprehensive study of the diesel vehicle parc to provide greater statistical significance.
2. Further investigation into the source of the apparent large increase in small particles for most vehicles at high speed.
3. Improved impactor measurements for size segregated mass measurements.
4. The application of the measurement methods to alternative engine and fuel technologies (such as gasoline, LPG, etc.).

Contents

1. INTRODUCTION	1
2. SUMMARY OF PHASE 1 REPORT	2
3. PHASE 2A - ASSESSMENT OF DIFFERENT TECHNIQUES AND PROCEDURES	4
3.1 EXPERIMENTAL	4
3.1.1 Test vehicle	4
3.1.2 Test fuel	4
3.1.3 Lubricating oil	4
3.1.4 Instrumentation	4
3.1.5 Sampling	5
3.1.6 Test conditions	6
3.1.6.1 Hot start steady-state road load.	6
3.1.6.2 Cold start steady-state road load	7
3.1.6.3 Free Acceleration Smoke tests	7
3.1.6.4 Cold start combined ECE plus EU drive cycle (NEDC)	7
3.1.6.5 Hot start combined ECE plus EU drive cycle (NEDC)	7
3.1.7 Conditioning	7
3.1.8 Test protocol	8
3.2 RESULTS	9
3.2.1 Regulatory emissions	9
3.2.1.1 Hot Steady state tests	9
3.2.1.2 Cold steady state tests	10
3.2.1.3 FAS tests	11
3.2.1.4 Hot start NEDC drive cycles	11
3.2.1.5 Cold start NEDC drive cycles	12
3.2.1.6 Conclusion	13
3.2.2 Particle sampling data	13
3.2.2.1 Steady state	13
3.2.2.2 Cold start steady state tests	15
3.2.2.3 FAS tests	16
3.2.2.4 NEDC drive cycles	16
3.2.2.5 Discussion	16
3.3 SUMMARY	18
3.4 RECOMMENDATIONS FOR PHASE 2B	19
4. PHASE 2B - APPLICATION TO VEHICLE PARC	20
4.1 EXPERIMENTAL	20
4.1.1 Selection of test vehicles	20
4.1.2 Instrumentation and sampling	21
4.1.3 Test conditions and protocol	22

4.2 RESULTS	22
4.2.1 CNC particle sampling effects	22
4.2.2 Steady state tests	23
4.2.3 FAS tests	24
4.2.4 New European Drive Cycle (NEDC)	25
4.2.5 Cycle correlations	28
4.3 DISCUSSION	28
4.3.1 Particle flux measurements	28
4.3.2 Particle mass measurements	28
4.3.3 Sampling considerations	29
4.3.4 Vehicle comparisons	29
4.4 SUMMARY OF PHASE 2B	30
4.4.1 Instrumentation	31
4.4.2 Vehicle and test effects	31
5. CONCLUSIONS	31
6. RECOMMENDATIONS	33
7. ACKNOWLEDGEMENTS	33

Appendix 1 - Description of particle measurement techniques

1. Introduction

Particulate emissions from motor vehicles equipped with diesel engines are of current concern from an air quality perspective. This kind of pollutant was initially regulated for light-duty vehicles through Directive 88/436/EEC. In the meantime the emission standards which have to be met by new vehicle types have been tightened.

Recent investigations into the impact of air pollution on human health have identified the smaller sized particulates which are respirable to be of particular concern in relation to their effect on human health. However, the measuring procedure for particulate emissions, as defined in current legislation, does not explicitly aim at the collection of particulates within a defined size range.

As more information from particulate studies has emerged, the possibility that vehicle particulate emissions will need to be regulated with regard to both particle size and number in addition to particle mass has also become evident. DGXI of the CEC have therefore commissioned a study to investigate how such measurements might be made to comply with future legislation.

This study comprises two phases:

Phase 1: Review of international literature, collection of data, proposition of measuring procedures, reporting of recommendations to the Commission. Particular emphasis was to be placed on information relating to health effects.

Phase 2: Experimental assessment of instrument performance.

The main objective is to provide proposals for changes of the current measuring procedure of particulate emission, if appropriate.

The results of Phase 1 have already been reported ("Review of international literature and recommendations for experimental programme", S P Bell, J J McAuchey, C J Dickens and D C W Blaikley, AEAT-3241, 1998). The main conclusions of that phase are summarised in the following section. Following on from that report an experimental work programme was agreed which comprised two main work packages.

The first package would be an in depth study on one vehicle (latest available technology) which would investigate:

- candidate instruments, e.g. current and in development;
- sampling regimes, e.g. raw (tailpipe/mini-diluter) through to high dilution ratios (CVS plus additional dilution system) and
- test cycle protocols, e.g. steady-state, transient and free acceleration

with the objective of determining correlations between raw, normal and high dilution ratio sampling systems for current and future particle size and concentration instrumentation over cycles representative of type approval conditions.

That work package would then provide the basis for the development of appropriate test protocols to be assessed on a larger range of vehicles in the second work package of the experimental programme. For this the vehicles would be selected to provide a comparison between direct and indirect injection engines incorporating changes in technology designed to meet different stages of technical legislation.

In addition to summarising Phase 1, the present report also describes in detail the first stage of the experimental programme (Phase 2a) and the application of its measurement procedures to a wider range of vehicles (Phase 2b). It also presents the conclusions and recommendations from the whole project.

2. Summary of Phase 1 report

Phase 1 involved:

- review of international literature, with particular emphasis on health effects
- collection of existing data
- proposition of measuring procedures
- reporting of recommendations for the experimental phase to the Commission.

Key findings of this study were as follows.

Concerning the relationship between health effects and exposure to inhalable particles, including those attributable to automotive particulate emissions, there is a consistency of epidemiological and other evidence suggesting that, in public health terms, methods of reducing particulate exposure should be encouraged. However, there are insufficient exposure and dosimetry data available to currently apportion the degree of risk from various sources. The evidence relating to mechanisms has advanced significantly in the last 2-3 years and indicates that the ultra-fine component of ambient particulate may be most significant, irrespective of chemical composition. Thus, as new epidemiological studies using different particle metrics report, greater certainty as to optimal areas for particulate control will emerge

Thus, it seems likely that the period to 2000/2001 will focus on the gathering of health effects data with respect to better defined, apportioned, population exposure data. On this timescale, review activities suggest there is unlikely to be a basis for new particle metric standards (e.g. surface area, number) before 2002 with implementation by 2005. Significant health-related and congestion costs have been attributed to road transport in urban areas and public and political opinion may require more rapid action on ultrafine particulate abatement than can be justified on potential cost savings alone. In this scenario, the use of markers such as particle number, which usefully discriminate automotive sources, could be accelerated, particularly for urban locations.

Concerning the choice of the particle size range/ranges to measure, these should ideally relate to the health effects of different sized particles. Additionally, consideration should be given to the ability to easily discriminate between chosen size ranges, and to the availability or future availability of suitable particle sizing instruments.

The most widely used mass based size range is known as PM_{10} (weight of particulate mass below $10\mu m$ aerodynamic diameter). Recently in the U.S., $PM_{2.5}$ has also been promulgated as a standard (weight of particulate mass below $2.5\mu m$). PM_{10} has been widely used for environmental monitoring, so it could be argued as appropriate to use this as a measure of particle emissions from vehicles. $PM_{2.5}$ has been less widely used so far, and may be superseded in future with newer standards, such as PM_1 (mass of particulate below $1\mu m$). The equipment required to measure PM_{10} , $PM_{2.5}$ or PM_1 can be relatively simple. In principle, all that is required is an impactor set to the required size, and a filter to catch the remaining particulate. It would be relatively easy, therefore, to alter the size cut-point of such measurements if required.

Available data shows that the majority of particles (by number) in vehicle emissions lie near $0.1\mu m$ ($100nm$), with no significant number of particles above $1\mu m$. It would therefore be appropriate to effectively concentrate on ultra-fine particles for number based measurements. Because there are as yet no hard medical facts supporting a cut-off of $100nm$, it was recommended that for the present experimental study number based particle emissions are classified as $<300nm$ and $>300nm$. $300nm$ was chosen partly due to its significance in lung deposition; particles above $300nm$ tend to be deposited in the upper airways by impaction, whereas particles below $300nm$ tend to reach the alveoli region of the lung where they are deposited due to diffusion. Separating particles about $300nm$ therefore roughly separates particles by their deposition site in the lungs. With respect to the future emergence of data relating to health effects it is recommended that the development of test protocols should recognise that this cut-off point may need to be changed.

Concerning the choice of instrumentation, it was concluded that the Scanning Mobility Particle Sizer (SMPS) is currently the most appropriate instrument for

the baseline measurement of the size distribution of particulate emissions from vehicles. (See Appendix 1 for a description of the relevant particle measurement techniques). However, a number of factors suggest that this instrument is not necessarily appropriate for application to future type approval test protocols. Its applicability to the measurement of particle size and concentration under steady-state conditions is well documented as being repeatable and reproducible. However, under transient conditions, whilst providing highly sensitive and time resolved information at a specific particle size band, it requires multiple drive cycles to be driven to provide an indication of particle concentration over the full spectrum of sizes of interest. Also, it is fairly expensive to purchase, and requires skill in the operation and interpretation of the data. It is therefore desirable that something far less sophisticated be developed for future regulatory testing. For these tests it is envisaged that such a device needs only to separate particles into preferably two size ranges, and then count the total number of particles in each range.

The recommendation for the experimental programme was therefore to use two simple condensation nucleus counters (CNCs), one with a pre-impactor set for 300nm (0.3µm). This would give a particle number concentration below 300nm and the total number concentration over all sizes. From this the number concentration above 300nm could easily be deduced. Some pre-dilution might be needed with these devices. It was also recommended that the SMPS be run in parallel to these devices to check their performance against the currently accepted industry standard and to explore whether a single size cut-off is appropriate or if two sizes provide a more robust measure.

It was proposed that the Phase 2 experimental programme would comprise two work packages as summarised in the Introduction section of the present report.

3. Phase 2a - assessment of different techniques and procedures

3.1 EXPERIMENTAL

3.1.1 Test vehicle

A Volkswagen Golf fitted with a 1.9 turbo-diesel direct injection engine was procured for this phase of the programme. This vehicle meets Euro 2 emissions limits (94/12/EC) and is representative of a relatively large part of the vehicle parc. The vehicle was selected anonymously and had covered ~6400km.

3.1.2 Test fuel

The test fuel for all tests was procured to a specification appropriate to year 2000. See appendix 3 in the Phase 1 literature review. It should be appreciated that this diesel fuel had a sulphur content of 300mg/km, which the vehicles were not optimised for.

3.1.3 Lubricating oil

Due to the influence oil formulation can have on particulate emissions from diesel engines it was recommended that an appropriate baseline reference oil be used in all vehicles. Whilst it could be argued that by 2005 synthetic oils may dominate in the vehicle parc, at this stage it was felt that a mineral reference oil would be more appropriate as more is known about the performance of these oils in relation to particulate formation. Therefore the oil used was to specification RL 189. This oil is a 15W40 mineral oil with a G4PD2 rating. Advice was taken on this matter from oil industry experts.

Lubricating oil and filter were changed prior to test.

3.1.4 Instrumentation

Particle number measurements were taken with a Scanning Mobility Particle Sizer (SMPS) and a Condensation Nucleus Counter (CNC) (see Appendix 1 for details). Because of the high particle concentration levels expected to be found in the dilution tunnel, the CNC sampled through a diluter, allowing dilutions of about 2000:1. The sample taken by the diluter was either direct from the sampling column, or passed through the lower stage of an Andersen impactor. This allowed the CNC to measure either all particles, or particles less than 300nm (aerodynamic diameter). It should be remembered that the SMPS measures particle size in terms of mobility diameter, which can differ from aerodynamic diameter depending on particle density and shape.

Particle mass measurements were collected with the lower stage of an Andersen impactor, giving particulate mass above and below 300nm (aerodynamic particle diameter).

The vehicle was fitted with an oxidation catalyst, so the temperatures before and after the catalyst were recorded.

Total particulate mass through the CVS was determined using filter papers. The smoke levels determined by a Celesco smokemeter (which is a fast response obscuration measurement device) were also logged, in addition to road speed, and modal determinations of CO₂, CO, NO_x and THC. The oil, pre- and post-catalyst, and Celesco temperatures were all logged onto a PC as a function of time. Also,

the differential pressure output from a pitot tube placed in the exhaust was logged in order to determine exhaust flow.

CVS gases were always logged modally. In addition, for steady state data and a selection of transient data, bag samples were collected and analysed.

