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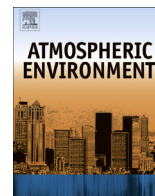
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## Detailed diesel exhaust characteristics including particle surface area and lung deposited dose for better understanding of health effects in human chamber exposure studies



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### H I G H L I G H T S

- DE properties vary significantly under the same DEP mass concentration exposure.
- Reporting detail DE characteristics is needed to explain observed health effects.
- DEP agglomerated structure has to be accounted for to not underestimate lung dose by surface.
- Use of size dependent effective density prevents overestimation of lung mass dose.
- Gas phase components known for carcinogenic and irritation effect should be reported.

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### A B S T R A C T

Several diesel exhaust (DE) characteristics, comprising both particle and gas phase, recognized as important when linking with health effects, are not reported in human chamber exposure studies. In order to understand effects of DE on humans there is a need for better characterization of DE when performing exposure studies. The aim of this study was to determine and quantify detailed DE characteristics during human chamber exposure. Additionally to compare to reported DE properties in conducted human exposures. A wide battery of particle and gas phase measurement techniques have been used to provide detailed DE characteristics including the DE particles (DEP) surface area, fraction and dose deposited in the lungs, chemical composition of both particle and gas phase such as NO, NO<sub>2</sub>, CO, CO<sub>2</sub>, volatile organic compounds (including aldehydes, benzene, toluene) and polycyclic aromatic hydrocarbons (PAHs). Eyes, nose and throat irritation effects were determined. Exposure conditions with PM<sub>1</sub> (<1 μm) mass concentration 280 μg m<sup>-3</sup>, number concentration 4 × 10<sup>5</sup> cm<sup>-3</sup> and elemental to total carbon fraction of 82% were generated from a diesel vehicle at idling. When estimating the lung deposited dose it was found that using the size dependent effective density (in contrast to assuming unity density) reduced the estimated respiratory dose by 132% by mass. Accounting for agglomerated structure of DEP prevented underestimation of lung deposited dose by surface area by 37% in comparison to assuming spherical particles. Comparison of DE characteristics reported in conducted chamber exposures showed that DE properties vary to a great extent under the same DEP mass concentration and engine load. This highlights the need for detailed and standardized approach for measuring and reporting of DE properties. Eyes irritation effects, most probably caused by aldehydes in the gas phase, as well as nose irritation were observed at exposure levels below current occupational exposure limit values

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given for exhaust fumes. Reporting detailed DE characteristics that include DEP properties (such as mass and number concentration, size resolved information, surface area, chemical composition, lung deposited dose by number, mass and surface) and detailed gas phase including components known for their carcinogenic and irritation effect (e.g. aldehydes, benzene, PAHs) can help in determination of key parameters responsible for observed health effects and comparison of chamber exposure studies.

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## 1. Introduction

Elevated exposures to diesel exhaust (DE) have been linked with variety of health outcomes including irritant effects, respiratory symptoms, immunologic, lung inflammatory, cardiovascular effects and lung cancer (Ris, 2007). Recently, DE was classified as a Group 1 carcinogen by the International Agency for Research on Cancer (Benbrahim-Tallaa et al., 2012). Levels of occupational exposure to DE are usually higher than the general population.

DE comprises a complex mixture of particle and gaseous components which vary depending on engine type, fuel characteristics, operating load and presence of emission abatement techniques. During chamber exposure studies DE characteristics can be influenced by generation and dilution systems used.

The DE particles (DEP) are almost exclusively found in the submicrometer fraction ( $<1 \mu\text{m}$ ). Two size modes can typically be distinguished, an accumulation mode and a nucleation mode (Burtscher, 2005). The volatile material in the hot exhaust can during dilution and cooling condense on the solid soot particles (accumulation mode) and/or through nucleation form new particles (nucleation mode). In general the chemical composition of DEP comprise elemental carbon, organic carbon, sulphur compounds and trace metals from lubricating oil and engine wear. Organic substances identified as adsorbed onto particles include C14–35 hydrocarbons (Ris, 2007). The oxygen to carbon (O:C) ratio of the particulate organic fraction of fresh DEP is typically low  $<0.1$  (Aiken et al., 2008).

In majority of human chamber exposure studies, conducted by research groups in Umeå University (Sweden), University of Edinburgh (UK), University of Washington (USA), Los Amigos Research and Educational Institute (USA), US EPA and University of North Carolina (USA), University of British Columbia (Canada) and Rutgers the State University of New Jersey (USA) (summarized in a review by Hesterberg et al., 2010 and in Appendix A), mass concentration of DEP ranged between 60 and  $300 \mu\text{g m}^{-3}$  and was frequently treated as the major exposure metrics. Apart from the particle mass concentration, the following characteristics have been indicated as important when linking with health effects: particle size, number concentration, surface area, chemical composition, solubility, volatile and non-volatile mass fraction, organic compounds including PAHs, soot core and metals (Giechaskiel et al., 2009; Oberdorster et al., 2005). Even if consensus on the most appropriate metrics has not been reached, correlation of particle surface area to inflammatory response has been recognized (Donaldson et al., 2001; Oberdorster et al., 2005). However particle surface area is not reported in published chamber exposure studies with one exception (Rissler et al., 2012). Respiratory tract deposition is an important link between exposure to DE and observed health effects but lung deposited dose is not reported in the exposure studies with one exception for one type of engine used in Umeå University studies (Rissler et al., 2012). In general DEP properties are not well characterized in chamber exposure studies.

