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CHARACTERISTION OF EXHAUST PARTICULATE EMISSIONS FROM ROAD VEHICLES

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INTRODUCTION

Following the amendments of the original Clean Air Act, US were the first to regulate particulate mass emissions from Diesel vehicles. In 1982 particulate emission standards for light duty vehicles (LDVs) were set at 0,6 g/mi (CONCAWE, 1995) while heavy duty vehicles (HDVs) followed in 1990 with a value of 0,6 g/bhp h. Emission limits for LDVs aim at a value of 0,01 g/mi in 2008 (Tier 2 standard) or a 98% reduction over a quarter of a century. European legislation followed later, but equally stringent, setting an emission standard value of 0,14 g/km for LDVs in 1992 (Directive 91/441/EEC) and aiming at a value of 0,025 g/km in 2005, reflecting an 82% reduction over a period of 13 years. In the same manner, HDV emission standards decreased from 0,36 g/kWh in 1992 (Directive 91/542/EC) to a foreseen 0,02 g/kWh in 2008 (Directive 99/96/EC), resulting in an overall 94% reduction over 16 years.

Such measures have propelled significant diesel engine advances to control emissions, including direct injection combustion, common rail injection systems and electronic management. Also, exhaust aftertreatment devices started appearing on diesel vehicles, ranging from oxidation to DeNOx catalysts and diesel particle filters. Finally, fuel quality has received severe refinements both with regard to sulphur and aromatics content. From more than 2000 ppm wt. sulphur and unlimited aromatics content in 1987 (Directive 85/716/ EEC), targets for 2005 are a sulphur content of 50 ppm wt. and a maximum 11% vol. aromatics (Directive 98/70/EC).

Those measures clearly indicate that the policy adopted so far, based on the regulation of particle mass emissions, has effectively motivated technology improvements which, in turn, warrant reduced emissions under most operating conditions. However, despite the significant technology improvements already achieved, current research at international level intensively focuses on a more detailed particle emission characterisation. In US, several large-scale projects overviewed by NRC and co-ordinated mainly by CRC (e.g. the Diesel Particulate Sampling Methodology project) have been set up to study diesel particle properties and their association with vehicle technology. Also, they investigate the effect of sampling procedure on a number of particle parameters, including size and number. In Europe, centrally co-ordinated research activities, including projects such as the GRPE initiative and the *PARTICULATES* project have parallel objectives to US studies.

Such efforts establish a discrete shift in the way particle emissions are determined, with mass representing only one dimension of the issue. It is evident that, since such projects are mainly co-ordinated by legislation authorities, future emission standards will most probably make use of their findings to regulate a number of particle emission dimensions. In this respect and in view of the new scientific targets and the potential upcoming emission legislation, aim of this work is to:

1. Identify the needs that called for a broader than the mass-based particle emission characterisation.
2. Explore the alternative measures that may be used for emission quantification, accounting for the importance of information provided and the possibility for their experimental determination.
3. Outline the fundamental criteria which need to be met in developing a particle measurement protocol to be used for policy implementation, on the basis of scientific information collected from on-going experience.
4. Demonstration of how selection of alternative measures and application of the general protocol guidelines can be used to characterise vehicle particle emissions.

VEHICLE PARTICLE EMISSIONS PERCEPTION SHIFT

The shift in the understanding of particle emissions and characterisation originated from two main reasons:

a. The need to understand and explain health effects associated with particle pollution.
b. The measurement challenges which have been arisen due to the evolution of air-quality standards and the continuous decrease of the mass-based vehicle emissions.

Those two reasons are analysed in the following paragraphs.

Particle characterisation requirements from medical studies

Although a detailed discussion of the health effects is not a priority in this work, it is useful to make a synopsis of the directions given from relevant studies. In that respect, epidemiological studies on the association of ambient particle pollution with health hazards showed a weak but consistent correlation of urban aerosol concentration with mortality (e.g. Vedal, 1997). In an effort to better correlate effects with ambient concentration, investigators used the mass of fine particles (i.e. <2,5 μm – $PM_{2,5}$) as a study parameter (e.g. Schwartz et al., 1996). Although the examined correlation was only moderately improved when using size classification criteria, there was an immediate consequence of those studies. First, they questioned whether total particle mass is the exclusive variable to associate with health effects. Secondly, they motivated legislation to consider particle size as an important control parameter. As a result, US EPA revised the National Ambient Air Quality Standards (U.S. Environmental Protection Agency, 1997) to include a day-average $PM_{2,5}$ standard on an annual basis. In Europe, EU Council (European Council, 1999) imposed the obligations to Member States to collect $PM_{2,5}$ data. Broadly speaking, epidemiology studies confirmed that particle size is a variable necessitating further attention.