3.1.5 Sampling

It is important to get the best sampling conditions for all proposed instruments. Before vehicle test work started, the sampling arrangements were considered to minimise sampling losses, and allow simultaneous isokinetic sampling from multiple instruments, for the particle measurement devices.

The sample system was designed to take one isokinetic sample from the dilution tunnel, and allow the measuring instruments to simultaneously sample from this. To satisfy these requirements the pipework extends horizontally from the dilution tunnel, with each instrument taking iso-kinetic samples from the pipe. An examination of the volumetric flow rate through the existing pipework (33.8 l min^{-1}), and the combined sample rates of the instruments likely to be used on the facility (0.3 , 20 and 20 l min^{-1} for the SMPS, CNC and Andersen Mk-II cascade impactor respectively) shows that these are of a similar magnitude. This similarity means that for the larger flow rates (CNC and Andersen Mk-II) the sample nozzles will occupy a large proportion of the pipe cross-sectional area (for iso-kinetic sampling), and that once one instrument has removed a sample of aerosol there will be relatively little particulate remaining for the other instruments to acquire a statistically representative sample. It was therefore decided that the Andersen would acquire its sample downstream of the sample points for the SMPS and CNC instruments.

The particle loss in the column is dominated by diffusion affecting the smaller particles. The ASTEC computer code gives a 98.4% transport efficiency for $0.01 \mu\text{m}$ aerodynamic diameter particles in the sampling column. This transport efficiency is very high and well within acceptable limits.

In determining the specifications for the sample lines that transport aerosol from the columns to the analysers, consideration had to be given to the volumetric flow rates at which the instruments operated and the diameter of their inlet ports. It was decided that the diameter of the sample lines from the columns to the instrument should be the same or similar to those of the instrument. Calculations were therefore undertaken using the diameters and flow rates for each analyser on the geometry shown in Figure 1. It was necessary to adjust the diameter of the sampling nozzles in certain cases to ensure isokinetic sampling where necessary. The results of these calculations are given for selected particle diameters in Table 1.

Table 1. Transport efficiencies for instrument sampling lines.

Analyser	Pipe Diameter	Nozzle Diameter Col 1	Transport Efficiency			
			0.01 μm (%)	0.1 μm (%)	1 μm (%)	10 μm (%)
	(mm)	(mm)				
SMPS	5	5	85.2	99.0	99.7	85.4
CNC	6	12	98.5	99.9	100.0	1.61
Anderse n	12	12	98.9	99.9	100.0	96.3

All of the transport efficiencies are high with the exception of the CNC's sampling line which decreases markedly above 5 μm aerodynamic diameter. However, in every other respect the transport efficiencies are very high and within acceptable levels. No one aspect of the entire sampling arrangement can be considered in isolation and the total transport efficiency needs to be considered for each instrument from the point where the particles are removed from the CVS tunnel.

Sampling for particulates included the use of the full flow CVS and dilution tunnel as used for regulatory testing. AEA Technology's CVS employs the PDP (positive displacement pump) principal which provides for up to 30:1 dilution ratio varying from 0 to 100% in 1% increments. Two settings, 65% and 90%, were used in the steady state tests. For high dilution testing, the SMPS sampled through an additional diluter, running at a dilution ratio of about 2000:1.

There is emerging data which suggests that the temperature of the sample line may have an effect on particulate size (e.g. higher temperatures may affect volatile components adsorbed onto particulate surface). This was investigated by running the sample line in an oven at elevated temperature (600°C) with the CVS operating at 65%.

Raw (tailpipe) sampling was also investigated, to determine whether dilution tunnel effects are significant and to explore this route as a possible in-service method of testing.

3.1.6 Test conditions

The vehicle was tested under the following conditions:

3.1.6.1 Hot start steady-state road load.

A series of hot start steady-state tests were carried out to investigate candidate instrument and sampling conditions at 3 road-load speed sites. AEA Technology's experience in measuring size distributions of particulates in vehicle exhaust indicates that only a limited number of steady-state conditions needed to

be monitored and so size scans were taken at idle, medium and high road load conditions. These are representative of some of the steady-state “plateaux” driven during the ECE 15 and EU legislative drive cycles and are shown in Table 2 below.

Table 2. Steady-state road load sites

Point	1	3	5
Speed kph.	idle	50	120
Gear	neutral	4	5

The vehicle was allowed to equilibrate at each condition before sampling commenced. A “dead man’s foot” was used to maintain the non-idle speeds. Each speed condition was studied twice for each sampling scheme, in the order idle, 50kph, 120kph, idle, 50kph, 120kph.

3.1.6.2 Cold start steady-state road load

In addition to the hot-start tests, we carried out cold start road load tests under different sampling conditions. The size distribution was scanned using the SMPS and the number of particles monitored using the CNC. This gave information on the changes in size distribution and number concentration during engine warm-up. Particulates by filter method and regulated gaseous pollutants were also measured during this test.

A driver (as opposed to a mechanical foot device) had to be used for these tests in order to counteract the effects of the engine warming up, so enabling a constant speed of 50kph to be maintained for ~15 to 20 minutes.

3.1.6.3 Free Acceleration Smoke tests

Free Acceleration Smoke (FAS) test cycles as defined in regulation 72/306/EEC were carried out with the SMPS instrument set at a fixed particle size. A series of 10 FAS tests were conducted for each run. Repeat runs were performed with the SMPS set to different size ranges.

3.1.6.4 Cold start combined ECE plus EU drive cycle (NEDC)

One cold start combined ECE plus EU drive cycle was monitored for each sampling condition. The SMPS instrument was set to a fixed size range of 45nm for each run.

3.1.6.5 Hot start combined ECE plus EU drive cycle (NEDC)

Hot start ECE plus EU drive cycles were performed with the SMPS instrument set at a fixed size range per test. These size ranges were centred on 10nm, 20nm, 45nm, 90nm and 150nm.

3.1.7 Conditioning

Vehicle and test equipment were presented and conditioned in accordance with the EPEFE test protocol for light-duty diesel engine vehicles.

The vehicle was subjected to 500 km road mileage conditioning following oil change to ensure all volatile components were driven off and that the oil was in a stable condition. 200 km of this mileage was carried out on reference test fuel.

Conditioning of the vehicle prior to hot start tests comprised running 3 extra-urban drive cycles and recording final bulk oil temperatures.

The dilution tunnel was conditioned at the start of each day to ensure tunnel background particulate levels were as low as possible. This comprises running the CVS system with the sample tube disconnected from the vehicle tailpipe and with the dilution air inlet restricted for 90% of its area. During this time SMPS scans are carried out until backgrounds become stable. Further conditioning included connecting the vehicle tailpipe and re-running SMPS scans.

All other test conditions complied with the requirements of the NEDC.

3.1.8 Test protocol

Information obtained during the literature survey indicated that a cut-point at 300nm was appropriate when monitoring transient emissions. The dynamic response of the SMPS and the candidate instrument were therefore monitored over the transient drive cycles. The CNC and Andersen impactor were run in parallel with the SMPS giving the opportunity to monitor particulate emissions > 300nm and <300nm under the same test conditions and over the same cycle.

A daily test schedule for transient runs comprised the following:

3 EU cycles for conditioning on the preceeding night
Cold start NEDC with SMPS set at 45nm
2 EU cycles for conditioning
Free acceleration smoke test with SMPS set at 10 nm
Hot start NEDC with SMPS set at 10 nm
Free acceleration smoke test with SMPS set at 20 nm
Hot start NEDC with SMPS set at 20 nm
Free acceleration smoke test with SMPS set at 45 nm
Hot start NEDC with SMPS set at 45 nm
Free acceleration smoke test with SMPS set at 90 nm

Hot start NEDC with SMPS set at 90 nm
Free acceleration smoke test with SMPS set at 150 nm
Hot start NEDC with SMPS set at 150 nm

The schedules were conducted under the following conditions, as a result of an analysis of the steady state data:

Day 1 - 65% CVS dilution;
Day 2 - high dilution;
Day 3 - high furnace temperature;
Day 4 - raw tailpipe.

3.2 RESULTS

3.2.1 Regulatory emissions

The CVS gas concentrations, particulate filter samples, Celesco smoke readings and measured temperatures from the different runs have been compared to evaluate the reproducibility of the vehicle running conditions. This is necessary in order to assess the significance of any differences observed in particulate distributions observed using the various techniques being tested.

3.2.1.1 Hot Steady state tests

Table 3 shows the reproducibility of steady state data taken on three separate days when sampling at CVS 65%, CVS 90% , CVS 65% with high temperature particle line and CVS 65% with high dilution particle sampling. Figure 2 shows that for the 50kph data the regulatory results were unaffected by the CVS operating conditions. This applies to all studied conditions and the results have therefore been treated together in Table 3. As the CVS 90% sampling had no significant effect on the results, it was not used for the transient experiments.

Table 3. Reproducibility of steady state data.

(Values are quoted \pm one standard deviation, with the percentage of the mean that this represents given in brackets.)

	Idle	50kph	120kph
CO ₂ (g/min)	21.3 \pm 0.6 (3%)	59 \pm 3 (5%)	329 \pm 12 (4%)
CO (g/min)	0.055 \pm 0.029 (52%)	0.007 \pm 0.007 (106%)	0.090 \pm 0.013 (14%)
THC (g/min)	0.026 \pm 0.006 (24%)	0.028 \pm 0.006 (22%)	0.033 \pm 0.005 (15%)
NO _x (g/min)	0.212 \pm 0.010 (5%)	0.17 \pm 0.02 (12%)	1.89 \pm 0.09 (5%)
Smoke K (m ⁻¹)	0.0012 \pm 0.0016 (130%)	0.041 \pm 0.009 (22%)	0.075 \pm 0.010 (13%)
Particulate (mg/km)	6.3 \pm 0.9mg/min (14%)	31 \pm 2 (9%)	108 \pm 21 (20%)
Exhaust flow (l/min)	~400	~570 (700)*	~2000 (2200)*
Temperatures			

Oil (°C)	99±2 (2%)	97.7±0.5 (0.5%)	112.3±0.5 (0.4%)
Catalyst In (°C)	132±6 (5%)	199±4 (2%)	415±7 (2%)
Catalyst Out (°C)	131±8 (5%)	194±4 (2%)	402±6 (2%)
Celesco (°C)	82±12 (14%)	93±6 (6%)	242±5 (2%)

* Values in brackets estimated using exhaust gas concentrations and fuel economy - see text.

The levels of CO and THC are very low due to the efficiency of the catalyst, so their reproducibility is very poor. The CO₂ levels are more representative of engine running conditions, and these are reasonably consistent.

The exhaust flow rates determined by pitot tube were quite noisy and approximate average values are quoted. These were compared to values estimated using a Bosch analyser (model ETT008.36, modified for catalyst work to read to 3 decimal places) to determine CO₂ and O₂ concentrations in the exhaust, together with the vehicle's own display of miles per gallon. This allowed the estimation of exhaust flow (indicated by an asterisk in Table 3) to be made. The two agree reasonably well.

The steady state temperatures are well reproduced on each occasion.

The smoke levels at idle are very low and so not very reproducible. This improves at higher speeds where the smoke levels increase. The amount of particulate emitted, as determined by capturing on filter papers, is fairly poorly reproduced. The levels of smoke and particulates are not as repeatable as the CO₂ because they represent the effects of relatively small changes in engine conditions, whereas the CO₂ is dependent on larger scale parameters. However, the reproducibility of smoke and particulates is of most relevance to the assessment of particle sizing and flux methods.

The raw exhaust data has not been included in the table above. For these data the temperatures, flow and smoke measurements were all in agreement with those above. However, the gaseous and particulate measurements are not directly comparable as not all of the exhaust is directed into the CVS by the funnel that is used for this set up. The gas analyses imply that the amount captured changes with engine condition, with ~80 to 90% captured at idle and 50kph, and only ~50 to 60% at 120kph. However, the two runs for each speed condition were in good agreement with each other and, taken together with the agreement in smoke and temperature levels, this suggests that the engine was running in a very similar manner to the previous runs.

The overall degree of reproducibility observed at steady state is influenced by the vehicle's engine management system. This can be seen, for example, in Figure 3, which shows the change in CO₂ concentration relative to the mean as a function of time at a steady state of 50kph. There is clearly a periodic variation in

measured CO₂ levels. The variation is much less periodic at other speeds, and appears to be an inherent engine management feature.