DE consist of a mixture of gaseous components and among them, the aldehydes (formaldehyde, acetaldehyde and acrolein), benzene, 1,3 butadiene, PAHs and nitro-PAHs are important due to

their potential carcinogenic effects (Benbrahim-Tallaa et al., 2012). Aldehydes, alkanes, alkenes, oxides of sulphur and nitrogen, are known to induce respiratory tract irritation given sufficient exposure. Gas phase characteristics specifically those having potential carcinogenic or irritation effects, as listed above, are not well characterized in the human exposure studies.

There is a need to pinpoint species-specific or combination of few DE characteristics that would explain observed health effects. To accomplish it detail DE characteristics should be reported as so far such trends are not easy to distinguish.

The aim of this study is to provide detailed knowledge on DE characteristics, indicated as important for health effects assessment. Additionally to compare to DE properties in conducted human exposures as well as highlight the range of DE characteristics that can be determined with known methods and instrumentation. In contrast to most of the human exposure studies we determined DEP surface area, size dependent effective density, fraction and dose of DEP deposited in the lungs as well as chemical composition of both gas and particle phase. These are the DE properties, which were indicated as important to quantify in order to understand the effects of DE on humans. An additional objective was to determine eyes, nose and throat irritation effects assessed via self-rating questionnaires and medical assessment.

This work is a part of a larger DINO project “Health effects of combined exposure to diesel and noise”, that aimed to determine influence of combined exposure to DEP and traffic noise on human health. Health effects of DE exposure in DINO study were reported by Xu et al. (2013) and concluded that short-term exposure to DE at  $\sim 300 \mu\text{g m}^{-3}$  caused temporary decline in peak expiratory flow in healthy subjects and the increase in leukocyte cell counts in peripheral blood indicated a systemic inflammatory responses.

## 2. Materials and methods

### 2.1. General methodology

In a laboratory chamber eighteen healthy volunteers (9 men and 9 women), all non-smokers of ages 40–66 (mean 51 years) were exposed twice to DE with high DEP concentration ( $\sim 300 \mu\text{g m}^{-3}$ ) and twice to filtered air (FA) with low particle concentration ( $\sim 2 \mu\text{g m}^{-3}$ ). The data presented here were extracted from the DINO project with four exposure conditions: 1) Reference exposure: FA and low traffic noise (46 dB(A)), 2) Diesel exposure: DE and low traffic noise, 3) Noise exposure: FA and high traffic noise (75 dB(A)), 4) Diesel and noise exposure: DE and high traffic noise.

In this study exposures were merged into two groups: 1) DE exposure that comprised of diesel and diesel and noise exposures and 2) FA exposure comprised of reference and noise exposures.

Test subjects (three at the one exposure, relaxed sitting) were exposed for three hours to each exposure scenario with at least one week interval between each scenario. The study was approved by the Regional Ethical Review Board.

The methodology used in DINO study is described in Wierzbicka et al. (2011).

## 2.2. Exposure chamber, diesel exhaust generation and dilution system

Details are presented in [Appendix B](#). In short, exposures took place in a 22 m<sup>3</sup> stainless steel chamber. Previous studies performed at Lund University proved that the chamber is suitable for human exposure studies ([Isaxon et al., 2013](#)).

The diesel exhaust was generated by an idling (900 rpm) Volkswagen Passat TDI (–98, 1900 cm<sup>3</sup>, 81 kW) placed outside the laboratory. The fuel used was Swedish Environmental Class 1 (EC1) diesel: sulfur content <10 ppm, cetane number min 51, aromatics 4% volume, PAHs < 0.02% volume. The initial boiling point was 180 °C and 95% volume boiling point was 340 °C. Diesel exhaust was diluted in a specifically designed two stage system, presented in [Fig. 1](#) and described in [Appendix B](#).

## 2.3. Measured characteristics, instrumentation and analysis

### 2.3.1. Particle number concentration, size distribution and mass concentration

The particle number concentration and size distribution (10–500 nm) was determined with a scanning mobility particle sizer consisting of a differential mobility analyzer (DMA, Vienna-type, 28 cm long) and a condensation particle counter (TSI Inc. 3010).

The PM<sub>1</sub> (<1 µm) mass concentration were determined with a Tapered Element Oscillating Microbalance (TEOM, R & P Inc., model 1400a). PM<sub>1</sub> cyclone and a nafion drier were installed upstream TEOM. PM<sub>2.5</sub> (<2.5 µm) for gravimetric analysis was collected during 14 DE and 2 FA, using cyclones (GK2.05 KTL) and sampling pumps (BGI 400S) ([Sallsten et al., 2006](#)).

### 2.3.2. Particle effective density and mass size distribution

For fresh diesel exhaust particles (porous agglomerates) effective density (definition and details in [Appendix C](#)) decreases with increasing size of agglomerated particles and differs considerably from bulk material density ([Maricq and Ning, 2004](#); [Park et al., 2003](#); [Rissler et al., 2013](#); [Virtanen et al., 2004](#)). To obtain mass size distribution on the basis of measured number size distribution information about size dependent effective density of particles is needed and was determined with a system that combines a DMA and Aerosol Particle Mass Analyzer (APM) in series, a so-called DMA–APM. For details see [Appendix C](#).

### 2.3.3. Particle surface area and size distribution

Particle surface area (definition in [Appendix C](#)) was calculated for three cases: 1) using Rissler's model ([Rissler et al., 2012](#)) that takes into account agglomerated structure of DEP (details in

[Appendix C](#)); 2) assuming spherical particles; 3) using Maynard's algorithm ([Maynard, 2003](#)), where particle surface area is estimated on the basis of measured particle mass and number concentrations, assuming specific geometric standard deviation (GSD) and known bulk material density.