In parallel, research that focused on the interaction of inhaled particles with lung cells to explain carcinogenic and other effects, raised the significance of additional particle properties. Donaldson et al. (1998) provide an up-to-date summary of a number of plausible mechanisms of lung injury due to particle inhalation. They mainly stress the fact that ultrafine (<100 nm) particles present higher toxicity than larger ones of the same bulk material. This effect was attributed to the larger number and surface area of ultrafine particles compared to the same mass of larger particles. Additionally small particles exhibit a high mass-specific surface area compared to larger ones, hence increasing the probability of interaction between particle and lung cells and subsequently the lung injury possibility. Although such hypotheses have not yet been verified by experiments (Osunsanya et al., 2001), the study of Donaldson et al. (1998) and studies referenced therein, brought on stage the significance of large ultrafine particle populations and the importance of their active surface on health impairment mechanisms. As a consequence, those parameters need to be explored by exhaust particle research to bring more detailed information to health studies.

Finally, it has been a general request (Health Effects Institute, 1999), that especially when referring to vehicle exhaust aerosol studies, chemical composition is explicitly addressed. This is necessary on the one hand to determine substances with direct mutagenic activity and (toxic) metal content and, on the other, to examine an exhaust-specific chemical marker selection for exposure analysis.

Implications from emission standards evolution

Inclusion of particle size as a parameter in air quality standard development, imposed by epidemiology studies, has in turn prompted investigators to study traffic emissions in the atmosphere taking size into consideration. Earlier studies (e.g. Kleeman and Cass, 1998) showed that airborne automotive exhaust particle are an overall ~10 % contributor to $PM_{2,5}$, but almost the exclusive contributor to the mass of ambient ultrafine particles. This outcome, together with the well-established result of high ultrafine particle number population from engines measured in the laboratory (e.g. Kittelson, 1998) initiated further studies aiming at the definition of particle number concentration. Traffic originating particles were studied by Hitchins et al. (2001) in various distances from main roads. They found that the mean size of particles lies in the range of 30 – 50 nm and that number concentration is highest close to the road, later decreasing due to coagulation. Shi et al. (2001) recorded ambient particle concentration originating from a mixed gasoline and diesel fleet and found that the median diameter is in the range of 20 – 30 nm, while a significant number of particles was found even below 7 nm. Wåhlin et al. (2001) showed that diesel traffic induced aerosol pollution comprises particles with a median diameter in the range of 20 nm and 30 nm when using fuel of moderate and low sulphur content respectively and that concentration decreased with fuel sulphur reduction. Finally, Kittelson et al. (1998) observed that the plume of diesel heavy duty vehicles forms a very discrete high-concentration nuclei particle mode under most of the ambient conditions studied, similar to atmospheric conditions. All relevant studies concluded that vehicle exhaust particles in the atmosphere appear as a distribution with a median size in the range of a few nanometer decades. Due to the specific interest of such ultrafine particles on medical research mentioned in the previous paragraphs, recording of the nanoparticle distribution receives increasing attention lately and is already considered as a minimum requirement for the study of engine particle emissions.

In parallel, the significant improvement of exhaust emissions imposed by progressively stringent emission standards brought measurement complications at the testing field. Complications mainly arise from the fact that the mass sampling procedure defined by the legislation imposes a sampling temperature below 52°C, which requires a substantial dilution to cool the exhaust gas. Therefore, the typical dilution ratios employed in the range of 10:1 – or higher – bring the total mass collected on the filter at very low levels. Given as an example only, Salvat e al. (2000) studied the emission behaviour of a production Euro III passenger car equipped with a particle filter. In their measurements, they established a close to ambient but discrete particle number concentration under most conditions examined. However, when measuring the emissions on a mass basis according to legislation, the mass collected on the filter was at the same level with the background concentration and a definite vehicle emission level could not be established. This is an indication that the mass measurement – at least as defined by current legislation – may not be sensitive enough for clean