Figure 4 shows the relationship between the smoke measured using the Celesco and the particulate sample collected on the filter paper. There appears to be a linear relationship within a given speed, but not necessarily between different speeds. This may be due to a change in the size and density of particles >300nm that contribute largely to the mass and the Celesco data. The Celesco smokemeter data when expressed in terms of m⁻¹ are proportional to the number of scattering/absorbing centres in the line of the LED light at 550nm. The constant of proportionality is a scattering/absorption efficiency term which will depend on the size and shape of particulates and the absorption characteristics of any oil droplets.

3.2.1.2 Cold steady state tests

Figures 5 to 7 show the changes in temperature, gas concentrations and smoke levels measured during a cold start, 50kph steady state test. The data is from one particular run with the CVS at 65% but are representative of the data from all the cold start steady state runs. The exhaust temperatures take ~6 or 7 minutes to stabilise; the oil temperature takes ~20 minutes. Exhaust gases take about 5 minutes to stabilise; the smoke readings take ~10 minutes to stabilise. After an initial spike on engine start, the smoke levels are low and then increase to a steady state.

The reproducibility of the data can be seen more clearly in Figures 8 and 9 which show the flux of CO and the smoke readings respectively for three cold start runs. There are some differences on the initial start up, but the data soon settle down to similar values.

3.2.1.3 FAS tests

Table 4 shows the reproducibility of the FAS data collected on different days. All data were collected with the CVS set at 65% (see 3.1.8). Within each day, 5 runs were carried out. The gases and smoke data were logged in real time and the results integrated over 100s. This gives the average behaviour from the whole FAS cycle rather than splitting it down into individual sections that would lead to much greater variation. It also enables the results to be compared against the collected particulate sample, which is an integration over all 10 individual FAS elements within a test.

Table 4. Reproducibility of FAS data.

(Values are quoted \pm one standard deviation, with the percentage of the mean that this represents given in brackets. Run sequence from 3.1.8)

	Day 1 (15/9/98)	Day 2 (16/9/98)	Day 3 (17/9/98)
CO ₂ (g/s)	2.05 \pm 0.05 (3%)	2.03 \pm 0.12 (6%)	2.03 \pm 0.03 (2%)
CO (g/s)	7.5 \pm 2.1 $\times 10^{-4}$ (28%)	6.5 \pm 0.8 $\times 10^{-4}$ (12%)	8.3 \pm 1.1 $\times 10^{-4}$ (13%)
THC (g/s)	1.9 \pm 0.5 $\times 10^{-4}$ (26%)	1.8 \pm 0.2 $\times 10^{-4}$ (11%)	1.9 \pm 0.3 $\times 10^{-4}$ (16%)
NO _x (g/s)	0.019 \pm 0.002 (12%)	0.018 \pm 0.002 (10%)	0.019 \pm 0.001 (7%)
Smoke K (m ⁻¹ s ⁻¹)	0.11 \pm 0.02 (16%)	0.10 \pm 0.01 (9%)	0.11 \pm 0.02 (16%)
Particulate (mg/s)	1.9 \pm 0.4 (20%)	2.00 \pm 0.07 (4%)	2.01 \pm 0.36 (18%)

The reproducibility of this data is at least as good as that from the steady state data. This is partially due to the overall levels being dominated by the very large transient peaks on which there will be better precision due to the large signals.

3.2.1.4 Hot start NEDC drive cycles

Tables 5 to 7 show the reproducibility of data collected from the transient cycles. Five cycles were driven each day. The gas data were logged modally (bag samples were collected for the first and last cycles and always showed reasonable agreement). The gaseous and smoke data can also be split into the respective EC and EU parts of the cycle (Tables 6 and 7). The CVS was operated at 65% for all tests (see 3.1.8).

The reproducibility of these data are generally better than observed for steady state or FAS data. This is probably due to the greater time period for data collection and changes in engine management strategy tending to cancel each other out overall.

Table 5. Reproducibility of NEDC drive cycle data.

(Values are quoted \pm one standard deviation, with the percentage of the mean that this represents given in brackets. Run sequence from 3.1.8)

	Day 1 (15/9/98)	Day 2 (16/9/98)	Day 3 (17/9/98)
CO ₂ (g/km)	136.5 \pm 0.6 (0.5%)	132.9 \pm 0.8 (0.6%)	132.6 \pm 2.2 (1.6%)
CO (g/km)	0.014 \pm 0.008 (54%)	0.033 \pm 0.004 (11%)	0.031 \pm 0.003 (11%)
THC (g/km)	0.014 \pm 0.002 (12%)	0.014 \pm 0.002 (16%)	0.015 \pm 0.002 (10%)
NO _x (g/km)	0.57 \pm 0.05 (8%)	0.56 \pm 0.01 (2%)	0.55 \pm 0.01 (2%)
Smoke K (m ⁻¹ /km)	10.6 \pm 2.0 (19%)	9.9 \pm 0.9 (9%)	11.5 \pm 0.6 (6%)
Particulate (mg/km)	54 \pm 3 (6%)	46 \pm 4 (10%)	55 \pm 6 (10%)

Table 6. Reproducibility of EC drive cycle data. Values are quoted \pm one standard deviation, with the percentage of the mean that this represents given in brackets.

	Day 1 (15/9/98)	Day 2 (16/9/98)	Day 3 (17/9/98)
CO ₂ (g/km)	165 \pm 3 (2%)	158 \pm 2 (1%)	158 \pm 4 (2%)
CO (g/km)	0.003 \pm 0.017 (600%)	0.039 \pm 0.007 (19%)	0.031 \pm 0.009 (29%)
THC (g/km)	0.022 \pm 0.004 (18%)	0.020 \pm 0.004 (21%)	0.020 \pm 0.003 (13%)
NO _x (g/km)	0.67 \pm 0.04 (6%)	0.63 \pm 0.03 (5%)	0.63 \pm 0.01 (1%)
Smoke K (m ⁻¹ /km)	15 \pm 4 (25%)	14 \pm 2 (11%)	17 \pm 1 (8%)

Table 7. Reproducibility of EU drive cycle data. Values are quoted \pm one standard deviation, with the percentage of the mean that this represents given in brackets.

	Day 1 (15/9/98)	Day 2 (16/9/98)	Day 3 (17/9/98)
CO ₂ (g/km)	123.8 \pm 0.6 (0.5%)	121.7 \pm 0.5 (0.4%)	121.0 \pm 1.8 (1.5%)
CO (g/km)	0.020 \pm 0.005 (26%)	0.030 \pm 0.002 (7%)	0.031 \pm 0.001 (4%)
THC (g/km)	0.011 \pm 0.001 (8%)	0.011 \pm 0.001 (12%)	0.012 \pm 0.001 (8%)
NO _x (g/km)	0.53 \pm 0.05 (10%)	0.52 \pm 0.01 (2%)	0.52 \pm 0.01 (3%)
Smoke K (m ⁻¹ /km)	9 \pm 1 (15%)	8.1 \pm 0.6 (8%)	9.3 \pm 0.4 (5%)

Figure 10 shows a reasonable relationship between measured smoke and collected particulate for these cycles. However, the range of data is fairly small.

3.2.1.5 Cold start NEDC drive cycles

Table 8 shows the reproducibility of the three cold start runs. The data show reasonably good consistency. Figures 11 and 12 illustrate the significant changes in gas emissions relative to hot start conditions for the two different parts of the NEDC cycle. This is largely due to the EC cycles where the engine is still quite cold. The CO₂, CO and THC are all higher for the cold start relative to the hot start, whereas the NO_x and smoke are lower. By the time the EU cycle is reached, the vehicle has almost warmed up and so the levels approach those from hot cycles. The overall level of particulates collected is very similar to that from the hot start cycles. This is probably due to most particle generation being from the EU cycle by which time the vehicle is hot.

Figure 13 shows the large (factor of 5) decrease in idle smoke levels observed as the vehicle warms up after a cold start, compared to the ~40% fall for the same data from a hot cycle. The data is taken from the Celesco smokemeter during the idling periods of the EC cycle and reflects the heating up of the catalyst (Figure 14). The slight fall observed in smoke for the hot cycles seems to indicate a degree of conditioning as the cycle progresses, even though the oil temperature is stable through this period. The catalyst temperatures, however, indicate a small degree of change (falling by ~20°C). The gaseous hydrocarbons at idle also fall

during cold cycles but not quite so dramatically, with levels decreasing by less than 50%, whilst they appear stable from hot start cycles.

Table 8. Reproducibility of cold start drive cycle data. Values are quoted \pm one standard deviation, with the percentage of the mean that this represents given in brackets.

	Bag sample	Total cycle	EC cycle	EU cycle
CO ₂ (g/km)	144 \pm 1 (0.8%)	149 \pm 5 (3%)	192 \pm 6 (3%)	130 \pm 4 (3%)
CO (g/km)	0.30 \pm 0.04 (13%)	0.30 \pm 0.04 (13%)	0.87 \pm 0.11 (13%)	0.04 \pm 0.01 (30%)
THC (g/km)	0.029 \pm 0.007 (25%)	0.057 \pm 0.009 (16%)	0.13 \pm 0.02 (17%)	0.024 \pm 0.004 (15%)
NO _x (g/km)	0.37 \pm 0.02 (4%)	0.49 \pm 0.04 (9%)	0.52 \pm 0.05 (9%)	0.47 \pm 0.04 (9%)
Smoke K (m ⁻¹ /km)		8.0 \pm 0.8 (10%)	11 \pm 1 (10%)	6.8 \pm 0.7 (10%)
Particulate (mg/km)	49 \pm 2 (5%)			

3.2.1.6 Conclusion

The vehicle shows good reproducibility from transient condition data. Changes implied by the techniques under test in particulate levels of less than 10% should be treated as due to vehicle variations. Steady state data are likely to be subject to greater variability due to the effects of electronic engine management systems.

3.2.2 Particle sampling data

3.2.2.1 Steady state

Particle mass measurements

Figures 15 and 16 show the mass samples collected with the Andersen impactor during the steady state testing. There is high variability in the mass collected, both between and within tests conditions. The only repeatable data was produced by the raw sample testing. These results highlight two problems with the measurements:

- 1) For the raw exhaust samples, the mass of material collected was high enough to make weighing errors insignificant. However, for the samples taken from the dilution tunnel, this was not the case.
- 2) The Andersen impactor is designed to sample from ambient pressure conditions. The dilution tunnel runs at a negative pressure (compared to

ambient), and it is possible that this prevented proper sealing between each stage.

The 'raw exhaust' mass data shows that most of the particulate mass from this vehicle was found below 300nm (from 70% to 80%). It should be pointed out that this 'raw' exhaust sampling was not carried out isokinetically, due to the variable exhaust flow. It is quite possible that there was under or over sampling of large particles. However, the regulatory filter measurements gave a mass of ~30mg/km at 50kph which compares well with the Andersen data. This suggests that for this condition the impactor is not losing significant numbers of particles affecting the mass, so the observation that ~30% of the mass came from particles >300nm is likely to be valid. However, the regulatory filter paper measurements gave 100mg/km at 120kph, and the Andersen masses were ~40% lower than this, suggesting some losses. However, there is also the uncertainty in the measurement introduced by having to use the pitot tube data to measure the exhaust flow.

Number measurements

Figures 17 to 19 show the total particle flux as measured by the SMPS and CNC at idle, 50kph and 120kph, for each of the five test conditions. Figures 20 to 22 show the SMPS scans for these tests.

At test condition 4 (high temperature), the background levels as measured by the CNC were of the same magnitude as the levels measured at 120kph under normal conditions, whereas the SMPS measured backgrounds were low across all test conditions. It is possible that very small particles (<7nm) were being produced by the high heat on the furnace ceramic tube. An alternative is that the small number of larger particles present in the air break down at high temperature into the small primary particles <7nm in diameter. Whatever the reason, the CNC data from test 4 is of little use.

During the 'raw exhaust' testing at 120kph, the CNC sample line rapidly filled with water. The high water vapour content is an inherent problem with raw exhaust sampling. The collected water 'bubbled' in the sample line, and it is possible that this increased particle losses.

Sampling conditions

The results above show that the sampling conditions tested have very little effect on the measurement. Test conditions 4 (high temperature) and 5 (raw exhaust) posed some problems, with very high background levels on the CNC for the high temperature tests, and with water collecting in the sample lines of the CNC

during the raw exhaust tests. It is also difficult to quantify raw exhaust emissions without a reliable real-time measure of exhaust flow.

Comparison of SMPS to CNC

The comparison of the SMPS to the CNC is best made over steady state tests, where the SMPS scans over its complete size range (over transient tests the SMPS can only monitor one selected size at a time).