Particle surface area distributions for estimation of total deposited particle surface area were obtained by combining the surface area of individual agglomerates as a function of particle size (based on Rissler's model using DMA–APM measurements) combined with SMPS data (details in [Appendix C](#)).

### 2.3.4. Particle morphology and primary particle size

Transmission Electron Microscopy (TEM) (100 kV PHILIPS CM10) was used to study the morphology of the particles. A Nanometer Aerosol Sampler (model 3089, TSI Inc.) was used to collect the particles onto carbon coated copper grids. From 30 randomly selected aggregates about 200 primary particles were sized to determine the size of primary soot particles, using a software "ImageJ" (Image Processing and Analysis in Java).

### 2.3.5. Particle chemical composition – PAHs, OC/EC

PM<sub>2.5</sub> collected on filters during 14 DE and 3 FA exposures (3 samples per each exposure) for gravimetric analyses were also analyzed for particulate-bound PAHs using high resolution gas chromatography and low-resolution mass spectrometry (HRGC/LRMS) ([Kliucininkas et al., 2011](#)).

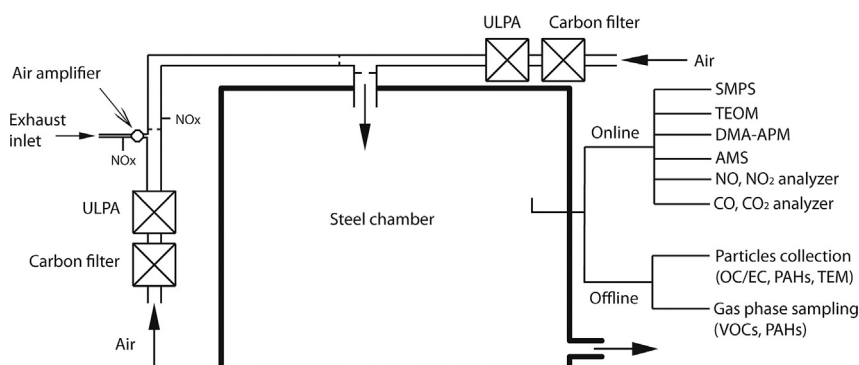
A set-up for PM<sub>1</sub> collection for organic (OC) and elemental (EC) carbon analysis was used ([Wierzbicka et al., 2005](#)). A thermal–optical method (NIOSH 5040 protocol), using a carbon analyzer developed by Sunset Laboratory Inc., was used for the analysis ([Johnson et al., 1981](#)).

With the High Resolution Time of Flight Aerosol Mass Spectrometer (HR-ToF-AMS, Aerodyne Research Inc.) the composition of the organic coating on the soot cores was investigated since evaporation occurs at 600 °C ([Jayne et al., 2000](#)). The atomic ratios were calculated through the algorithm of [Aiken et al. \(2008\)](#). The mass spectra were analyzed with IGOR pro 6 (Wavemetrics, USA) using SQUIRREL 1.51 and PIKA 1.10.

### 2.3.6. Gas phase analysis – NO<sub>x</sub>, CO, CO<sub>2</sub>, VOCs and PAHs

Concentrations of NO and NO<sub>2</sub> in the exposure chamber was measured with an Eco Physics CLD 700AL chemiluminescence NO<sub>x</sub> analyser (0.001 ppm resolution, 1% linearity). The CO levels were monitored with a Maihak Unor 9 CO monitor (resolution 0.1 ppm). CO<sub>2</sub> was monitored with a Riken Keiki model RI-411A.

Measurements of gaseous PAHs and volatile organic compounds (VOCs) (as listed in [Table 4](#)) were done using active sampling with Perkin Elmer tube samplers, filled with Tenax TA (Scientific



**Fig. 1.** Schematic of dilution system and the instruments used for the particle and gas phase characterization.

Instrument Services, Inc., USA). The samples (3 samples per exposure, in the same exposures as for particle bound PAHs) were analyzed using an automatic thermal desorber (ATD, Unity Markes International Limited) and HRGC/LRMS (Kliucininkas et al., 2011). Measurements of 1,3-butadiene were performed using ultra diffusive samplers (SKC Inc., USA) filled with Carbopack X (Supelco, USA) in 7 DE and 2 FA exposures with 3 samples per exposure (Strandberg et al., 2005). Active sampling of formaldehyde and acetaldehyde was performed using pumps and Sep-Pak 2,4-dinitrophenylhydrazine (DNPH)-impregnated silica cartridges (Waters, USA) (Sallsten et al., 2006) in 6 DE and 2 FA exposures (2 samples per exposure).

#### 2.4. Lung deposition and dose

The deposited dose with respect to particle number, mass and surface area was calculated according to:

$$\text{Dose} = \text{TDF} \cdot C_{\text{in}} \cdot t \cdot Q \quad (1)$$

where TDF is total deposited fraction, defined as  $\text{TDF} = \sum \text{DF}(d_{\text{me}})$ , and  $\text{DF}(d_{\text{me}})$  is the deposition fraction at particle mobility diameter  $d_{\text{me}}$ ,  $C_{\text{in}}$  is inhaled concentration by number, mass or surface area,  $t$  is exposure time,  $Q$  is inhaled volume flow.

$\text{DF}(d_{\text{me}})$  was estimated on the basis of the experimentally determined and parameterized function for diesel soot particles (see Appendix C) reported in Rissler et al. (2012).

