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Size Resolved Particle Number Emission Factors of Motorway Traffic Differentiated between Heavy and Light Duty Vehicles

Carmen Nickel[1†], Heinz Kaminski[1], Bryan Hellack[1], Ulrich Quass[1], Astrid John[1], Otto Klemm[2], Thomas A.J. Kuhlbusch[1,3*]

[1] *IUTA e.V., Air Quality & Sustainable Nanotechnology Unit, Bliersheimer Str. 60, 47229 Duisburg, Germany*
[2] *University of Münster, Climatology Working Group, Robert-Koch-Str. 26, 48149 Münster, Germany*
[3] *CENIDE, Centre for NanoIntegration at the University of Duisburg-Essen, Germany*

ABSTRACT

It is well accepted that airborne particles can induce adverse health effects dependent on the source, composition, morphology and size. Studies indicate that ultrafine particles (diameter < 100 nm) are of specific importance. Therefore, upwind and downwind field measurements of particle number size distributions (14–750 nm), nitrogen oxides, PM_{10} and PM_1 mass concentrations were performed to derive information on sources of those types of particles from motorways. The measurement stations were located at a motorway in a rural area with flat terrain and unhindered air flow situation. The mean particle number concentration was 20,900 #/cm^3 downwind and 3,400 #/cm^3 upwind of the motorway. The highest total particle number concentration at the downwind station was 141,000 #/cm^3. About 90% of these particles were < 100 nm. The measured data were used to derive size-dependent emission factors (EF) using the NO_x tracer method. This method is based on listed NO_x EF (HBEFA, 2010). The average total particle number EF per vehicle was determined to be 3.5×10^{14} particles/km. The average particle EF was 2.1×10^{14} particles/km and 11.8×10^{14} particles/km for light duty vehicles (LDV) and heavy duty vehicles (HDV). The higher EF for HDV is mainly caused by particles with diameters below 50 nm. The comparison of EF from the literature show the importance of the particle size range investigated. Especially particles at the lower size detection limit contribute to total particle number concentrations and hence determine the EF significantly. In the EURO V directive, particle number emission limits of 6×10^{11} particles/km were set for diesel passenger cars. This value is defined for non-volatile particles > 23 nm. The EF for the given size range (> 23 nm) determined in this study were significantly higher with 1.0×10^{14} for LDV.

Keywords: Emission factor; Emission limit; Particle number concentration; Ultrafine particles; Motorway traffic; Light duty vehicle emissions; Heavy duty vehicle emissions.

INTRODUCTION

Epidemiological and toxicological studies have shown associations between adverse health effects and airborne particulate matter (Dockery *et al.*, 1993; Monn and Becker 1999; Oberdörster *et al.*, 2000; Schaumann *et al.*, 2004; Baulig *et al.*, 2009; Peters *et al.*, 2009). Recent improvements in measurement techniques allowed the study and demonstration of correlations between ultrafine particles (UFP, particle diameter d < 100 nm) and adverse health effects (Peters *et al.*, 1997a, b; Donaldson *et al.*, 1998, Oberdörster *et al.*, 2000, 2001; Stölzel *et al.*, 2007). UFP can penetrate to the alveolar region of the lung (Daigle *et al.*, 2003) and, depending on particle characteristics and personal constitution, up to 80% of them may deposit in the respiratory tract (CRP, 1995; Löhndahl *et al.*, 2007). Consequently, the studies of potential effects of UFP have gained increasing interest also linked to their specific properties such as high surface area, high mobility and their potential to reach secondary target organs in the human body (Oberdörster *et al.*, 2002, 2004; Kreyling *et al.*, 2009).

In urban areas, combustion processes, mainly traffic emissions, are a major source of anthropogenic UFPs (Imhof *et al.*, 2005; Tsang *et al.*, 2008; Hagler *et al.*, 2009) while secondary particle formation processes from precursor gases, such as VOC, SO_2, NO_x and NH_3, are the main source of ultrafine particles in rural and remote areas (Birmili and Wiedensohler, 2000). Various epidemiological studies indicate association between adverse health effects and traffic related particles, mainly diesel exhaust emissions (Hoek *et al.*, 2002; Schwartz *et al.*, 2005; Maynard *et al.*,

* Corresponding author. Tel.: +492065418267
E-mail address: tky@iuta.de
† Corresponding author. Tel.: +492065418209
E-mail address: nickel@iuta.de

2007; Sydbom *et al.*, 2001).