At idle there is a fair agreement between the two instruments, although there are some outliers (probably due to the very low particle concentrations at this condition).

Except for test 4 (where the CNC backgrounds were very high) at 50kph the total number concentration measured by the SMPS and by the CNC are in good agreement.

At 120kph the CNC measured a much higher particle flux than the SMPS (about 20x higher) during the tests conducted using the dilution tunnel. For test condition 5 (raw exhaust sampling) there were sampling issues that throw the CNC data into doubt. The CNC measurements at this condition are actually lower than the corresponding SMPS data.

Particle number size split

Alternate measurements were made with the CNC of either total particle number (dilution only), or of particle number below 300nm (impaction of particles >300nm and dilution). The results at 50kph showed a reduction of ~30% when sampling through the impactor (using raw exhaust data), implying 30% of the particles by number are >300nm. However, when equivalent SMPS measurements were made through the impactor, the particle number concentration dropped by a similar amount (~25%). Because the SMPS was not measuring particles >300nm, this implies that the reduction of particle number by the impactor was mainly due to losses of particles <300nm (probably by diffusional losses). This implies the number of particles >300nm is insignificant under these conditions.

At 120kph the CNC showed a reduction of 50% when sampling through the impactor, compared to reduction of 25% as measured by the SMPS. This seems to imply that there is a significant number of particles above 300nm. However, due to a number of reasons it is unlikely that this is the case:

- The Andersen impactor mass measurements show at most 30% of the mass is associated with particles > 300nm. The SMPS data shows that of those particles <300nm, the mean size is ~80nm. If we assume a particle size of 80nm for those <300nm and a worse case scenario of 300nm for big particles,

then those of 300nm comprise only 3% of the total number of particles if they account for 30% of the mass if the particle densities are the same. On this basis, these larger particles would not account for the increase in particle flux observed by the CNC at 120kph relative to the SMPS. However, the fibrilous nature of particulate agglomerates means that particle densities are unlikely to be the same.

- At 50kph the SMPS and CNC measurements have strong agreement, so most particles (by number) probably fall within the SMPS size range (7nm - 320nm)

It is possible, therefore, that the particles seen by the CNC at 120kph but not seen by the SMPS are in fact smaller than the SMPS lower size limit (7nm). Further, if this is the case then a large proportion of these will be lost in the impactor due to diffusional losses.

Repeatability

Figures 17 to 27 show the repeat data for the SMPS and CNC. The repeatability of both instruments is generally good.

3.2.2.2 Cold start steady state tests

Figure 28 shows the SMPS measured flux for successive scans over a cold start steady state 50kph test at four sampling conditions. The average flux over hot start tests was 1.9×10^{12} /s. The average for the cold start testing was slightly higher at 2.1×10^{12} /s, with no obvious effects from the different test conditions.

Figure 29 shows 6 sequential SMPS scans during a cold start steady state test, showing very little change in distribution as the engine warms.

The differences between hot and cold start tests are probably insignificant.

3.2.2.3 FAS tests

CNC and SMPS measurements were taken over FAS tests. Figure 30 shows particle flux measurements as an average of the flux over the complete FAS test, with Figure 31 showing SMPS data. There was very little difference between tests with the CVS at 65% and high dilution tests, and the measurements are fairly repeatable. The SMPS data shows that the size distribution is similar to that seen during the steady state tests.

It should be noted that for raw exhaust measurements, pitot tube measurements of exhaust gas velocity had to be used in order to determine dilution ratios and, therefore, number concentrations and exhaust particle fluxes. These measurements were very noisy and occasionally showed large spikes in the data, which translates to noisy particulate measurement data. This was especially

true during transients which the pitot tube appears incapable of following accurately.

3.2.2.4 NEDC drive cycles

Measurements were taken over NEDC drive cycles at four conditions; CVS 65%, high dilution, high temperature and raw exhaust. The high temperature measurements were affected by high backgrounds, as seen in the steady state tests. Figures 32 and 33 show the average particles per kilometre measured by the CNC over the ECE and EU sections of the drive cycle respectively. There is very little difference between the 65% CVS measurements and those at high dilution, but for the raw exhaust there are big differences. This is mainly due to the problems associated with measurement of the exhaust flow.

Repeatability over the ECE part of the cycle is reasonable, but is much worse over the EU cycle. Most of the variation comes during the 120kph section of the EU cycle (figure 34).

3.2.2.5 Discussion

In this work, the CNC measured more particles at 120kph than did the SMPS. With such a discrepancy between the two instruments, it is important to ensure that the particles measured by the CNC are real particles rather than anomalies caused by the measuring/sampling regime or by the measuring nature of the instrument. Three possible causes of these differences are:

- Spontaneous formation of droplets in the diluter
- CNC measurement errors caused by a discrepancy between its count mode and opacity mode.
- Real particles outside the size range of the SMPS

Spontaneous formation of particles in the diluter

In the diluter, the hot sample air is diluted by cooler dilution air. If there is a large amount of volatile material in gaseous phase in the sample air, there is the possibility of spontaneous formation of droplets. These droplets would then be counted by the CNC, as they are indistinguishable from solid particles. However, during the high dilution tests, the SMPS also sampled through the diluter, and did not see an increase in particle concentration. If droplets are being formed, they are outside the size range of the SMPS.

CNC opacity and count mode

The CNC works in two modes, count mode (low concentrations) and opacity mode (high concentrations). In count mode, individual particles are seen as pulses by the CNC optics. As the concentration increases, these pulses begin to merge together, so individual particles can no longer be measured. The instrument then switches to opacity mode. In opacity mode, the particle concentration is

determined by measuring the light scattered by the particles. Opacity mode is less accurate than count mode, and more susceptible to errors. Figure 35 shows the relationship of particle count to particle opacity, clearly showing how as opacity increases, particle count initially increases, and then decreases.

The CNC operated in count mode at 50kph and idle, but opacity mode at 120kph. The concern was that the large particle concentration measured at 120kph was due to an opacity problem. This would have to be checked at the start of Phase 2b and particle dilutions adjusted if necessary.

Real particles outside the size range of SMPS

The good agreement between the SMPS and CNC at 50kph shows that at this condition there were few particles (by number) >300nm. At 120kph however the CNC saw many more particles than the SMPS. Likewise, over the drive cycles the CNC saw a greater increase over the 120kph section of the EUDC than did the SMPS. These measurements imply that at high speeds the vehicle was producing a significant number of particles outside the measuring range of the SMPS (7nm - 300nm), but measurable by the CNC. The question arises as to how many of these particles are greater than 300nm and how many less than 7nm.

During Phase 1 of this study (literature review) a lot of SMPS data was examined. Much of this data was measured using the SMPS in its 14nm to 720nm range. None of this data showed any significant particle emissions above 300nm. Other Phase 1 data showed mass distributions of vehicles emissions, with peaks at ~200nm and secondary peaks at ~1µm to 4µm, but with never more than 50% of the particulate mass > 1µm. Because of the cubic relationship between particle diameter and particle volume, the volume of a 70nm particle (mean size measured by SMPS) is only 0.5% of that of a 1µm particles (Table 9). Assuming spherical particles and constant density, if 50% of particulate mass was associated with 70nm particles, and 50% with 1µm particles, 99.5% of particles by number would be at 70nm, with only 0.5% at 1µm.

Table 9. Volume ratio of a 70nm particle to particles from 700nm to 4000nm.

Size (nm)	Size (µm)	Volume ratio of a 70nm particle
700	0.7	1.00%
1000	1	0.49%
2000	2	0.12%
4000	4	0.03%

It has generally been observed that the particles are not solid spheres, but rather chain agglomerates and it is possible that they become further removed from the solid sphere model as the size increases. However, even if this reduced the

particle density by a factor of 10, still only 5% of particles by number would be associated with the larger particles.

The measurements made by the CNC through the impactor (cut off 300nm) at 120kph were inconclusive. Impactors are designed to characterise particle size by mass, and although it is known that a significant number of small particles (<30nm) will collect on inappropriate stages due to diffusion, their mass is so small that this effect is insignificant. However, for particle number measurements this effect is substantial.

For these reasons, it is reasonable to measure total number of particles as opposed to number of particles above and below 300nm.

Andersen mass measurements

The mass measurements made by the Andersen impactor from the dilution tunnel did not collect enough mass to allow for accurate weighing of the stages. The technique's validity was shown during the raw exhaust tests, where enough material was deposited on each stage. It is likely that the sensible way to go would be to design a high flow, one stage (plus filter) impactor specially for this role. At a higher flow more material can be collected for better weighing accuracy. The scope of this work package does not allow for such an impactor to be designed and produced. For this reason mass measurements for Phase 2b continued to be made with an Andersen impactor only over drive cycles, where there is more chance of collecting sufficient material.

Sampling

All tests seem to show that dilution method has little effect on the particulate number measurements. This simplifies matters in that a straightforward use of the CVS system in its standard mode can be used without the addition of extra equipment. The use of the raw exhaust was investigated as a potential in-service method. This method certainly has advantages for particle mass measurements using an impactor, but the results are difficult to quantify without the use of a reliable exhaust flow monitor.

3.3 SUMMARY

The main findings of Phase 2a are summarised below.

Instrumentation results

- The CNC + diluter data agrees with SMPS data for idle and 50kph conditions
- At 120kph the CNC + diluter measures particles missed by the SMPS
- There are no significant particle emissions by number above 300nm, therefore a total particle number instrument is sufficient. This also circumvents the diffusional losses of small particles that occur if an impactor is used.

- There are significant particle emissions by mass both above and below 300nm.
- The Andersen impactor gave good results from raw exhaust measurements but the masses collected from diluted exhaust were too small to give reliable data.

Effect of test conditions

- The different sampling schemes studied in this work had no significant effect on particles observed from the vehicle, either in terms of flux or size distribution.
- Cold start steady states did not show significant differences to hot start steady states in terms of particulates.
- Apart from an initial sharp increase in particle count on engine start, cold start NEDC show little difference in particulate emissions to hot start cycles.

3.4 RECOMMENDATIONS FOR PHASE 2B

As a result of the work of Phase 2a, the following recommendations were made for the Phase 2b programme:

- use a CNC type device in all particle mode only;
- use the Andersen impactor with the mass collected split for particles above and below 300nm;
- use the range of cycles studied in Phase 2a except the cold start 50kph steady state and idle conditions, as these do not add any significant value to the measurements;
- use standard CVS 65% sampling conditions; and
- use SMPS at a single size as a baseline with which to compare the behaviour of the CNC.

4. Phase 2b - application to vehicle parc

4.1 EXPERIMENTAL

4.1.1 Selection of test vehicles

Vehicles were selected in the context of relevant technology levels as classified by the existing regulatory framework.

Regulation of light duty vehicle exhaust emissions was first implemented within the EU in 1970 under the original ECE 15 regulation and its equivalent EC directive 70/220/EEC. Pollutants regulated were carbon monoxide (CO), unburnt hydrocarbons (THC) and oxides of nitrogen (NO_x). This directive has been amended 4 times and only applied to gasoline powered vehicles up to and including amendment 3 (78/665/EEC). Amendment 4 (83/352/EEC) also required diesel engined vehicles to be subjected to the emissions test. Up to and including amendment 4, only gaseous emissions were required to be regulated.

Further reductions in gaseous emissions were proposed from amendment 4 and also, for the first time, a limit was placed on particulate emissions from diesel powered vehicles, resulting in a new ECE directive being adopted as Regulation 83 (88/76/EEC). This included the regulation of particulate emissions by mass, adopting US sampling procedures with limits specified in a separate EC directive, 88/436/EEC. These limits were:

1.1 g/test (equivalent to 0.27 g/km) for Type Approval

1.4 g/test (equivalent to 0.35 g/km) for Conformity of Production

Implementation dates were:

New models: 1.10.1989

All production: 1.10.1990

Whilst further reductions in CO and HC+NO_x were proposed in 89/458/EEC, in practical terms these were not implemented by any member state in anticipation of the new Consolidated Emissions Directive (91/441/EEC) which not only covered exhaust emissions (including aspects of durability) but also included limits for evaporative emissions. Also, whilst all previous emissions tests had been carried out over the ECE 15 driving cycle up to a maximum speed of 50 kph, the new directive introduced an additional higher speed cycle known as the Extra Urban Drive Cycle (EUDC) requiring vehicles to be tested up to 120 kph under simulated road load conditions.