#### 2.5. Eyes, nose and throat irritation symptoms – self-ratings and medical examination

At every exposure each subject filled in a self-rating sensory symptoms questionnaire before and after the exposure, as well as after 15, 75 and 135 min of exposure. In the questionnaire a visual-analog scoring scale with verbal anchors at the endpoints were used. Replies were coded as ranging from 0 to 100 (Osterberg et al., 2004). Additionally medical examination of signs of eyes, nose and throat irritation including redness and secretion were performed before and after exposure by the same physician.

The generalized estimating equations model (GEE) procedure in SPSS, version 18.0, was used to specify a repeated measures model. All outcomes were compared with FA exposure. Statistical significance refers to  $p < 0.05$  in medical examination. Multiple comparisons (three times) were performed in self-rated eye irritation, statistical significance refers to  $p < 0.017$  according to Bonferroni correction.

### 3. Results and discussions

Summary of exposure characteristics are presented in Table 1. Average  $\text{PM}_{10}$  mass concentration during all DE exposures was  $276 \pm 27 \mu\text{g m}^{-3}$  with particle number concentration of  $3.9 \times 10^5 \pm 0.5 \times 10^5 \text{ cm}^{-3}$ . Average number size distribution of all DE is presented in Fig. 2. The size distribution was unimodal and average CMD during all DE (mean of means) was  $89 \pm 9 \text{ nm}$  with GSD  $1.98 \pm 0.12$ . Used generation set-up and dilution system ensured repeatability of exposure conditions, the details are given in Appendix C. Shape and structure of DEP in dominating size i.e. around 100 nm can be seen from TEM images shown in Fig. 3.

Number concentrations in DE exposure studies (see Appendix A) with idling engine and mass concentration about  $300 \mu\text{g m}^{-3}$  vary between  $9.5 \times 10^5$  and  $4.3 \times 10^6 \text{ cm}^{-3}$  (Barath et al., 2010; Lucking et al., 2008; Mills et al., 2007; Salvi et al., 2000). Average number concentration in our study  $3.9 \times 10^5 \text{ cm}^{-3}$  belongs to the lower range within the quoted studies. Note that reported number concentration depends on lower size detection limit (for measured

**Table 1**

Summary of exposure characteristics given as average values of all averages during DE and FA exposures.

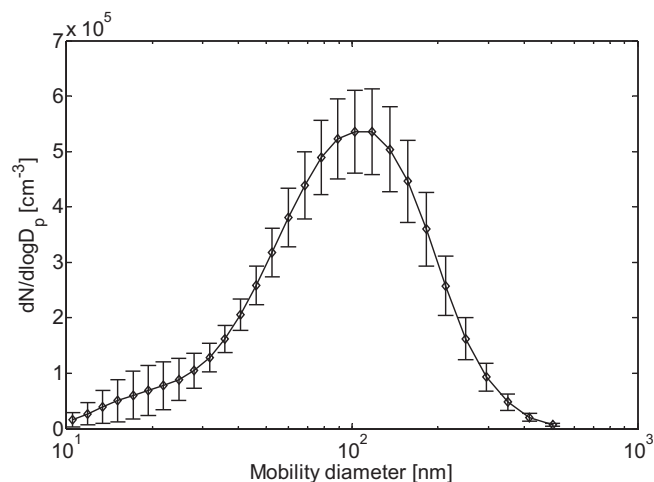
	DE	± Std	FA	± Std	Unit
Particle mass concentration ( $\text{PM}_{10}$ , TEOM)	276	±27	~2 <sup>a</sup>	±2	$\mu\text{g m}^{-3}$
Particle number concentration (10–500 nm, SMPS)	$3.9 \times 10^5$	$\pm 0.5 \times 10^5$	14	±16	$\text{cm}^{-3}$
Count median diameter (CMD)	89	±9	107	±24	nm
Geometric standard deviation	1.98	±0.12	2.30	±0.44	
Particle elemental carbon fraction (EC/TC <sup>b</sup> )	82	±3			%
Particle organic carbon fraction (OC/TC <sup>b</sup> )	18	±3			%
NO	$9.8 \times 10^3$	$\pm 1 \times 10^3$	3	±1	ppb
NO <sub>2</sub>	$1.3 \times 10^3$	$\pm 0.4 \times 10^3$	2	±1	ppb
CO	7.1	±2.2	0.3	±0.2	ppm
CO <sub>2</sub>	$1.9 \times 10^3$	$\pm 0.1 \times 10^3$	950	±70	ppm
VOCs <sup>c</sup>	710	±14			$\mu\text{g m}^{-3}$
Gas phase PAHs <sup>c</sup>	$7.5 \times 10^3$	$\pm 0.2 \times 10^3$			$\text{ng m}^{-3}$
Particle phase PAHs <sup>c</sup>	60	±1.2			$\text{ng m}^{-3}$
Temp	22.7	±0.7	22.7	±0.8	deg C

<sup>a</sup> Estimated value, see Appendix B.

<sup>b</sup> TC is total particulate carbon.