vehicle technologies of today and the future. Hence, other sampling configurations or particle properties need to be selected for the accurate determination of vehicle exhaust levels.
Historically, only exhaust particulate emissions from diesel vehicles have been regulated. Two thorough studies from ACEA (1999) and CONCAWE (1998) showed that despite the significant reduction of diesel vehicle emissions, gasoline vehicles of respective Euro standards, still exhibit two orders of magnitude lower emission levels, with respect to mass-based emissions. However, both studies consistently demonstrated that, at high speed, gasoline vehicles produce high number concentrations of ultrafine particles, while their mass weighted emissions remain low. Additionally, Patschull and Roth (1995) showed that small 2-stroke engines are very significant contributors of particle number populations, reaching equal to or even higher than diesel engine concentrations (~10^9 cm^{-3}). Those particles mainly originate from unburned hydrocarbons of fuel and lubrication oil. Such studies make clear that in order to increase the reliability of emission inventories, particle characterisation needs to be extended to spark ignition engines taking also into account the particularities of their emissions.
A final consequence of mass emission standard reduction is that the contribution of other vehicle-related sources namely road, tyre, break and clutch attrition becomes increasingly important. There is a great range of emission factors reported in the literature from those sources because of the variety of materials, conditions and particularities of the determination methodologies utilised. Some, only moderate, emission factor values may be used from recent publications to estimate the contribution of non-exhaust particulates to total particle emissions. Muschack (1990) estimates road-wear (for non-studded tyres) at 3,8 mg/vkm. Rauterberg-Wulff (1999) reports a tyre-wear rate of 6,1 mg/vkm for 4-wheel passenger cars. Garg et al. (2000) provide a range of 3,2 – 8,8 mg/vkm for break wear, while no consistent clutch wear rates could be found in the literature. Based on the moderate values selected, non-exhaust vehicle particle rate is estimated at ~16 mg/vkm. Uncertainty granted, this value is at the same order with today's (Euro III – 50 mg/vkm) and especially year 2005 (Euro IV – 25 mg/vkm) exhaust emission standards. It is evident that the contribution of non-exhaust particles in emission inventories cannot be neglected anymore. Although the present work does not further deal with non-exhaust particulates, it is made clear that due to their increasing interest and because current sampling legislation deals only with exhaust emission, future sampling and analysis procedures need also to provide methods for the characterisation of such particles.

ALTERNATIVE METRICS FOR PARTICLE CHARACTERISATION

Size

It is made evident that total particle mass needs to be complemented by additional information to better evaluate vehicle particle emissions and pollution potential. Particle size has received much interest, because of the different health and environmental implications expected for the wide size range covered from particle exhaust emissions. However, size of nanometer and ultrafine particles is an ambiguous quantity. In principle, the size of an actual particle is expressed as the equivalent diameter of a spherical particle of an equal property with the examined particle. Based on this definition, size is recorded with instruments and techniques which may quantify the selected property. Diameters historically used and with practical interest include:

- The mobility diameter (D_p), which is the diameter of a sphere with the same mobility (settling velocity per unit external force). Mobility diameter does not depend on particle mass and is useful when an external force field (mainly diffusional) is applied to size particles. This is the diameter determined with diffusion batteries (Dolan et al., 1980). In the case of an external electrical field, the electrical mobility diameter is proportional to mechanical mobility and number of elementary charges on the particle and it is recorded with electrostatic classifiers. Mobility diameter is significant for ultrafine particles, determining the depth that particles reach in the lung and their deposition locations (Hinds, 1999).
- The aerodynamic diameter (D_a), which, in strict definition according to Hinds (1999), is the diameter of a sphere of unit density (1 g/cm^3) with the same settling velocity. In practical terms, it is the diameter of a unit density sphere with the same inertial behaviour and is most useful for particles above 0,1 μm, where inertia is important. Usually exhaust particle aerodynamic diameter is recorded with impactors. Based on their aerodynamic diameter, particles are distinguished to respirable and non-respirable. Also aerodynamic diameter determines, to a large extent, the residence time of particles in the atmosphere.
- Diameters determined with optical observation, using several microscopy and light scattering techniques (Willeke and Baron, 1992). In case of agglomerated particles, a fractal-like diameter (D_f) can provide the particle level of agglomeration. Fractal-like dimension can be determined directly with optical techniques (Willeke and Baron, 1992) and indirectly with combination of different aerosol properties (Skillas et al., 1998). In practical applications, fractal-like dimension can be used to deduce the active particle surface from mobility diameter and to correlate properties recorded with different instruments (e.g. aerodynamic diameter from impactor with mobility diameter from electrostatic classifier).