Subsequently more stringent limits for 1996 onwards were adopted as Directive 96/12/EC. Separate limits were given for gasoline- and diesel-fuelled vehicles and effectively represented a reduction of 55% in particulate mass emissions from diesel vehicles.

Within this Directive, Conformity of Production limits were brought into line with Type Approval limits.

Implementation dates were 1 January 1996 for new models and 1 January 1997 for all production. Slightly less stringent limits are currently in place for direct injection (DI) engines, with full compliance by 30 September 1999.

Particulate limits are specified as follows:

Indirect injection (IDI) engines	0.08 g/km
Direct injection engines	0.10 g/km (until 30 Sept 1999)
Direct injection engines	0.08 g/km (from 1 Oct 1999)

A group of 9 vehicles in total were procured anonymously for study. Three were fitted with older technology IDI engines originally type approved to 88/436/EEC. The remaining six vehicles were split between IDI and DI vehicles (including the vehicle used in Phase 2a) and complied with directive 96/12/EC.

All vehicles complying with directive 96/12/EC were fitted with oxidation catalysts, those complying with 88/436/EEC were not.

Table 10 shows the vehicles used for testing in this phase of work. It was intended to re-test the Golf TDi used in Phase 2a, but this vehicle was no longer available.

Table 10. Vehicles used for testing.

Make	Model	Engine size (l)	Mileage at start	Reg. number	Fuel system	EC regulatory standard
VW	Passat TDi	1.9	2638	S420SKJ	DI	94/12/EC
VW	Polo DL	1.9	3568	S290SKJ	DI	94/12/EC
Mercedes*	C220	2.2	32501	R14PRY	IDI	94/12/EC
Ford	Fiesta	1.8	11568	R435ETX	IDI	94/12/EC
Citroen	Xantia	1.9	5317	S47ECD	IDI	94/12/EC
Peugeot	405 TD	1.9	121099	L420 KWL	IDI	88/436/EE C
Citroen ⁺	ZX	1.9	125681	K613YM A	IDI	88/436/EE C
Ford	Mondeo	1.8	103450	L285PW	IDI	88/436/EE

				B		C
--	--	--	--	---	--	---

* Automatic transmission

+ Engine re-conditioned at 60000 miles

The same test fuel and lubricating oil used in Phase 2a were employed in these tests. Each vehicle had an oil and oil filter change. They were drained of fuel and filled with the test fuel. They were then conditioned by on-the-road driving. They were prepared for tests by driving 3 EU drive cycles on the evening preceding their testing.

4.1.2 Instrumentation and sampling

For the steady state tests, the SMPS and CNC were run in parallel. Over the repeat drive cycles and FAS tests it was decided to run the SMPS at one size only, 50nm. This allows the repeatability of the CNC measurements to be compared to the repeatability of the SMPS measurements. The Andersen impactor was configured to collect particulate above and below a size of 300nm over the drive cycles.

The same CVS and gas analysis equipment employed in Phase 2a were used in this work. CVS gases were always logged modally - at 1s intervals for all runs apart from FAS tests when 0.2s logging was used. Bag analysis was used for cold start NEDC cycles. Smoke was measured using the Celesco smokemeter and regulatory filter samples were collected.

A single standard sampling scheme was employed - CVS sampling set at 65%.

An initial experiment was performed on the VW Passatt to check whether the CNC in opacity mode gave rise to the large number of particles observed at 120kph relative to the SMPS during Phase 2a. With the vehicle at 120kph, the dilution for the CNC was varied to change operating modes between count and opacity modes in order to make a direct comparison between the two.

4.1.3 Test conditions and protocol

Steady state (50 and 120kph), cold and hot start NEDC cycles, and FAS tests were studied for each vehicle, as in Phase 2a.

Two vehicles were tested per day. Each vehicle underwent the tests in the following sequence:-

Day 1 Cold start NEDC
 50kph steady state
 120kph steady state
 FAS test
 Hot start NEDC
 2 EUDC for conditioning

Day 2 Cold start NEDC
FAS test
Hot start NEDC

On the second day, the next two vehicles were received for preparation for the next day's testing.

The VW Passat and VW Polo only had one cold start each, as time was spent initially investigating particle sampling issues arising from Phase 2a.

4.2 RESULTS

4.2.1 CNC particle sampling effects

For Phase 2a, the CNC operated in count mode at 50kph and idle, but opacity mode at 120kph. The concern was that the large particle concentration measured at 120kph was due to an opacity problem. To check this, a small experiment was performed at the start of the Phase 2b work. A vehicle was run at 120kph, and a CNC sample taken. The dilution ratio was increased slowly, allowing the CNC to switch from opacity mode to count mode. Figure 36 plots the measured particle flux and the dilution ratio. As the dilution ratio increases, the flux (corrected for dilution) remains fairly constant. As the dilution ratio reached ~4500, the CNC switched from opacity to count mode, with no obvious change in the measured particle flux. This shows that the high concentrations measured by the CNC at 120kph are real particles or droplets, and not a result of an opacity anomaly.

4.2.2 Steady state tests

Tables 11 and 12 show the CVS data for the two steady state conditions. The filter particulate data is shown in Figures 37 and 38, the data for the Golf collected in Phase 2a being included. Error bars from Phase 2a have been included for the Golf as an indication of the likely uncertainties in the rest of the data.

It is very noticeable that the Fiesta shows the lowest particulate emissions at 50kph, but the highest at 120kph, perhaps reflecting different priorities in engine optimisation parameters.

Table 11. 50kph steady state data.

Vehicle	CO ₂ (g/km)	CO (g/km)	THC (g/km)	NO _x (g/km)	Particulate (mg/km)	Smoke (m ⁻¹ /km)
Passat	95.6	0.019	0.010	0.377	20.0	no data
Polo	83.2	0.065	0.009	0.174	19.9	5.47
Mercedes	113.6	0.028	0.026	0.193	31.7	7.56
Fiesta	90.1	0.027	0.000	0.288	6.2	1.94
Xantia	103.7	0.000	0.007	0.496	14.8	3.46
Peugeot 405	96.7	no data*	0.011	0.397	11.5	2.52
Citroen ZX	91.4	no data*	0.000	0.448	31.2	4.38
Mondeo	96.5	0.165	0.000	0.159	20.0	4.03
Golf⁺	70.8	0.008	0.034	0.204	31.2	2.95

* CO analysers were not operational

+ Phase 2a data

Table 12. 120kph steady state data.

Vehicle	CO ₂ (g/km)	CO (g/km)	THC (g/km)	NO _x (g/km)	Particulate (mg/km)	Smoke (m ⁻¹ /km)
Passat	184	0.065	0.011	1.28	128	7.0
Polo	165	0.035	0.016	0.65	100	10.1
Mercedes	230	0.056	0.005	1.05	62	5.6
Fiesta	179	0.046	0.011	0.37	243	14.0
Xantia	219	0.021	0.003	1.22	55	4.9
Peugeot 405	200	no data*	0.016	0.89	49	6.0
Citroen ZX	183	no data*	0.054	0.90	220	5.8
Mondeo	207	0.149	0.022	1.01	113	4.6
Golf⁺	165	0.045	0.017	0.95	108	2.3

* CO analysers were not operational

+ Phase 2a data

The relationship between smoke and particulate mass measured from all vehicles is rather uncertain (Figure 39). It seems to show the same trend as in Phase 2a, with different gradients being observed at high and load speed, but there is a fairly large spread in the data.

Older technology vehicles (Peugeot 405, Citroen ZX and Ford Mondeo) tend to show more CO and THC, but particulate and NO_x levels are not necessarily worse than new technology.

Figures 40 and 41 show the SMPS and CNC flux data for the steady state tests. As in Phase 2a, the CNC and SMPS data at 50kph agree well. At 120kph, as in Phase 2a, the CNC measures higher particle concentrations than the SMPS for most vehicles. However, for the Peugeot, the CNC and SMPS are in good agreement at 120kph. For all the Mondeo tests the CNC measurements are very low, and it is possible that there was a problem with the diluter during these tests.

Figure 42 shows the mean particle diameter measured using SMPS at the two steady states. All vehicles give similar size distributions, although most vehicles show a tendency for the particle size to increase at higher speed.

There is very good correlation between the particle flux determined using the CNC compared to the SMPS at 50kph (Figure 43). Particle flux does not appear to correlate to regulated particulate mass (Figure 44) or to measured smoke (Figure 45). This is not particularly surprising because the mass comprises particles with a wide range of overall densities due to their different degrees of small particle agglomeration. This shows that the two measures are complementary rather than comparative.

4.2.3 FAS tests

Table 13 shows the regulatory FAS data from the different runs for each vehicle. Some vehicles had an additional FAS run and this data is included. The gas data from the second run on the Polo was not collected due to a computer fault. One set of CO data for the Peugeot 405 was not collected due to an analyser malfunction. Note that the three sets of Golf data are averages from several runs on three separate days from Phase 2a.

Figure 46 shows the filter particulate data for the different runs. The third particulate sample for the Fiesta appears to be anomalously low. There is no apparent reason for this. Otherwise there is reasonable reproducibility between runs, but without the statistical significance of the Golf data where 5 separate runs were averaged for each point in Figure 46.

Figure 47 shows the rather good relationship between smoke and regulatory filter mass measurements using the data from all vehicles.

Figures 48 and 49 show the total flux over the FAS test as measured by the CNC and SMPS respectively (remembering that the SMPS is only measuring 50nm particles). For each vehicle two FAS tests were performed, indicated by the light and dark bars on the charts. The repeatability of the particle flux measurements is quite good, with the CNC repeatability being on a par to that of the SMPS.

As examples, Figures 50 to 51 show the CNC and SMPS scans for each FAS test for the Citroen Xantia. The CNC scans are much sharper than those of the

SMPS and reflect more the behaviour seen with the Celesco smokemeter. This is due to the relatively long time it takes for the particles to travel through the SMPS, giving rise to lateral diffusion which smudges the FAS peaks. The transfer time through the diluter into the CNC is much less, and so the FAS peaks remain sharp.

There is no evidence of a relationship between particle flux and filter mass measurements (Figure 52), although the CNC and SMPS show a good correlation with each other (Figure 53), except for the CNC measurements for the Mondeo which appear anomalously low as was also observed in the steady state tests.

Table 13. FAS test data from CVS.

Vehicle	CO ₂ g/s	CO g/s	THC g/s	NO _x g/s	Particulate (mg/s)	Smoke (m ⁻¹ /s)
Passat	2.53	1.70E-03	4.11E-04	1.78E-02	3.82	0.225
Passat	2.34	1.48E-03	3.97E-04	1.72E-02	5.03	0.206
Polo	2.25	2.80E-03	8.40E-04	5.18E-03	5.92	0.439
Polo	-	-	-	-	3.68	0.224
Mercedes	2.85	4.50E-02	1.05E-03	8.32E-03	4.17	0.262
Mercedes	3.17	1.48E-02	0.00183	8.30E-03	4.45	0.23
Mercedes	2.95	9.51E-03	1.29E-03	7.93E-03	2.63	0.174
Fiesta	2.09	1.43E-02	4.68E-04	6.05E-03	4.50	0.347
Fiesta	2.02	2.60E-03	3.80E-04	5.94E-03	2.53	0.211
Fiesta	2.21	2.30E-03	3.87E-04	6.79E-03	0.83	0.166
Xantia	2.87	3.90E-03	6.67E-04	1.00E-02	6.87	0.354
Xantia	2.73	3.20E-03	5.16E-04	9.21E-03	3.02	0.219
Xantia	2.73	2.80E-03	4.31E-04	9.08E-03	4.17	0.229
Peugeot 405	2.79	-	1.44E-03	8.65E-03	5.28	0.353
Peugeot 405	2.75	3.59E-02	1.60E-03	9.12E-03	7.20	0.506
Citroen ZX	2.03	1.22E-02	2.30E-03	7.33E-03	10.42	0.498
Citroen ZX	1.99	1.11E-02	2.09E-03	6.95E-03	7.00	0.282
Mondeo	2.03	1.11E-02	1.32E-03	4.37E-03	4.52	0.263
Mondeo	2.15	1.14E-02	1.38E-03	5.24E-03	3.52	0.208
Mondeo	2	9.36E-03	1.01E-03	5.18E-03	3.00	0.218
Golf	2.05	7.50E-04	1.90E-04	1.90E-02	1.90	0.11

Golf⁺	2.03	6.50E-04	1.80E-04	1.80E-02	2.00	0.1
Golf⁺	2.03	8.30E-04	1.90E-04	1.90E-02	2.01	0.11

⁺ Data from Phase 2a, average of 5 separate FAS tests.