<sup>c</sup> Given as total of measured compounds listed in Table 3.

size ranges see Appendix A). In some studies the particle size resolved information are missing i.e. single or bi-modal size distribution, CMD, GSD, mass median diameter (MMD), which are important for estimation of the fraction of particles deposited in the lungs. Size of the DEP during exposures in our study (CMD 89 nm) was bigger than the size reported for idling engines in Umeå studies for Volvo TD45, 4.5 L (CMD 55 nm) (Barath et al., 2010) and for Volvo TD40, 4.0 L (CMD 75 nm) (Rissler et al., 2012), in case of the latter bi-modal size distribution was observed with a nucleation mode (CMD 16 nm). In our study only a slight trace of a nucleation mode is visible in Fig. 2. Presence of nucleation mode in a study with Volvo TD45, 4.5 L, can be explained by the use of a larger heavy duty engine in comparison to our study (1.9 L, passenger car), resulting in higher exhaust temp and larger emission of lubricating oil, which under dilution conditions favor nucleation of new particles (Maricq, 2007). The same type of a diesel fuel with low sulphur content (<10 ppm) was used in both studies. Presence of nucleation mode reported by Rissler et al. (2012) explains the higher number concentration in comparison to our study at the same mass concentration.



**Fig. 2.** Average number size distribution (mean of means) for all DE exposure with bars denoting standard deviation.

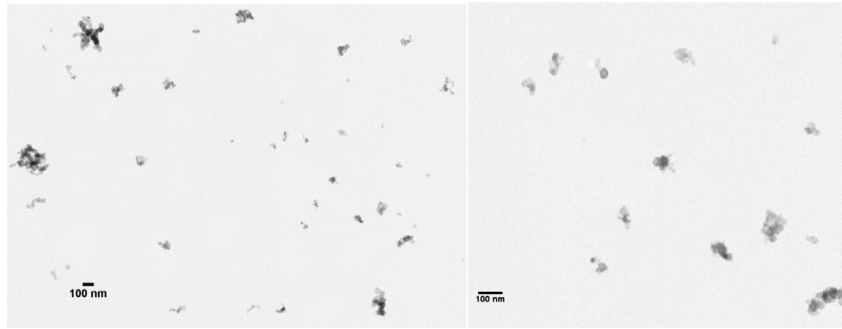


Fig. 3. TEM images of the diesel particles.

Mass concentration of DEP in reported exposure studies, typically ranged between 100 and 300  $\mu\text{g m}^{-3}$  and is reported mainly as  $\text{PM}_{10}$  ( $<10 \mu\text{m}$ ), in some studies as  $\text{PM}_{2.5}$  (Carlsten et al., 2008) or total particulate mass (Sawant et al., 2008). With increasing knowledge on diesel particle characteristics it is known that DEP are smaller than 1  $\mu\text{m}$  (Burtcher, 2005; Maricq, 2007) thus to capture the mass concentration of particles emitted from diesel engines  $\text{PM}_1$  concentrations are appropriate and were used in this study. In our study average mass concentration  $\text{PM}_1$  (TEOM) were comparable to  $\text{PM}_{2.5}$  (gravimetric) within  $\pm 1\%$ .

Fig. 4 shows differences in mass size distribution for two cases: 1) when size dependent effective density determined with DMA–APM system was used and 2) when density 1  $\text{g cm}^{-3}$  (constant throughout whole size range) was assumed. In Table 2 average mass concentration and MMD are given. Differences seen in Fig. 4 illustrate the importance of having information about the size resolved effective density in order to not overestimate the mass concentration. Assumption of particle density 1  $\text{g cm}^{-3}$  resulted in overestimation of average mass concentration in comparison to measured values (TEOM) by 261% (Tables 1 and 2). Whereas average mass concentration calculated with size dependent particle density equals 276  $\mu\text{g m}^{-3}$  and agrees with TEOM measured values.

In Table 2 particle surface area was calculated for three cases as described in section 2.3.3. Despite the recognition of the importance of the surface area of the particles in their toxicity, surface area is not reported in the conducted exposure studies (Hesterberg et al., 2010 and Appendix A). Taking into account agglomerated structure of DEP resulted in 1.7 times higher particle surface area

( $3.5 \times 10^{-4} \text{ cm}^2 \text{ cm}^{-3}$ ) in comparison to assumption of spherical particles ( $2.1 \times 10^{-4} \text{ cm}^2 \text{ cm}^{-3}$ ), whereas use of Maynard's algorithm resulted in estimation of surface area ( $1.0 \times 10^{-4} \text{ cm}^2 \text{ cm}^{-3}$ ) that is about half of that estimated on the basis of spherical particles.

Lung deposited fraction and deposited dose during 3 h of DE exposure by particle number, mass and surface area were calculated and presented in Table 3. For surface area two scenarios were considered i.e. 1) assuming spherical particles, and 2) taking into account surface area of the agglomerated DEP. Lung deposited dose by particle number, mass and surface area in general is not reported in the exposure studies with one exception for one type of engine used in Umeå University studies (Rissler et al., 2012). Assumption of spheres resulted in underestimation by 37% of deposited dose by surface area in comparison to when surface area of agglomerates is accounted for. Deposited dose by mass was, on contrary, overestimated by 132% when unit density was used in comparison to estimations with size dependent density. Apart from described methods, instruments measuring lung deposited particle surface area are also available (namely Nanometer Surface Area Monitor model [NSAM TSI Inc] and portable instrument Partector Nanoparticle Dosimeter [Naneos Particle Solutions gmbh] but their accuracy for agglomerated particles (typically dominating DEP surface area) remains to be investigated.