Number Concentration

Number concentration is a second metric requested for particle characterisation. In the case of vehicle emissions, number concentration is a more sensitive tool compared to mass to study vehicle effects, because it mainly depends on

ultrafine particles which, although form large number populations, only little contribute to the total mass emitted (Ntziachristos et al., 2001). The most widespread ultrafine number concentration determination devices are the Condensation Particle Counter (Kersten et al., 1991) and the Electrical Low Pressure Impactor (Marjamäki et al., 2002). They both provide the concentration of particles from a few nanometers to the micrometer region in near-real time, although they operate on distinctly different principles. Number concentration information is necessary as input to atmospheric transport or mechanistic aerosol models (e.g. Kleeman and Cass, 1998) to provide the end effect of technology measures to urban air quality.

Surface Area

Particle surface is the means by which particles interact with their environment. This may involve (photo)-chemical reactions and gas-to-particle conversion, liquid adsorption/desorption processes and particle-solid interaction. By such mechanisms particles transform and age in the atmosphere and act on the human body. It should be expected that relevant effects become more prominent as surface area increases. This becomes more important when a specific surface area is considered, e.g. per particle mass or number concentration. As with number concentration, determination of the specific surface area also depends on the surface definition and the method utilised to quantify area.

For particle surface definition, BET method is a widespread analytical technique (Willeke and Baron, 1992). For automotive exhaust aerosols though, other methods are more common involving ion diffusion on the surface and area determination by the current produced by the aerosol stream. Ion production can be achieved in several ways, e.g. by a radioactive source (e.g. Epiphaniometer – Gäggeler et al., 1989) or from a corona discharge (e.g. Nanomet – Kasper et al., 2000). Such methods make use of several assumptions, such as that the rate of ion attaching on the particles is proportional to the surface area available for interaction with the environment and that the charging efficiency is known. Despite the uncertainties, such techniques give the possibility to obtain some measure of particle surface area from vehicle exhaust particles in near real time.

Particle Nature and Chemical Speciation

When vehicle exhaust sampling is performed at high temperature, a large fraction of the ultrafine mode is lost (Ntziachristos and Samaras, 2000). This is a clear indication that part of the nano-size population consists of "secondary" particles from the condensation of volatile species. On this basis, and in the addition to the decision of a suitable metric for particle characterisation, there has been a discussion whether solid and secondary particulates should be equally treated. Concerns are raised due to the possible differences of primary and secondary particles with regard to the behaviour in the atmosphere (e.g. Väkevä et al., 1999) and their health impairment mechanisms (CONCAWE, 1999). The same discussion also touches upon the strong dependence of nanoparticle formation on sampling conditions - namely residence time, dilution ratio and sampling temperature (Abdul – Khalek et al., 1999) - and the complications that this brings when reproducing results in different experimental facilities. This has led several studies to remove condensed particles from vehicle exhaust samples and only focus on solid material (e.g. Mayer et al., 1996). Evidently, such decision relaxes to a large extent the dependence of particle concentration from sampling conditions. However, it also strips significant information which, based on the evidence of atmospheric studies discussed in the previous session, relates to ultrafine particles actually occurring in the atmosphere (e.g. Wåhlin et al. 2001). Perhaps, a more comprehensive approach would be to separate volatile from solid particles, collecting information on both. Available devices for separating the two phases include the thermodesorber or thermodenuder which adsorbs volatile species, permitting solid particles to pass through (Burtscher et al., 2001, Mikkanen et al., 2001) and the catalytic stripper (Abdul-Khalek et al., 1995) which catalytically oxidises and removes from the sample any volatile organic species.

CHALLENGES FOR A PARTICLE MEASUREMENT PROTOCOL DEVELOPMENT

The development of a strict measurement protocol is a prerequisite to obtain a multidimensional particle characterisation. Such a protocol should fully define measurement conditions and data handling in order to produce meaningful results. In accordance to gaseous emissions measurement protocols, such a multidimensional particle characterisation protocol should address the following general guidelines:

1. Robustness: All significant variables relating to test execution, emission quantification and results reduction must be controllable.
2. Flexibility: The range of measurement parameters can be selected in order to adjust the sensitivity of the measurement and accommodate different emission levels.
3. Reproducibility: This does not by definition imply measurement repeatability – which also depends on the condition of the emission source –; it refers to the ability to reproduce testing conditions.