4.2.4 New European Drive Cycle (NEDC)

Figures 54 to 61 show the CVS data collected from hot and cold NEDCs. The repeat runs are consistent with each other apart from the particulate filter samples for the hot cycles for the Fiesta and Mercedes. These appear to be anomalously low and coincide with the anomalous results found for the last FAS test for these cars. Note that the data for the Golf is from Phase 2a, the two hot runs shown being the average of runs on two separate days, and the cold run data being the average of all the cold runs carried out for the Golf.

Vehicles generally give slightly more particulates from cold cycles relative to hot start ones (Figure 54). There is roughly a factor of 3 spread in particulate emissions, the Fiesta giving the lowest and the Citroen ZX (the oldest car) the highest.

Smoke levels are very similar overall between the hot and cold runs, with the VW vehicles showing the largest amount of smoke (Figure 55).

CO₂ levels, which are a good indication of fuel consumption, show similar levels for many vehicles, but with the Mercedes showing significantly higher fuel consumption than most vehicles (Figure 56). Cold start cycles generally give an increase in CO₂ emission of ~9% relative to hot starts.

CO levels vary greatly between vehicles and usually show much greater levels for cold starts relative to hot (Figure 57). Older technology vehicles give the most CO emission.

THC emissions also vary greatly between vehicles, with the oldest cars showing the highest levels (Figure 58). Cold starts generally give much higher levels of THC. There is a very good correlation between the levels of CO and THC (Figure 59), both being an indication of incomplete combustion. There is also a degree of correlation with filter mass (Figure 60) but this is noisier.

NO_x emissions are very similar for hot and cold cycles in most cases (Figure 61), but occasionally show slightly lower levels for cold starts (e.g. Xantia, Golf, Citroen ZX), possibly due to lower combustion temperatures.

There is a reasonably good relationship between smoke and filter particulate data for all vehicles except the Citroen ZX, which shows lower smoke, or higher

particulate, than might be expected (Figure 62). The two points below 20mg/km are the anomalously low particulate measurements for the Fiesta and Mercedes.

Figures 63 and 64 show example particle flux traces over repeat drive cycles for the SMPS and CNC respectively for the Citroen Xantia. Each figure contains two cold start tests and two hot start tests. Both instruments give similar repeatability over the cycle. Points to note are that over the first part of the ECE the cold start tests give slightly higher particle fluxes (seen by both instruments). The major difference between the two instruments comes at the high speed section of the EU drive cycle. Here the CNC measured flux rises in proportion to the rest of the scan by a greater margin than seen by the SMPS. This ties in with the divergence seen between the two instruments at 120kph steady state. The final point to note is that the repeatability of the CNC scan is similar to that of the SMPS scan, until the 120kph section, where a greater variation is seen. It should be noted that this large relative increase for the CNC data at the end of the EUDC was not observed for the Peugeot 405, Citroen ZX and Mondeo (the older cars).

Figures 65 and 66 show the average particles per kilometre over each section of the drive cycle for the Citroen Xantia, for SMPS and CNC respectively. These graphs clearly show the higher emissions at cold start over the first section of the ECE. Looking at the average value over the total cycle the difference between hot start and cold start is less.

Table 14 below summarises the average particle per kilometre and the relative standard deviation over the repeat tests as measured by the SMPS (at 50nm) and the CNC (total particle count) for each vehicle tested. These standard deviations have been calculated using both hot start and cold start test data. Ideally figures should be calculated for hot and cold starts separately, but only having two cold start tests per vehicle would make the statistics suspect.

In general the CNC has a slightly higher relative standard deviation (of about $\pm 10\%$) over repeat tests than the SMPS. However, considering the number of tests carried out on each vehicle the differences between CNC and SMPS standard deviations are marginal and may be due more to vehicle reproducibility than measurement technique effects.

Table 14. Average particles per kilometre over whole drive cycle

	SMPS at 50nm (normalised)		CNC	
	Average	\pm Std Deviation	Average	\pm Std Deviation
Passatt	1.74E+14	31%	1.73E+14	12%
Polo	3.76E+14	10%	2.60E+14	14%
Mercedes	2.54E+14	8%	1.63E+14	18%
Fiesta	2.21E+14	5%	2.11E+14	6%
Citroen	2.74E+14	6%	2.09E+14	11%

Xantia				
Peugeot	2.40E+14	6%	1.66E+14	10%
Citroen ZX	3.06E+14	7%	2.30E+14	2%
Mondeo	3.16E+14	9%		

Figure 67 shows the data of Table 14 plotted as a function of vehicle.

There is some degree of relationship between the SMPS and filter mass measurements, particularly if the VW Passat data are ignored (Figure 68). There appears to be very little correlation between the CNC and filter mass data (Figure 69) - the CNC measurements seem to be relatively insensitive to vehicle. Although regulated particulate mass varied by a factor of ~3, CNC particle fluxes varied by ~50%, so any relationship is within the noise of the measurements.

The CNC and SMPS show a good correlation (Figure 70) excepting the Mondeo CNC data which seems to be anomalously low for all measurements.

Figure 71 shows the average particle mass above and below 300nm collected with the Andersen impactor over an NEDC. From this we can see that for all vehicles tested there was more mass associated with particles below 300nm than for those above 300nm.

Table 15 shows the total mass collected and the relative standard deviation of these measurements. This highlights the poor repeatability of these measurements. The Andersen consistently measures less particulate than regulatory measurements, suggesting that there may be some sample loss.

Table 15. Total mass collected by Andersen impactor over NEDC drive cycles

Vehicle	Total mass [mg/km]	Relative Standard Deviation
Passat	12	32%
Polo	38	60%
Mercedes	18	88%
Fiesta	16	42%
Citroen	34	24%
Peugeot	28	54%
Citroen ZX	36	18%
Mondeo	26	42%

4.2.5 Cycle correlations

In order to assess the relevance of each test to representative type approval conditions a comparison has to be made against one of the tests deemed to be representative. For this purpose the hot NEDC has been chosen as the baseline.

Figures 72 to 75 show the relationship between the regulated particulates measured from the different tests compared to the hot NEDC cycle. As can be seen, there is a relationship between the two steady states examined and the NEDC cycle (Figures 72 and 73), with the exception of a couple of outliers. The Fiesta at 120kph seems particularly anomalous. As might be expected, the cold cycle shows a strong correlation to the hot cycle data (Figure 75). However, the FAS test shows little or no correlation (Figure 74), suggesting that it is not representative of how a vehicle may behave under type approval testing.

Figures 76 to 79 show the CNC and SMPS flux data from the different tests compared to the hot start cycle. There is a good correlation for the cold start cycle, and a fair one for the 120kph data. There may be a correlation for the 50kph and FAS data but it is very noisy.

4.3 DISCUSSION

4.3.1 Particle flux measurements

The CNC effect at 120kph (measuring more particles than the SMPS) was seen with all vehicles tested, except the Peugeot. For this vehicle, the CNC and SMPS agreed well at both 50kph and 120kph. During the drive cycle testing, the CNC saw a high burst of particles at the 120kph section of the EUDC for all vehicles except the Peugeot, the Citroen ZX and the Mondeo. These three vehicles were the oldest vehicles used in these tests, which suggests that this could be an engine effect or an after treatment effect.

4.3.2 Particle mass measurements

The Andersen mass measurements were not very repeatable. This could be rectified by taking larger samples to get more mass on each of the two stages. It is also possible that the design of the impactor caused sampling problems. The Andersen is designed to take samples from ambient pressures, so each stage is just sealed with an 'O'ring. The dilution tunnel, however, runs at below ambient pressure, and it is possible that under these conditions the Andersen was leaking.

These tests did show, however, that there is particulate mass both above and below 300nm. It would therefore be sensible to design an impactor specially for this type of measurement. Moreover, an impactor could be designed to sit in the standard regulated filter sample points found on most dilution tunnels.

4.3.3 Sampling considerations

The standard CVS sampling technology used in this work for the particulate measurements imposes no special problems and it is envisaged, therefore, that

the methodologies developed in this work will be directly applicable to other CVS technologies such as critical flow venturi (CFV) systems.

The work in this programme has concentrated exclusively on diesel vehicles, although a range of technologies within that vehicle parc were studied. Special consideration must be given as to whether the procedures adopted in this work will be applicable to alternative engine technologies and fuels. For SI engines and gas fuelled vehicles, the particulate mass levels are generally much lower than diesel equivalents. It is unlikely that a mass impactor will be able to collect enough sample to make quantitative measurements of particulate mass split between size ranges. However, as the mass from these vehicles is so low, this may not be an important parameter. Of greater importance are the particle numbers emitted and their size distributions. The CNC will still be an appropriate technique to use to collect the total numbers of particles emitted from these vehicles, and the sampling schemes and drive cycles employed are likely to remain unchanged. However, measurements on representative samples of these technologies need to be made for confirmation.

4.3.4 Vehicle comparisons

Steady state

Figures 37 and 38 show the steady state levels of regulatory particulate emissions collected from 50 and 120kph steady states respectively. There is no obvious effect of the different engine technologies. CO and THC levels seem to be higher for older technologies, but NO_x levels seem unrelated (Tables 11 and 12).

The particle flux data at 50kph (Figure 40) show similar degrees of variation in flux between vehicles as do the particulate mass measurements, but with the vehicles ranked in different orders. It is noticeable, for example, that the Mercedes gives relatively large particulate mass but small numbers of particles. The variation at 120kph is generally similar (Figure 41), although it is noteworthy that the Peugeot 405 does not appear to give the very large increase in small particles observed with the CNC (the CNC Mondeo data is suspect). Perhaps this is a reflection of the vehicle age (the Citroen ZX had a reconditioned engine), but without more data it is difficult to be sure.

FAS tests

Figure 46 shows the particulate mass emissions from the range of vehicles for the FAS tests. The older technology vehicles (Peugeot 405, Citroen ZX and Ford Mondeo) tend to show slightly higher particulate levels than the newer vehicles. This is also true for the levels of CO and THC, with the exception of the Mercedes which emits particularly high levels of these pollutants.

Figure 48 shows the particle flux measured using CNC during FAS tests. The variation between vehicles is of a similar magnitude to the particulate

measurements, but the order of merit has changed, and there does not appear to be a correlation with vehicle technology.

NEDC

Figure 54 shows the particulate mass emissions from drive cycles. Although an older vehicle (the Citroen ZX) gives the most particulate, two new VWs are the next highest polluters. The smallest vehicle, the Ford Fiesta, consistently gives the lowest particulates except at high speed. Whilst the limits applied to vehicles originally type approved to directive 88/436/EEC were relative to being driven over the urban cycle and were much higher than those applied to vehicles approved to 96/12/EC, all vehicles tested over the NEDC gave cold start particulate emissions within their original type approval limits. It is noteworthy that, with the exception of the Citroen Xantia, all vehicles, including those fitted with DI engines, gave particulate emissions within the limits relevant to the current legislation for IDI engines i.e 0.08 g/km, although it must be appreciated that all tests were carried out on diesel fuel with a sulphur content of 300mg/kg.

In terms of smoke, the newer technologies appear to be worse in most cases than the old (Figure 55). The levels of CO and THC are again significantly higher for the old technologies (Figures 57 and 58). NO_x emissions appear to show no correlation (Figure 61).

Figure 67 shows the particle fluxes measured from the drive cycles. The variation in flux determined using the CNC is relatively small and may be insignificant - in other words, all diesel vehicles studied in this work give more or less the same number of particles from the NEDC. However, at the high speed part of the EUDC, all vehicles except the Peugeot 405, Citroen ZX and Ford Mondeo (although the Mondeo data is suspect) gave much greater increases in CNC signals relative to SMPS. These three vehicles are the oldest and do not have catalysts.

General implications of technology developments

The results from such a small sample of vehicles should be treated with caution. However, in terms of high speed/load particulate number emissions, vehicles fitted with modern fuel injection equipment and emissions management systems to reduce NO_x and particulates appear to perform worse than older technology vehicles. Hence the introduction of number-based regulations limiting ultrafine particulate emissions might compromise the technologies which are currently deployed or under development by the industry to meet the existing planned stages for regulatory emissions limits. Therefore it is possible that either additional aftertreatment measures or a trade off with mass-based regulatory limits might have to be considered in parallel with the introduction of number-based regulatory limits for particulate emissions.