CO and  $\text{NO}_2$  levels were kept below the Swedish occupational exposure limit values (AFS, 2011:18) given for exhaust fumes as 8 h averages, 20 ppm and 1000 ppb, respectively. Recalculating the 3 h average exposure concentrations of CO (7.1 ppm) and  $\text{NO}_2$  (1300 ppb) to 8h averages, were below the limits. Concentrations of

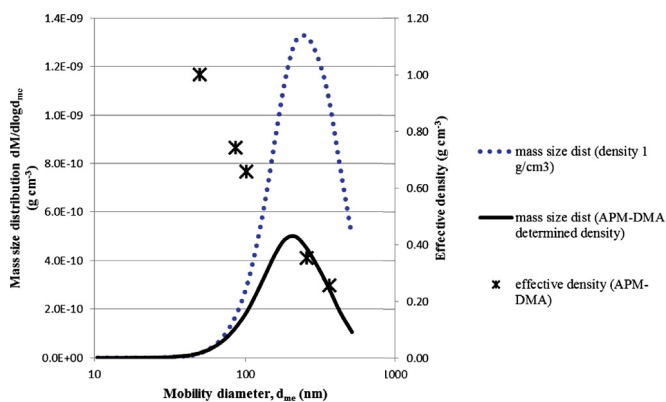


Fig. 4. Differences in mass size distribution when effective density determined with DMA–APM system (size dependent) is used and if density 1  $\text{g cm}^{-3}$  is assumed (constant throughout whole size range). Size dependent effective density as measured by APM–DMA system is also plotted.

Table 2

Particle characteristics calculated on the basis of SMPS measurements (10–500 nm), size dependent density determined from DMA–APM measurements and Rissler et al. (2012) model for surface area estimation of soot aggregates.

	DE	$\pm$ Std	Unit
<i>Particle mass concentration and size distribution</i>			
Assuming density 1 $\text{g cm}^{-3}$			
Mass concentration	719	$\pm 164$	$\mu\text{g m}^{-3}$
Mass median diameter (MMD)	229	$\pm 9$	nm
Geometric standard deviation	1.59	$\pm 0.01$	
Size dependent density (DMA–APM)			
Mass concentration	276	$\pm 56$	$\mu\text{g m}^{-3}$
Mass median diameter (MMD)	195	$\pm 8$	nm
Geometric standard deviation	1.65	$\pm 0.01$	
<i>Particle surface area</i>			
Assuming spherical particles	$2.1 \times 10^{-4}$	$\pm 0.4 \times 10^{-4}$	$\text{cm}^2 \text{ cm}^{-3}$
Aggregates – according to model by Rissler et al. (2012)	$3.5 \times 10^{-4}$	$\pm 0.7 \times 10^{-4}$	$\text{cm}^2 \text{ cm}^{-3}$
According to Maynard's algorithm	$1.0 \times 10^{-4}$		$\text{cm}^2 \text{ cm}^{-3}$

**Table 3**  
Lung deposited fraction and dose during 3 h of DE exposure.

		Deposited fraction	Deposited dose during 3 h exposure
Number		0.45 ± 0.03	$2.8 \times 10^{11} \pm 3.5 \times 10^{10}$ (particles)
Surface area	Agglomerates	0.27 ± 0.01	151.9 ± 27.7 (cm <sup>2</sup> )
	Spheres	0.29 ± 0.01	95.7 ± 16.5 (cm <sup>2</sup> )
Mass	Size dependent density	0.27 ± 0.01	118.5 ± 21.6 (μg)
	Density 1 g cm <sup>-3</sup>	0.24 ± 0.01	275.5 ± 57.2 (μg)

NO<sub>2</sub>, NO and CO in other exposure studies with idling engine and DEP mass concentration about 300 μg m<sup>-3</sup> vary between 0.2 and 1.9 ppm for NO<sub>2</sub>, 0.4–4.5 ppm for NO and 3–27 ppm for CO (see Appendix A). Concentrations in our study belong to higher range within the quoted studies i.e. 1.3 ppm NO<sub>2</sub>, 9.8 ppm NO (higher value than in other studies) and 7.1 ppm CO.

The levels of VOCs and PAHs are presented in Table 4. High levels were found for formaldehyde (400 μg m<sup>-3</sup>), benzene (24 μg m<sup>-3</sup>) and 1,3-butadiene (18 μg m<sup>-3</sup>). The levels in the FA exposures were less than 2% of the DE except for toluene, xylenes, styrene and phenanthrene (about 25%) and for fluorene and anthracene (about 40–50%). Levels of naphthalene and benzo(a)pyrene in our study were comparable with levels found by Sobus et al. (2008) using the same sampling method. Lower levels of both these substances were however found by Sawant et al. (2008). We found relatively higher levels of some of the other particulate PAHs (benzo(a)anthracene, chrysene, benzo(a)fluoranthene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene) compared to levels found by Sobus et al. (2008). We also found somewhat higher levels of benzene and toluene.

**Table 4**  
Mean concentrations of VOCs and PAHs during DE exposures. N denotes number of exposures when measurements were carried out (three samples at each exposure). Mean is given as mean of means during exposures. SD denotes standard deviation.

	N	Mean	SD
<i>VOCs (μg m<sup>-3</sup>)</i>			
Formaldehyde	6	400	97
Acetaldehyde	6	200	63
1,3-Butadiene	7	18	3.5
Benzene	14	24	4.8
Toluene	14	25	5.5
Ethylbenzene	14	5.7	1.5
<i>m,p</i> -Xylene	14	13	4.4
<i>o</i> -Xylene	14	5.3	1.3
Styrene	14	4	0.7
4-Ethyltoluene	14	5.2	1.2
1,3,5-Trimethylbenzene	14	3	0.9
1,2,4-Trimethylbenzene	14	5.9	1.2
1,2,3-Trimethylbenzene	14	2.8	0.5
<i>PAHs (ng m<sup>-3</sup>) gas phase</i>			
Naphthalene	14	7200	1100
Acenaphthalene	14	180	58
Acenaphthene	14	<6	—
Fluorene	14	15	7
Phenanthrene	14	95	23
Anthracene	14	15	12
<i>PAHs (ng m<sup>-3</sup>) particulate phase</i>			
Fluoranthene	14	16	3.1
Pyrene	14	18	4.1
Benzo(a)anthracene	14	2.8	0.4
Chrysene	14	5.7	0.9
Benzo (b)fluoranthene	14	3.6	0.6
Benzo(k)fluoranthene	14	3.6	0.7
Benzo(a)pyrene	14	0.9	0.2
Perylene	14	0.1	0.02
Ideno(1,2,3-c,d)pyrene + dibenzo(a,h)anthracene	14	4	1
Benzo(g,h,i)perylene	14	4.8	1.1