Exhaust emissions show mainly bimodal size distributions, with a nucleation and a soot carbon related agglomeration mode. The nucleation mode (< 50 nm) is mainly dominated by volatile organic and sulphur compounds, which are formed during exhaust dilution and cooling, as well as solid carbon and metal compounds (Kittelson, 1998; Chow and Watson, 2007). The nucleation mode shows the highest particle number concentration, whereas the accumulation mode (50–300 nm), dominated by carbonaceous agglomerates, contributes most significantly to the mass concentration. The composition of the exhaust emissions changes in dependence on the fuel, engine technology and exhaust treatment, but also the choice of sampling and measurement techniques have a significant influence on the determined results. Dilution and cooling processes for example determine the amount of volatile materials which adsorb or condense onto existing solid particles or nucleate and form "new" particles (Kittelson, 1998). Accordingly, a comparison between different situations and different techniques is only feasible to a limit extend. This becomes most evident when comparing dynamometer test results in the laboratory sampling under controlled conditions and field measurement in real world situations.

The concern about the potential size dependent effect of particles for adverse health effects stimulated research work characterizing particle number size distribution of exhaust emissions (Giechaskiel *et al.*, 2012). Only a few studies are available calculating particle size dependent emission factors (Birmili *et al.*, 2009, Wang *et al.*, 2010). The determination of size dependent real world EF allows the calculation of particle size dependent traffic emission contributions and its linkage to toxicological and epidemiological studies.

The majority of the studies determined total particle number emission factors for different traffic situations and different vehicle types (HDV and LDV) and were conducted without size dependency. Furthermore, the size ranges as well as the correction techniques used to account for dilution of the aerosol, vary widely. Most studies which calculated number emission factors were conducted in tunnel systems and the tunnel ventilation was used to calculate dilution conditions (Kirchstetter *et al.*, 1999; Gidhagen *et al.*, 2003; Kristensson *et al.*, 2004; Geller *et al.*, 2005; Ban-Weiss *et al.*, 2010). Furthermore, different model systems were applied for the calculation of the dilution of the exhaust emissions (Jamriska and Morawska 2001; Corsmeier *et al.*, 2005; Birmili *et al.*, 2009; Wehner *et al.*, 2009) or the NO_x tracer method was used for the calculation of the dilution, like by Imhof *et al.* (2005) and Wang *et al.* (2010).

In 2011 the European Commission introduced a particle number emission limit of 6×10^{11} particle/km for diesel passenger cars. The EU limit applies to non-volatile particles, therefore, the aerosol is diluted and heated to e.g., 350°C (JRC Report, 2011) in a dynamometer test stand. Additionally, a cut off size of > 23 nm is used to achieve comparability between different instruments and measurement situations. This measuring approach is rather different from ambient air measurements, because the main parts of the exhaust emissions are (volatile) particles in the nucleation mode. Accordingly, these values cannot directly be compared to onsite measurements in ambient conditions without pre-treatment of the aerosol.

This study combines the NO_x tracer method the emission factors of HBEFA 3.1 (2010) and the calculation of size dependent number emission factors (14–750 nm) for light and heavy duty vehicles of a motorway in Germany.

METHOD

Upwind and downwind measurements of the particle number size distribution and further parameter like PM_{10}, PM_1 and NO_x were conducted at the motorway A61 in Germany.

Measurement Station

The measurement period was from 16.12.2006 through 07.01.2007. The monitoring stations were climatized to 20°C and set-up both sides of the A61 motorway, between the interchanges Meckenheim and Bad Neuenahr, Germany (Fig. 1).

The distances between the measuring stations and the first lane of the motorway were 13 m for the side with traffic direction Cologne (north western direction) and 3 m for the traffic side direction Koblenz (south eastern direction), respectively.

The local meteorological parameters wind speed, wind direction, temperature and humidity were recorded according to the VDI guideline 3786 (2004) using a Campbell Scientific meteorological station. Fig. 2 summarises the temperature and the relative humidity for the study period. The average temperature was 4°C (SD 4°C) and relative humidity 94 (SD 7).

The stations were characterized by a flat terrain and unhindered air flow conditions. The main wind direction was from the southwest (Fig. 1), almost perpendicular to the highway. Accordingly, the north-eastern station - direction Cologne - was influenced by traffic during most of the time. For clear upwind and downwind situations (southwest or northeast), the upwind station represented the background concentrations and the downwind station the traffic-influenced concentrations, respectively. The concentration differences between traffic-influenced downwind and upwind (background) stations provide the concentrations attributable to local traffic emissions.

The average vehicle volume combined for both directions was 79,500 vehicles per day during the experimental period. Traffic data was provided by the Federal Highway Research Institute (Bundesanstalt für Straßenwesen, BASt) in nine categories and with one-hour temporal resolution. For this study, the nine vehicle categories were merged into the two classes, Light Duty Vehicles (LDV) and Heavy Duty Vehicles (HDV). LDV was defined as petrol and diesel passenger cars, light commercial vehicles and motorbikes and HDV as trucks with gross weights over 3.5 t (with or without trailers) and busses. The average HDV fraction was 18%, with high variations depending on the time of day and on the day of the week (2% to 78%).