4.4 SUMMARY OF PHASE 2B

4.4.1 Instrumentation

1. The CNC + diluter measurements agree well with SMPS under most conditions.
2. At 120kph, the CNC measures particles missed by the SMPS for most vehicles. These particles are likely to be smaller than the 7nm lower limit of the SMPS.
3. Both flux measurement techniques give good reproducibility.
4. The Andersen impactor gives poor results due to the small amounts of particulate collected. Improvements in the impactor design and using a higher sample volume should substantially improve this.

4.4.2 Vehicle and test effects

1. There was a variation of a factor of ~5 in particulate measurements between vehicles under steady state conditions.
2. There does not appear to be any distinct correlation between measured particulate emissions and engine technology for steady state measurements.
3. Older engine technology tends to give slightly more particulate mass, CO and THC emissions from FAS tests. Particle flux appears to bear little relation to engine technology.
4. Very little variation was observed between vehicles in particle numbers emitted during the NEDC. However, older vehicles appear to show far fewer of the apparently small particles detected during the high speed portion of the EUDC.
5. For the NEDC, smoke, CO and THC levels tend to be higher for older vehicles, though particulate mass does not seem to correlate in the same way.
6. For particulate mass, the two steady states and the cold start NEDC relate well to the hot start NEDC. The FAS test data does not clearly relate to the hot start NEDC.
7. For particle number measurements, the 120kph steady state and the cold start NEDC relate well to the hot start NEDC. The 50kph and FAS tests do not relate well to the hot start NEDC.

5. Conclusions

There is evidence that there is a link between particulate exposure and health, and that the ultra-fine component of ambient particulate may be most significant.

In view of the uncertainty over causal links it is not known whether number, mass or both are the most appropriate particulate markers.

The range of sampling conditions studied in this work do not have a significant effect on particle flux and sizing measurements. This means that standard CVS operating conditions can be used which retains the simplicity of the current standard sampling arrangements. It also means that future in-service testing could be based on raw exhaust measurements, although accurate exhaust flow measurements would be required.

Cold start NEDC and steady state data show reasonable correlations to hot start NEDC data. FAS data appear to show very little correlation to other driving conditions, suggesting it has limited usefulness as an in-service measure by proxy of type approval particulate measurements. There is no obvious reason to change from the standard cold start NEDC that is currently used at type approval.

CeleSCO smoke obscuration measurements are reasonably well related to particulate mass measurements under most conditions, suggesting it is a fairly good indication of the particulate mass emissions under a given driving condition.

The CNC + diluter shows very few particles (by number) above 300nm and can therefore be used in all particle mode as an indicator of ultrafine particulate number.

The CNC + diluter technique is in very good agreement with SMPS except for high speed (~120kph), where the CNC shows a much larger number of particles for most vehicles. The work carried out so far seems to suggest that this is caused by a large increase in very small (<7nm) particles. Both techniques gave good reproducibility. These results indicate that the CNC + diluter can be used reliably to determine particle number emissions.

The CNC and SMPS particle fluxes cannot be used as an indication of particulate mass or vice versa. This is not surprising given the complex nature of particle density and mass distribution, and the two measurements are in fact complementary.

The Andersen impactor shows that there is up to ~30% of mass arising from particles bigger than 300nm. However, measurements with the present arrangement were not totally reliable due to the small samples collected. An improved impactor design, together with an increase in volume sampled, would be necessary to improve this measurement procedure.

The different diesel vehicles studied gave a range of emission levels that did not always correlate to any particular engine technology. The number of particles emitted during a NEDC showed little variation between vehicles. However, more vehicles and engine technologies need to be studied before sensible limits on particle number emissions can be imposed.

6. Recommendations

Based on the work in this report the following preliminary recommendations are made as a route for incorporating size-related particulate measurements into type approval protocols:

1. Until the health position is clarified further, measure particulate in terms of mass and number.
2. As the number of particles above 300nm was found to be insignificant, total particle flux is a convenient indicator of ultra-fine (e.g. <300nm) particle flux.
3. A CNC with diluter is an appropriate technique to measure total particle number flux.
4. It is appropriate for diesel vehicles to split the particulate mass at 300nm.
5. To achieve a split in mass range an impactor can be used, though improved design is necessary.
6. Standard CVS sampling conditions can be used.
7. There is no reason to change from using the standard cold start NEDC.

However, this work, by its very nature, was restricted to a limited range of vehicles and engine technologies. It is necessary to test the appropriateness of the above procedures on a wide range of vehicles and fuels. Although the procedures used to measure particle number flux are likely to be applicable to the range of technologies currently available, the absolute levels of particle numbers may change significantly between engine and fuel technologies. Further clarification work is therefore needed.

1. A more comprehensive study of the diesel vehicle parc to provide greater statistical significance.
2. Further investigation into the large increase in the number of (apparently) very small particles at high speed for some vehicles is needed. The reliability and source of this increase needs to be determined. This could be facilitated

by a rigorous investigation into the mechanisms and capabilities of aerosol measurement techniques in the size range below 10nm.

3. Improved impactor design and measurements are needed to make definitive proposals regarding the implementation of a size segregated mass measurement.
4. The application of the methods developed in this work to alternative fuel and vehicle technologies is needed to test their overall applicability, and to determine sensible regulatory levels of particle measurements.

7. Acknowledgements

Thanks to Gerry Cole, Roger Page and Tony Reading for running the engine test facility, Denise Knight for running the particle sampling equipment and to Frank Lamberts, the CEC project manager for this programme, for his comments and advice.

APPENDIX 1

Description of particle measurement techniques

SMPS & CNC

Electrical mobility aerosol analysers such as the Scanning Mobility Particle Sizer (SMPS) are based on the movement of gas-borne particles carrying a known electric charge towards an electrode of opposite charge. These techniques are widely used in the laboratory, especially in particle formation studies, since they have the potential for very good size resolution in the range 0.01 to 1.0 μm diameter. However, their application in the field is largely restricted to the measurement of low concentrations of stable aerosols, such as may be found in environmental pollution monitoring. Electrical mobility analysers are fairly expensive to purchase, and require skill in the operation and interpretation of the data. However, for diesel particulate measurements they offer in principle the capability of near-real time measurements over the applicable size range since the measurement of size distributions typically requires approximately 1 minute.

Electrical mobility (Z_p) is defined in terms of the velocity component (U_e) that a charged particle experiences under the influence of an external electric field of strength E_e :

$$Z_p = \frac{U_e}{E_e}$$

Z_p is expressed in terms of $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ if U_e and E_e are in units of cm s^{-1} and V cm^{-1} respectively. When a charged particle attains terminal velocity in an electric field, the electric force is balanced by the drag force on the particle. Under Stokesian conditions (sub-micron particles), the expression:

$$Z_p = \frac{n_p e C_c}{3 \pi \mu D_p \chi}$$

is obtained, where n is the number of charges on the particle, e is the elementary unit of charge (4.803×10^{10} statcoulombs), C_c is the Cunningham slip correction factor, μ is the gas viscosity, D_v is the volume equivalent diameter of the particle and χ is the dynamic shape factor (unity for spheres). The above equation provides a means of size-separating particles of known charge on the basis of their differing electrical mobilities.

Electrical mobility analysers are widely used to size particles in the range 0.004 to $1\mu\text{m}$ volume equivalent diameter. Particles much smaller than this lower limit are difficult to charge, whereas multiple charging becomes a problem with micron-sized and larger particles. Electrical mobility analysers are the only high resolution techniques for particles smaller than about $0.1\mu\text{m}$ volume equivalent diameter.

Differential mobility analysers (DMAs) have been developed to capture the narrow range of particles that have a common trajectory within an electrical mobility analyser. They typically consist of an Electrostatic Classifier as the mobility analyser, which is coupled to a continuous flow condensation nucleus counter (CNC). The aerosol flow rate entering the instrument ranges from 0.1 to 1.0 l min^{-1} , although is normally operated at the flow rate of the CNC (0.3 l min^{-1}). Particles larger than $1\mu\text{m}$ aerodynamic diameter are initially removed in a single-stage impactor, since they may carry more electrostatic charge than the data reduction procedure permits, resulting in the propagation of large errors throughout the measured size distribution. The aerosol is then passed through a bipolar charge equilibrator consisting of a ^{85}Kr radioactive source contained within the electrostatic classifier section. Emerging particles carry a Boltzmann distribution of charges (the overall charge is zero, but the aerosol contains well-defined proportions of particles carrying ± 1 , ± 2 , ± 3 charges etc. The electrode voltage is initially set to a low positive potential; particles that have a narrow range of high electrical mobilities (smallest particles) enter the gap and are collected by the detector as a 'monodisperse' aerosol. As the electrode voltage is increased, the sizes of particles exiting the electrostatic classifier also increase, since the electrical mobility of the particles that enter the gap at the base of the electrode decreases. As the particle size increases above $0.05\mu\text{m}$ volume equivalent diameter, the aerosol begins to consist of several monodisperse sub-fractions corresponding to the different negative charge levels allowed by the Boltzmann charge equilibrium. Thus, the signal recorded by the detector during the measurement sequence corresponds to the actual number-size distribution, modified by the presence of a known proportion of multiply-charged particles. The analyser software corrects for these multiply-charged particles up to 6 charges per particle.

Scans can be achieved in as little as 60 s at very high resolution (64 channels per decade of size). In the SMPS the electric field strength in the electrical classifier section is varied monotonically, at the same time making particle number

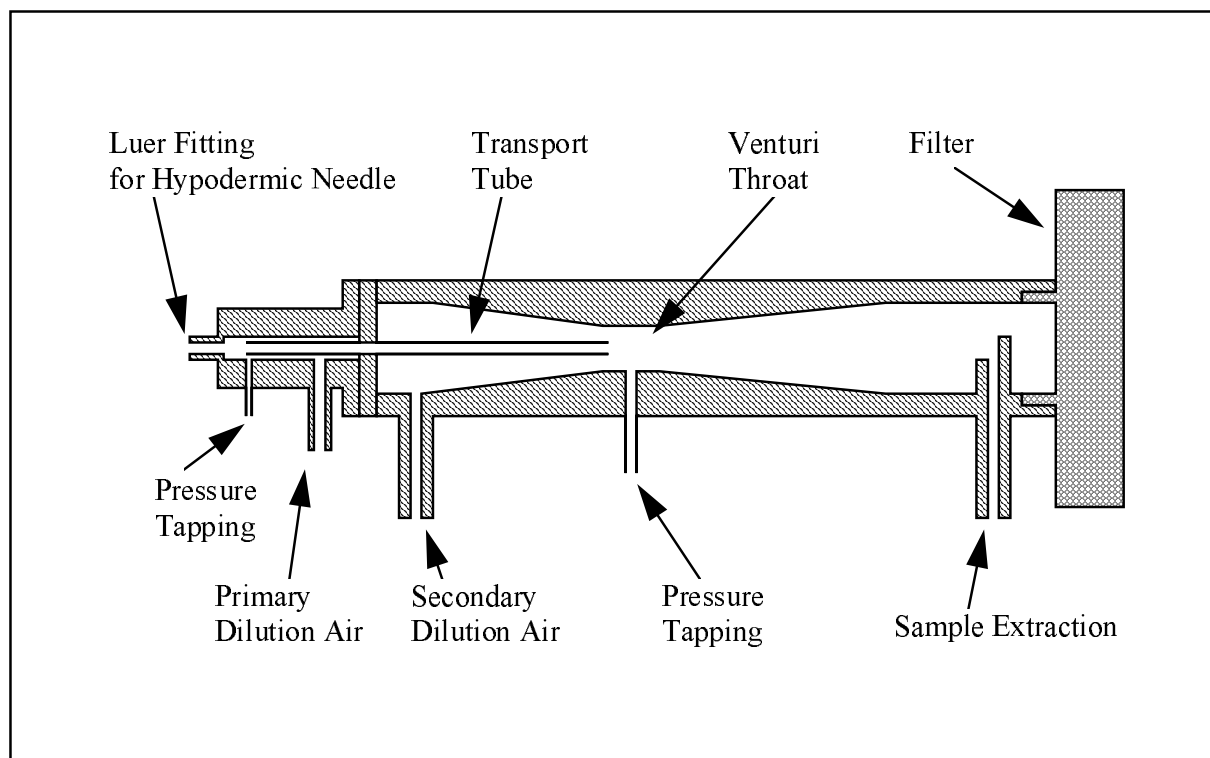
concentration measurements in rapid succession (as much as 10 times per second) using a condensation nuclei counter. The measurement cycle consists of repeated ramps of the central electrode voltage on an exponential scale, increasing from a defined minimum value to maximum field strength, then decreasing the field strength back to the minimum value. The particles entering the sample extraction slot of the classifier will have a monotonic variation in electrical mobility if the electric field strength is varied monotonically. Hence, after making allowances for the finite transit times of the particles within the classifier and from the extraction slot to the condensation nucleus counter, the entire size distribution of the incoming aerosol can be scanned both accurately and rapidly.