In addition, or in extension to those requirements, a particle specific protocol should also address the aerosol particularities. Time-dependent effects occur in the atmosphere associated with the mixing process and with exhaust aerosol and gas exposure to light and atmospheric reactants. This process, generally called particle ageing, is not possible to simulate in the laboratory due to the associated complexity, but can be approached by selection of sampling parameters (dilution ratio, temperature and residence time). Usually questions are raised whether measured quantities can represent inhaled atmospheric aerosol. Structuring a measurement protocol requires decision on the ageing level

wished to be represented. Different approaches include the representation of fresh aerosol immediately after emission in the atmosphere, a typical ageing between emission and inhalation at a pavement distance and daily ageing to approach average atmospheric condition.
Time in particle characterisation is important also from a sampling point of view. Although gaseous emissions behave as inert gases during sampling and measurement, high aerosol concentrations in the exhaust undergo a dynamic process which may decisively modify the mean size and concentration of the distribution. Mechanisms of time-dependent effects are particle losses on the sampling boundaries due to settling, diffusiophoresis and thermophoresis, particle-to-particle interaction due to coagulation and gas-to-particle conversion. Time constants are such that long sampling times, such as sampling from bags may dramatically decrease concentrations due to losses and particle interaction. In contrast, too short times of a few milliseconds may not be sufficient for an equilibrium concentration to be reached (Abdul-Khalek et al., 1999). From a sampling point of view, once a desired equilibrium condition has been reached, particle analysis should immediately follow. Precautions should also be taken to ensure a smooth transfer of the sample from the extraction point to the instruments in order to avoid boundary interference.
Another aerosol particularity concerns the dependence of particle property on the specific instrument selected to record it. This has been made particularly obvious in the case of particle size, where different size expressions (diameters) are derived from size classification instruments. This dependence may provide difficulties when comparing results or deriving conclusions obtained from different instruments. Obviously, definition of a sampling protocol cannot leave out the instrument selection, which should be based on grounds of both instrument applicability and relevance of output information for the particular study.

EVALUATION OF ALTERNATIVE METRICS

Sampling and Measurement Set-Up

In the framework *PARTICULATES*, an EU 5th Framework Programme sponsored project, a specific particle measurement protocol was developed, which attempted to answer to most of the guidelines outlined above. The following paragraphs present the application of this protocol in order to evaluate a range of alternative particle characterisation metrics. The evaluation relies on testing the behaviour of several metrics to reflect technology improvement and examine their sensitivity to vehicle operation changes.