The A61 motorway is mainly frequented by long distance

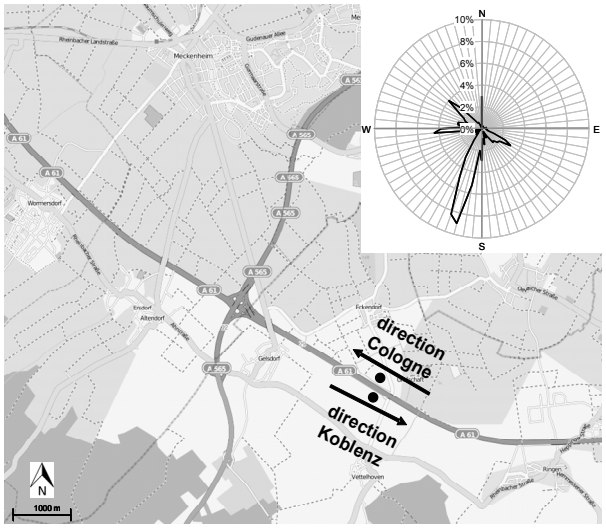

Fig. 1. Location of the measurement sites at the A61, black dots indicates the measuring stations (Source: OpenStreetMap 2010). The wind rose inset of relative frequencies of wind directions visualises the main wind direction during the three weeks measurement period.

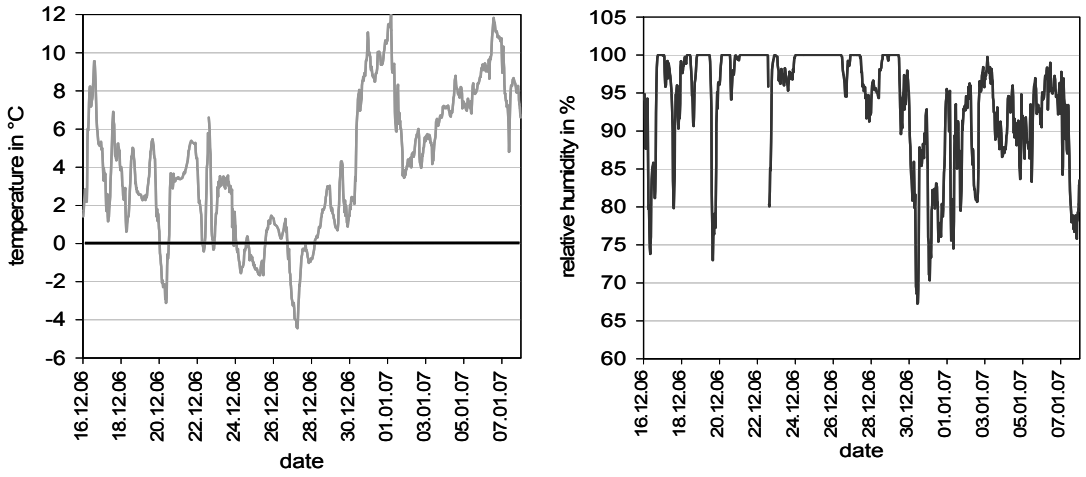

Fig. 2. Ambient temperature (left) and relative humidity (right) during the measuring period.

transport, which is reflected by the high HDV fraction during the weekdays. Note further that HDV traffic is legally limited over weekends to passenger cars, light duty vehicles and urgent transport goods HDV.

The measurements were conducted two weeks before and one week after New Year's Eve leading to 35% lower HDV traffic compared to the annual mean. No differences in traffic volume were observed for LDV.

Instrumentation

The particle number size distributions were measured at both sides of the motorway with scanning mobility particle sizers (TSI SMPS Model 3936) each. The SMPSs were operated with 0.3 L/min aerosol flow and 3 L/min sheath flow to cover the size range from 14 nm through 750 nm electrical mobility particle diameters. The up-scan times were 240 seconds, the retrace times were 20 seconds. Scan intervals started every 6 minutes. All size spectra were corrected for multiple charges of particles, diffusion losses in the instruments, and diffusion losses in the sampling line according to the equation for the transport efficiency in a laminar tube flow (Hinds, 1999). The comparability and size calibration of the SMPSs was checked prior and directly after the measurement campaign and found to be within 5%.

Measurements of NO_x were conducted with a time resolution of 30 min at both sides of the motorway, using a NO_x Monitor Model ML 9811. The PM_1 and PM_{10} mass concentrations were monitored continuously using a Tapered Element Oscillating Microbalance (TEOM) Model R&P 1400 AB, respectively. Periodic maintenance of the instruments and function tests were carried out for quality control.

Data Preparation

For data analysis 30 min average concentration data of particle number size distributions, NO_x, PM_1, and PM_{10} were used. The hourly traffic counts were divided by two to derive 30 min values assuming constant traffic flow. The

experimental data set of 1,104 half-hour averages comprised the whole three week measurement period.