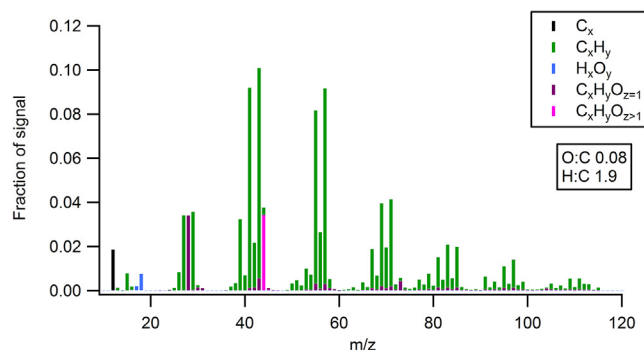
During DE exposures particle EC comprised on average 82% (ranging from 78 to 86%) of the TC, and particle OC fraction accounted for 18% (14–22%) of the TC, see Table 1. Barath et al. (2010) at PM<sub>10</sub> about 300 μg m<sup>-3</sup> reported similar finding during European urban cycle but opposite during idling i.e. EC/TC 5.5% and OC/EC 94.5%.

The high resolution aerosol mass spectrum is dominated by hydrocarbon fragments (Fig. 5). By adding all detected fragments in the mass spectra elemental analysis was performed. The results show that on a molar basis the organic fraction of the DEP consists of 63% hydrogen, 34% carbon and 3% oxygen. The O:C ratio is 0.08 and the H:C ratio is 1.9. Concentrations of nitrogen were negligible in the organic fraction. The total organic matter to total OC ratio (AMS data) was 1.3. Using this ratio, results from the OC/EC analysis and assuming that the particle core is pure elemental carbon the particle organic matter concentration was estimated to be ~60 μg m<sup>-3</sup>. The total concentration of particle PAHs, detected with offline methods (Table 1) is 0.06 μg m<sup>-3</sup>, that is about 0.1% of the total particle organic matter concentration and about 0.02% of PM<sub>1</sub> mass, which is in line with values reported by Ris (2007) who quoted that PAHs and their derivatives comprise <1% of DEP mass.

Information about the remaining major part of the organic mass was obtained from the AMS. This part was dominated by hydrocarbon fragments in the C<sub>x</sub>H(2x + 1) and C<sub>x</sub>H(2x - 1) series (*m/z* = 27, 29, 41, 43, 55, 57 etc). The mass spectrum is similar to the primary organic aerosol in several previous DE emission and tunnel studies (Chirico et al., 2011). The low O:C ratio is very much in contrast to that found in aged oxidized organic aerosol (typically 0.3–0.8) typically dominating in remote regions and downwind urban areas. As DE is exposed to UV light at atmospheric conditions for several hours, the organic coating changes and O:C increases. Thus AMS results show that DEP composition in this study, is representative for primary DEP (fresh emissions) relevant for exposures at occupational settings, at street level and within a city.

Mean changes in eye irritation during DE and FA exposures are presented in Fig. 6. The increment in the eye irritation score was 4.8 in DE exposure versus 0.8 in FA exposure (*p* = 0.046) at 75 min, and 11.5 versus -0.2 (*p* = 0.011) at 135 min.

Irritation in nose, eyes and throat, were also found in the physical examination. Exposure to DE tended to increase the odds of occurrence and/or increase of signs such as redness, secretion and swelling in the eyes (odds ratio [OR] = 3.1, 95% CI 0.97–9.84, *p* = 0.057). There were 19 reports regarding to occurrence and/or increase of such signs in eyes, 11 after DE exposure and 8 after FA exposure. DE exposure increased the risk of occurrence and/or

**Fig. 5.** A high resolution aerosol mass spectrum with division of the organic fragments into five categories dependent on their composition. The categories are fragments containing: only carbon; only carbon and hydrogen; only hydrogen and oxygen; carbon and hydrogen and exactly one oxygen atom; and carbon, hydrogen and more than one oxygen atom.