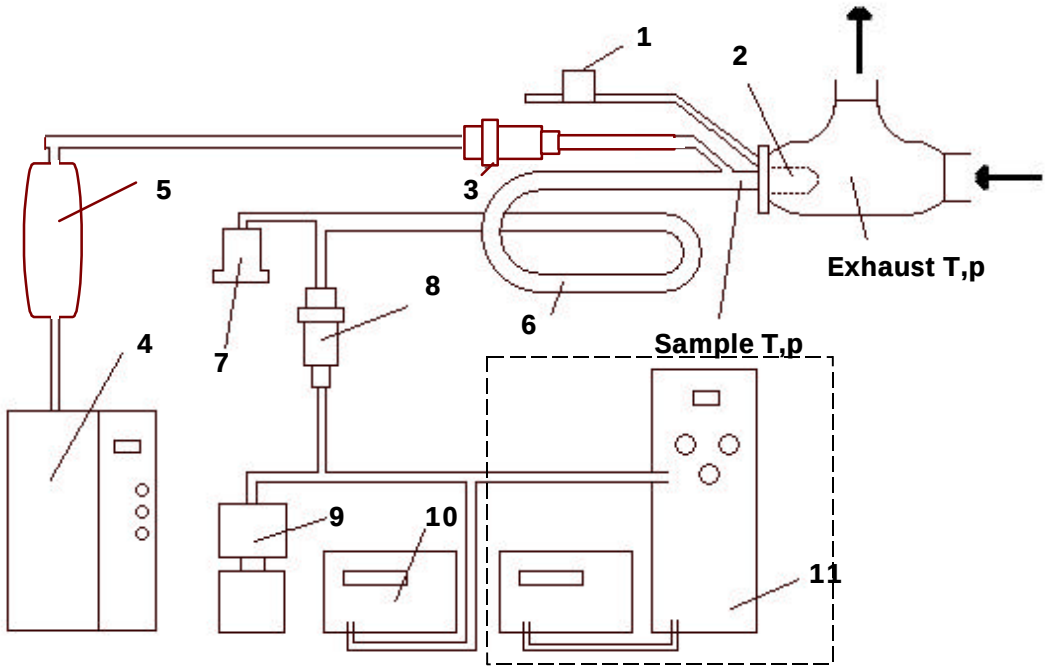

Figure 1: Experimental set-up developed in the *PARTICULATES* project: 1 Mass flow controller for the dilution air, 2 Probe and Primary dilutor, 3 Secondary dilutor (optional), 4 ELPI, 5 Thermodenuder, 6 Ageing chamber, 7 Impactor for mass measurement, 8 Secondary dilutor, 9 Diffusion charger, 10 CPC, 11 DMA + CPC (for steady state tests only).

Figure 1 presents the overall experimental set-up for diluting, sampling and measuring vehicle exhaust aerosol. The sampling conditions selected reflect a short-term ageing at moderate dilution ratio and ambient temperature. The primary dilution ratio is adjusted to a nominal ratio of 12:1 and is achieved with rapid turbulent mixing of the exhaust gas with purified air (time constant ~1 ms). A constant dilution temperature is forced at 32°C. After primary dilution, aerosol is allowed to stabilise for a couple of seconds before analysis, necessary to equilibrate the concentration and

homogenise the diluted sample. These sampling conditions were defined in the framework of a parametric analysis study, specifically designed for this purpose. The selection of the dilution conditions was rather based on the need to define repeatable and relatively stable conditions, in particular as regards production of 'secondary" (nano-) particle production.
The request for short residence time is met by the handling of the sample after ageing/stabilisation. Short lines of few milliseconds residence time and of conducting material have been used for all instruments to minimise losses. Flow remains turbulent in all critical transfer lines while sharp cross-area expansions or contractions and bends have been avoided. Additionally, the initial cooling of the exhaust gas simultaneously with dilution to almost ambient temperature decreases thermal gradients in the sampling system.
A number of instruments have been used to determine several aerosol properties. A TSI SMPS 3936 system has been used to scan particle concentration in the range 10 nm – 1 μm mobility diameter. In case of transient tests, a TSI CPC 3010 model is used to record total particle concentration above 10 nm. The current produced by a unipolar corona-type charger which diffuses ions on particles is used to monitor the diffusion-active particle surface area. The device has been calibrated to convert current values to surface area. This integrated surface area magnitude is generally referred to as "Fuchs" particle surface. Non-volatile (solid) aerosol properties are separated by means of a Dekati Thermodenuder. Solid particle size distribution and number concentration in the range 30 nm – 1 μm aerodynamic diameter are then monitored downstream of the thermodenuder with an Electrical Low Pressure Impactor (ELPI) of the same manufacturer. Ejector dilutors are used to provide additional dilution before the instruments in cases where concentration exceeds instrument range. Also, the legislated CVS method has been used to collect mass of particles on teflon-coated filters and determine PM concentration by weighing the filters on a Mettler Toledo microbalance (precision 10^{-7} g).