Diluter

The diluter design is a two stage unit. A schematic diagram of the unit is shown in Figure A1.

The aerosol is sampled through a hypodermic needle and is immediately mixed with primary dilution air. The diluted aerosol then flows through a transport tube to the throat of a Venturi where it mixes with secondary dilution air. It is the Venturi that creates the reduced pressure that draws the sample in through the hypodermic needle. This allows the sample to be extracted and the diluted sample to be delivered to the measurement instrument all at atmospheric pressure.

Figure A1 Ultrafine Diluter Design



The flow in the hypodermic needle is designed to be laminar and this has a two-fold advantage because the flow rate can be accurately determined from the pressure drop along the length of the needle and secondly because the particle losses due to Brownian diffusion are well known. At the exit of the needle the flow a turbulent jet is formed with the primary dilution air and this promotes good mixing to ensure complete dilution. The rapid initial dilution is designed to lower the concentration of the sample to reduce the effects of concentration dependent processes such as agglomeration. Secondary dilution occurs in the Venturi throat and turbulence in this section of the diluter assures rapid and complete mixing before the flow is expended gradually to recover dynamic pressure. A sample of the diluted aerosol is then removed for analysis by the measuring instrument (in this case CNC).

Andersen impactor

The Andersen II Cascade Impactor is designed to measure the mass-weighted size distribution of aerosol particles in the approximate size range from 0.4 to 10 μm aerodynamic diameter. The instrument is a multistage, multijet device that fractionates particles into size classes based on their inertia. An impactor stage consists of a series of jets beneath which is situated a flat collection medium at a preset distance. Aerosol passing through each jet is directed against the collection medium causing the fluid streamlines to be deflected through 90°. Particles with high inertia are unable to follow the streamlines and impact on the collection medium while particles with a sufficiently low inertia follow the streamlines and miss the collection plate (see Figure A2).

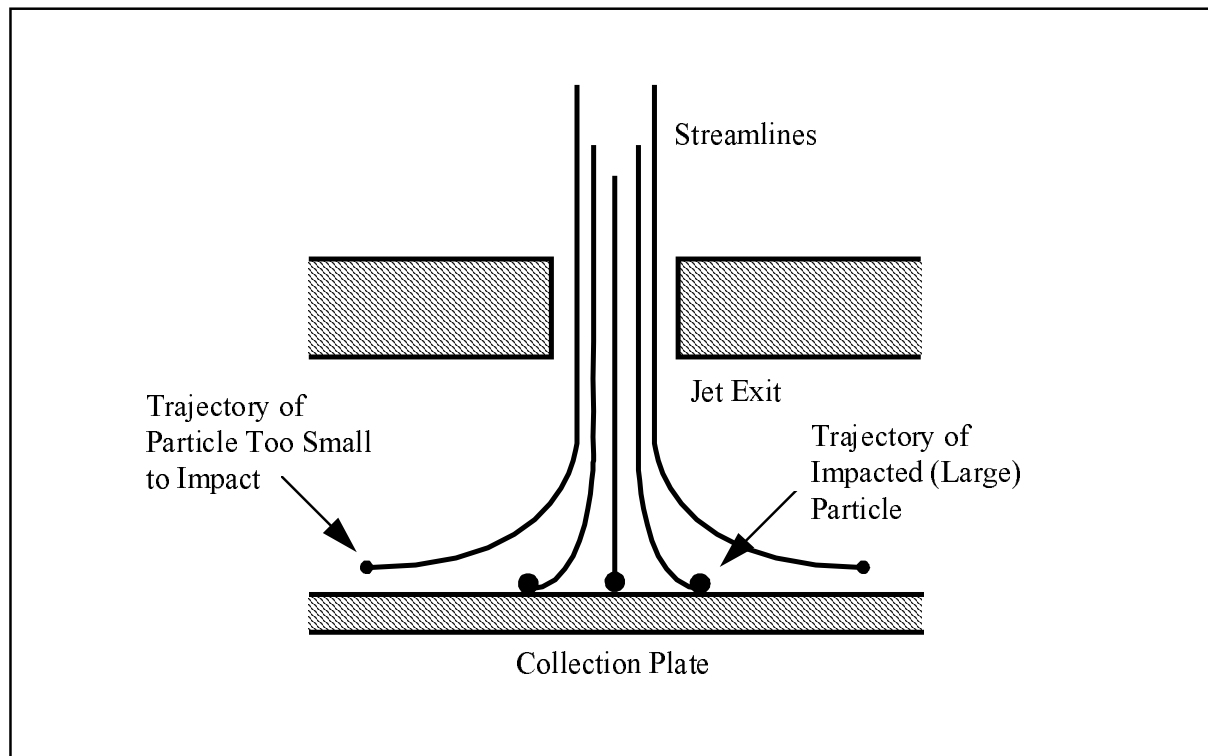


Figure A2 Single Impactor Stage.

A collection efficiency can be defined for each stage as the proportion of particles that enter the stage that are collected by the impaction plate. The ideal collection behaviour of an impactor stage is a step function; particles that are greater than a critical size will be collected whereas smaller particles will pass through the stage unaffected. The actual nature of the collection efficiency curves is, however, S-shaped. In a well-designed impactor the mass of undersized particles that are collected is approximately equal to the mass of oversized particles that are not collected and this allows the size at which 50% of the particles are collected (d_{50}) to act as the representative size of an ideal impactor stage (see Figure A3).

Impactors measure the inertia-based aerodynamic diameter (d_{ae}) of aerosol particles, defined as the diameter of a sphere with a density of 103 kg m^{-3} that has the same settling velocity as the particle under consideration:

$$d_{ae} = d_v \left[\frac{\rho_p}{\chi \rho_0} \frac{C(d_v)}{C(d_{ae})} \right]^{\frac{1}{2}}$$

where d_v is the volume equivalent diameter of the particle, ρ_p and ρ_0 are the particle and reference densities respectively, χ is the dynamic shape factor and $C(d_v)$ and $C(d_{ac})$ are the Cunningham correction factors given by:

$$C = 1 + \frac{2.52\lambda}{d}$$

for particles greater than $0.1 \mu\text{m}$ where λ is the mean free path of the gas and d is the particle diameter.

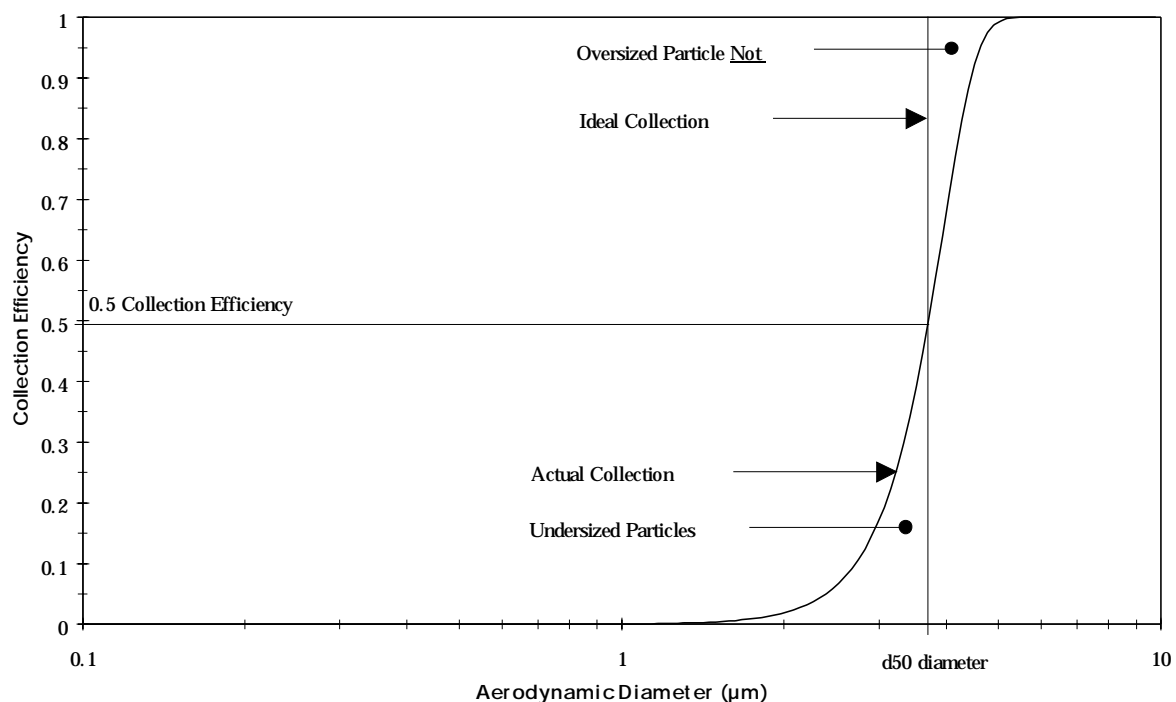


Figure A3 Ideal and Actual Collection Behaviour.

The cascade impactor consists of several impactor stages in series each with a greater collection efficiency with respect to aerodynamic diameter. The Andersen impactor is an 8-stage cascade device that is constructed in three parts: the entry cone, the collection stages and plates and the back-up filter. The instrument is normally operated at 28.3 l min^{-1} although other flow rates and a variety of media can be used on the collection plates (metal, foil, glass, filter paper etc.). The internal construction of the impactor is shown in Figure A4.

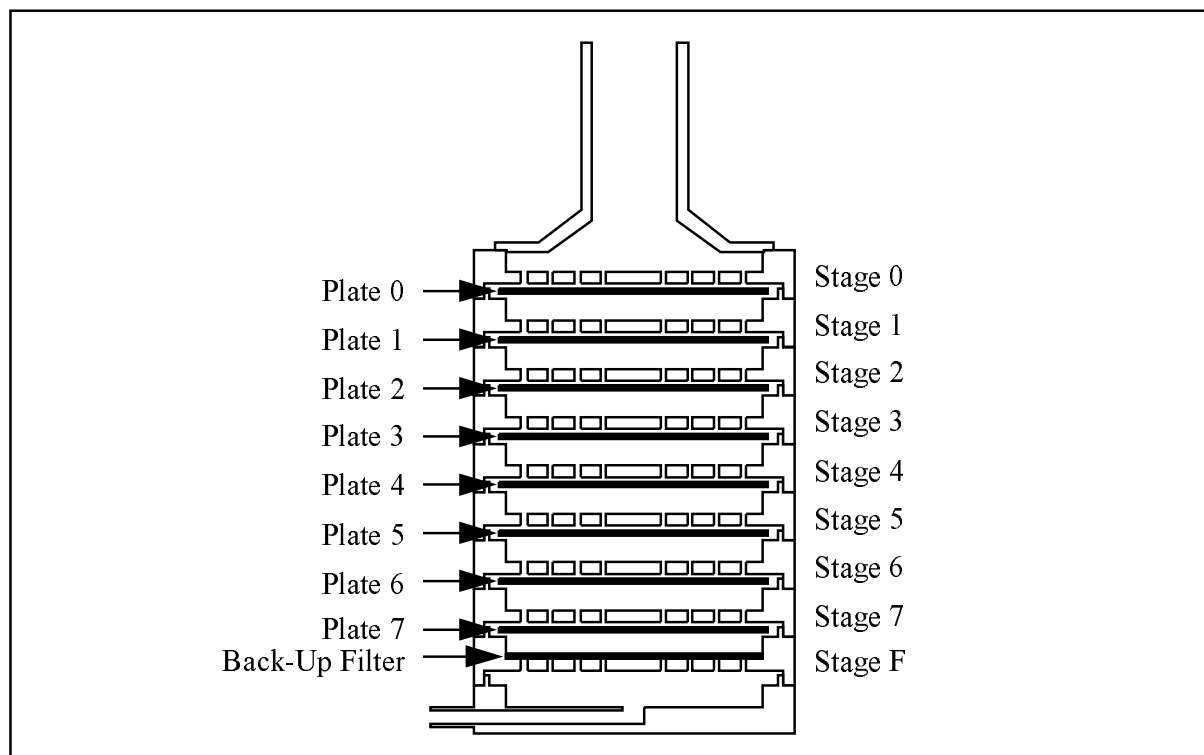


Figure A4 Andersen Mark-II Cascade Impactor.