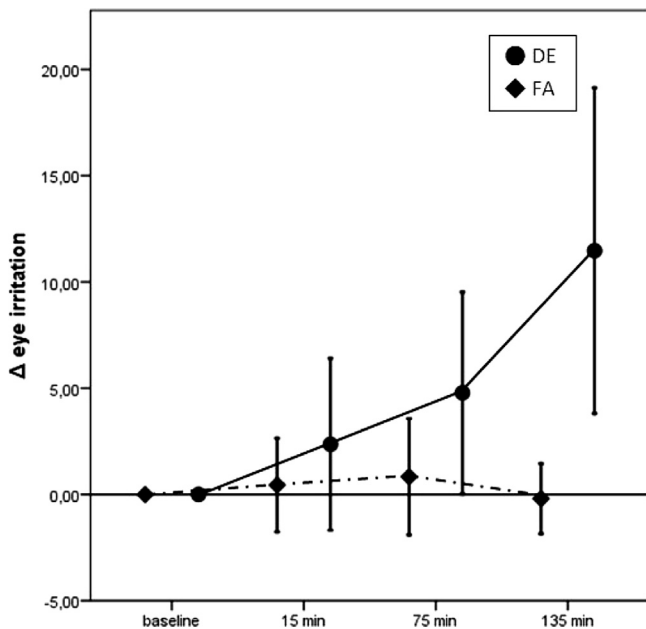


Fig. 6. Changes ( $\Delta$ ) in eye irritation for DE exposure (circles and lines) and FA exposure (diamonds and broken lines). The figure shows the changes (mean and 95% confidence intervals) of before, 15 min, 75 min and 135 min of exposure.

increase of signs such as redness, secretion and swelling in nose (odds ratio [OR] = 6.4, 95% CI 2.47–16.80,  $p < 0.001$ ). There were 26 reports regarding to occurrence and/or increase of such signs in the nose, 20 after DE and 6 after FA exposures. However, there were few reports regarding to occurrence and/or increase of such signs in the throat, one after DE and two after FA exposures. The average formaldehyde level ( $400 \mu\text{g m}^{-3}$ ) during exposures was similar to levels found in one study by Rudell et al. (1994),  $500 \mu\text{g m}^{-3}$ . However Rudell et al. (1999) and Sawant et al. (2008) reported lower levels about 40 and  $60 \mu\text{g m}^{-3}$ , respectively. Slight eye irritation was reported in human volunteers exposed for 4–5 h to about  $300\text{--}400 \mu\text{g m}^{-3}$ , but not  $150 \mu\text{g m}^{-3}$  (Andersen and Mölhave, 1983; Lang et al., 2008). Objective measures (moderately reddened conjunctiva and average blinking frequency) were only affected after 4 h exposure to  $600 \mu\text{g m}^{-3}$  with exposure peaks of  $1.2 \text{ mg m}^{-3}$  (Lang et al., 2008). In our study self-rated irritation of the eyes was higher during DE than FA exposure, with a statistically significant difference after 135 min. The increase was modest; at most 11.5 points on a VAS scale ranging from 0 to 100. However, signs of eye irritation at the medical examination tended to be more pronounced after DE exposure. Increase in nasal irritation and lower respiratory tract symptoms was reported at  $260 \mu\text{g m}^{-3}$  of formaldehyde by Wilhelmsson and Holmstrom (1992). Aldehydes emitted from diesel, in the present study, are the most likely cause of eyes, nose and throat irritation, although exposure to submicron particles may also have contributed.

#### 4. Conclusions

We determined detailed DE characteristics that are important to quantify in chamber exposure studies due to recognized relevancy when linking with health effects.

Comparison of DE characteristics reported in conducted chamber exposure studies (Appendix A) showed that DE characteristics (number concentration, EC/TC, OC/TC,  $\text{NO}_2$ , NO, CO, formaldehyde) vary to a great extent under the same DEP mass concentration and engine load. Thus the same DEP mass concentration and engine

load do not infer the same DE exposure characteristics which highlights the need for detailed and standardized approach for reporting of DE properties.

Scientific evidence on DEP characteristics, which are  $<1 \mu\text{m}$  (Burtcher, 2005), does not justify usage of  $\text{PM}_{10}$  mass concentration, instead  $\text{PM}_1$  or  $\text{PM}_{2.5}$  should be used.

When estimating the lung deposited dose, use of the size dependent effective density (in contrast to assuming unity density) reduced the estimated respiratory dose by 132% by mass.

Three ways of estimating DEP surface area were presented and can be used depending on level of available particle characteristics. Estimated DEP surface area, that accounted for agglomerated structure of DEP, was  $3.5 \times 10^{-4} \text{ cm}^2 \text{ cm}^{-3}$  and was the highest in comparison to two other ways of estimation where less details about DEP characteristics are known. Accounting for agglomerated structure of DEP prevents underestimation of lung deposited dose by surface area by 37% in comparison to assuming spherical particles.

Obtained from AMS measurements, the mass spectra and low O:C ratio (0.08) show that DEP composition in this study, is representative for primary DEP (fresh emissions) relevant for exposures at occupational settings, at street level and within a city.

Adverse nose and eyes irritation symptoms were observed in healthy volunteers only after 135 min. This indicates that observed effects occur at levels that are lower than Swedish occupational exposure limit values (AFS2011:18, 2011) given for exhaust fumes for CO and  $\text{NO}_2$  as indicators for 8 h averages (CO 20 ppm,  $\text{NO}_2$  1 ppm) and American Conference of Governmental Industrial Hygienists (ACGIH, 1996) Threshold Limit Values (respirable particulates  $3 \text{ mg m}^{-3}$  and  $\text{NO}_2$  3 ppm).

Reporting detailed DE characteristics that include DEP properties (such as unified metrics of  $\text{PM}_1$  or  $\text{PM}_{2.5}$  mass concentration, number concentration, size resolved information, surface area, chemical composition, lung deposited dose by number, mass and surface) and detailed gas phase characteristics including components known for their carcinogenic and irritation effect (e.g. aldehydes, benzene, PAHs) can help in mapping and comparison of chamber exposure studies and determination of key DE parameters responsible for observed health effects.

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#### Appendices A, B and C. Supplementary material

Supplementary material related to this article can be found at <http://dx.doi.org/10.1016/j.atmosenv.2013.11.025>.

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