Measurement Results and Discussion

Three different Diesel passenger cars of improving technology were selected for particle emission tests. The older car is equipped with an IDI turbocharged engine corresponding to an emission level before the introduction of any EU regulation. This vehicle is referred to as "Euro 0" to compare it with later "Euro"-based emission conventions and corresponds to a late-80s emission level. The second vehicle is a turbocharged DI one, complying with "Euro I" emission standards (Directive 91/441/EEC), which imposed a 180 mg/km limit value for PM and corresponds to mid 90s emission behaviour. Finally, a modern "Euro III" (Directive 98/69/EC Step 1) vehicle completes the test group, equipped with a common-rail engine and an oxidation catalyst and emitting less than 50 mg/km PM. The test vehicles come from different mass production European manufacturers and each on its time has obtained a good fraction of the diesel passenger car market. All tests were conducted with year 2000 specifications fuel (according to 98/70/EC) with a maximum sulphur content of 350 ppm wt. and 11% vol. PAH content.
Several tests over urban, extra urban and steady speed conditions were conducted to evaluate vehicle performance. Results given in the following figures correspond to mean values from one mixed driving (urban and extra-urban) cycle with engine start from ambient temperature (cold-start), two urban driving cycles and three steady speed tests (50 km/h, 90 km/h, 120 km/h). Results are presented in non-dimensional terms, normalised over the average performance of all vehicles and all operation conditions. This index so derived demonstrates the relative difference in emission level between the three vehicles. The sensitivity of each variable on vehicle operation condition is given with the error bars associated with each point. The error bars correspond to the 95% confidence interval for the six operation conditions tested. Examination of the figures can give generic trends regarding the dependence of proposed metrics on vehicle technology and operation.
Figure 2 compares the PM and active surface levels for the three vehicle technologies. As expected, PM reduction is consistent with technology improvements, with the older car emitting three times more than the recent technology. PM data show little dependence on vehicle operation condition. However, Fuchs surface is not equally consistent with technology improvement. The Euro III vehicle emission level substantially overlaps with the Euro I one, exposing a substantial dependence on operation condition.
Significant overlapping occurs also in the total number concentration and median particle size shown in Figure 3. Probably the most significant outcome of Figure 3 is the strong dependence of the metrics used on vehicle operation condition: there is an over two-fold difference between operation conditions for the same vehicle. In contrast, the difference between Euro 0 and Euro III is no more than 40% as regards mean concentrations and only a few percent units as regards median particle size. Practically, since dependence on operation condition is so strong, the relevant position of the mean concentration and median size for each vehicle is rather a matter of operation condition selection than absolute difference of the vehicle emission levels.
The strong dependence of total particle metrics with operation condition is obviously the outcome of the nuclei mode particularities. Specific engine operation and fuel combination can provide high concentration of volatile species, which, under moderate sampling temperature, condense to form a high concentration of ultrafine (<100 nm) particles in addition to the size distribution of solid-core particles. Formation of the nuclei mode increases particle concentration significantly with a subsequent decrease of the mean size. The occurrence or not of the high nuclei-mode is responsible for the sensitivity of proposed metrics with vehicle operation conditions.

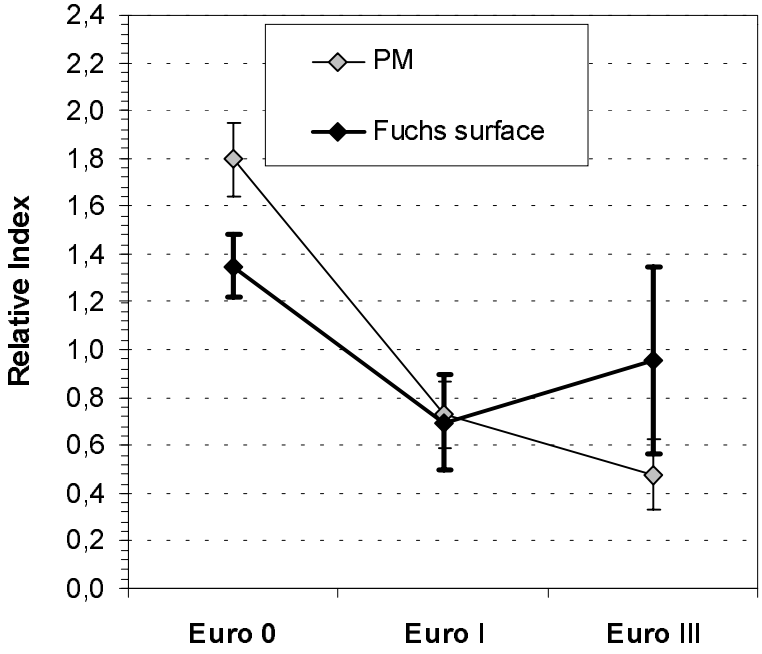

Figure 2: PM concentration and diffusion active surface of emitted aerosol.

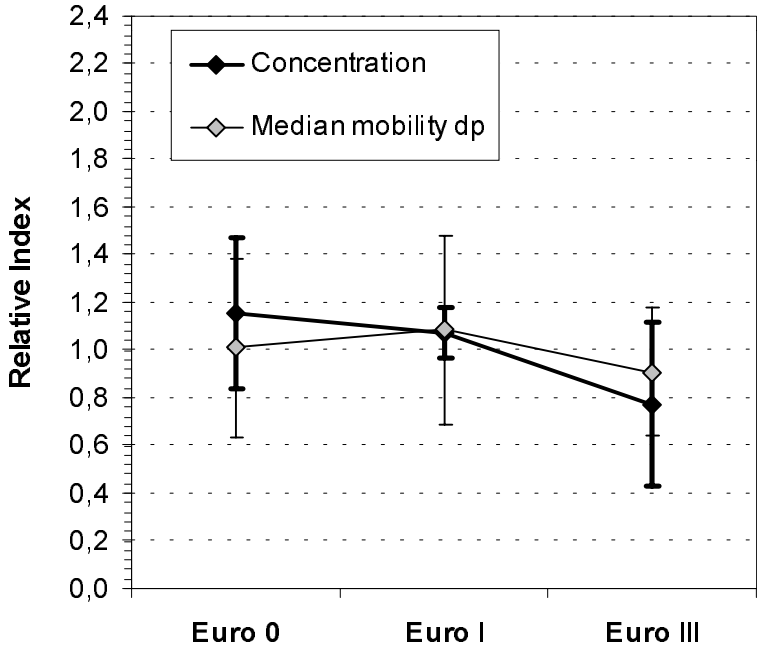

Figure 3: Number concentration and median size (based on electrical mobility – measured over steady state tests only) of particles in the range 7 nm – 1 μm.

In the case of solid particles (Figure 4) technology improvement is clearly demonstrated with a consistent, almost 10-fold, reduction of number concentration between the Euro 0 and Euro III vehicles. Additionally, little sensitivity is shown to vehicle operation condition. On the other hand solid particle median size exhibits the same behaviour as median mobility based particle size. In this case, a difference of only a few percentage units is observed for the three vehicles, not consistent with technology improvement. Furthermore, solid particle mean size dependence on operation condition is negligible. Mean size of solid particles seems to vary little for diesel vehicles.

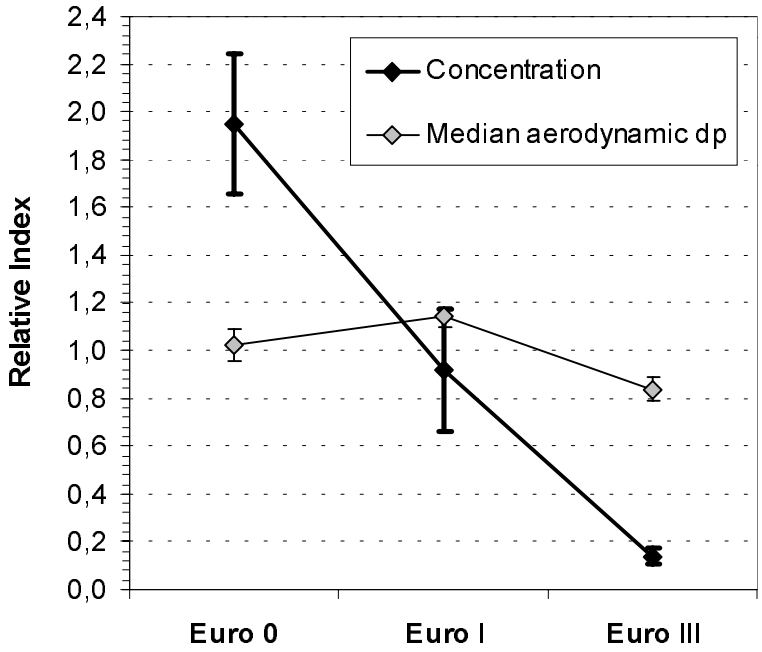

Figure 4: Number concentration and median size (based on aerodynamic diameter) of solid particles in the range 30 nm – 1 μm.

SUMMARY AND CONCLUSIONS

Based on the above observations, the following conclusions can be drawn:

1. PM reductions brought by technology improvements are not consistently accompanied by decreases in total particle number concentration and surface area. Hence, the introduction of additional metrics seems justified for more effective particle emission understanding and control.
2. All magnitudes referring to total particle emissions show a higher sensitivity to vehicle operation condition than to vehicle technology. This observation is associated with the formation of nuclei mode, which is not obvious from PM information. Metrics such as total particle concentration and mean particle size is necessary to fully explore emission performance under different operation conditions.
3. Technology improvement is strongly reflected to solid particle number concentration. This may provide an additional, more sensitive measurement complementing PM mass emissions.
4. Median size is the least sensitive metric to either vehicle technology improvements or vehicle operation condition. This information can potentially be used to distinguish diesel particle emissions from other sources.

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