For diurnal analysis, the data measured during the public holidays (25.12.2006 and 26.12.2006, New Year 2007), were marked to be weekend values, which lead to a total of eleven weekend days and twelve workdays. The correlations between particle number, NO_x, PM_1, PM_{10} and the traffic flow were analysed using the difference between the traffic-influenced (downwind) station and the background (upwind) station. These differences are attributed to the local traffic emissions. Only the data taken during clear upwind/downwind and dry meteorological conditions were used. The reduced data set was 31% (= 341 half hour data sets) of the original data set.

The calculated size segregated emission factors for the particle number are based on the NO_x emission factors of the HBEFA 3.1 (2010) assuming the site specific driving conditions and average fleet composition for Germany for 2006. More specifically, the EF_{NOx} for LDV was calculated using the average of diesel and petrol passenger cars, light commercial vehicles and motorbikes with speed of 120 km/h. For HDV, an average speed of 80 km/h, the official speed limit, was employed. The speed chosen for the vehicles were in accordance to those measured.

The emission classes Euro V/5 and VI/6 are included in this new HBEFA 3.1 (2010) version as well as modern exhaust reduction techniques such as soot particle filters for diesel cars and trucks. These values assume a "business as usual" (HBEFA, 2010) traffic situation with free flow conditions for a motorway within a rural background area in Germany. The traffic situation "business as usual" is characterised by a mixture of various vehicle sub-segments like different emission concepts (EURO 1,2,3,4 etc.), within a vehicle category, as a result of the fleet composition model, for the basic year 2006 in Germany. Average NO_x emission factors $EF_{LDV,NOx}$ for light duty vehicles of 1.047 g/km and $EF_{HDV,NOx}$ for heavy duty vehicles of 5.459 g/km were derived based on the information given above.

Calculation of Particle Emission Factors (EF)

With the measured particle size data, emission factors (EF) were calculated for the entire vehicle fleet and, in addition, separate emission factors for heavy duty vehicles (HDV) and light duty vehicles (LDV), with a high resolution of the particle size spectrum. The NO_x-tracer method according to Gehrig *et al.* (2004) was used to determine size dependent emission factors for the different size classes of the 30 min average SMPS data. This approach assumes similar dispersion of particles and gases during their transport from the point of emission (tailpipe exhaust) to the point of measurement. The differences of the particle concentrations downwind minus upwind (ΔN_i) and the traffic counts (n_{fleet}) were used along with the dilution factor (v) to calculate the particle number emission factors for the entire vehicle fleet, $EF_{fleet,\,i}$ in i particle size classes (Eq. (1)).

$$EF_{fleet,\,i} = \frac{\Delta N_i \times v}{n_{fleet}} \quad (1)$$

The $EF_{fleet,\,i}$ for individual size classes i and the entire vehicle fleet are the respective sums of the emission factors for light duty vehicles, $EF_{LDV,\,i}$ and those for heavy duty vehicles, $EF_{HDV,\,i}$. Therefore, Eq. (1) can be re-arranged according to Eq. (2):

$$\Delta N_i = \frac{EF_{fleet,i} \times n_{fleet}}{v} = \frac{EF_{LDV,i} \times n_{LDV} + EF_{HDV,i} \times n_{HDV}}{v} \quad (2)$$

Using a similar approach for NO_x with the known EF (HBEFA, 2010) for LDV and HDV, the dilution factor v is computed according to Eq. (3) for each 30 min time interval:

$$v = \frac{EF_{LDV,\,NOx} \times n_{LDV} + EF_{HDV,\,NOx} \times n_{HDV}}{\Delta NOx} \quad (3)$$

This dilution factor is used in Eq. (1) and in Eqs. (4) and (5). Next, in order to compute the emission factors $EF_{LDV,i}$ and $EF_{HDV,i}$, two data sub-sets, in which either HDV dominated the particle emissions (time periods when the fraction n_{HDV}/n_{fleet} were > 85% - $n_{HDV(highHDV)}$ and $n_{LDV(highHDV)}$ in Eq. (5)), or, alternatively, LDV (n_{LDV}) dominated the emissions, were utilised. For the cases of high LDV contributions, the concentration differences, $\Delta N_{i,high\ LDV}$, can be, in analogy to Eq. (2), interpreted as sum LDV and HDV contributions to the total particle emission (within each size bin i):

$$\Delta N_{i,highLDV} = \frac{EF_{LDV,i} \times n_{LDV(highLDV)} + EF_{HDV,i} \times n_{HDV(highLDV)}}{v} \quad (4)$$

here, $n_{LDV(highLDV)}$ is the fraction of LDV, at time periods when LDV dominates > 85% and $n_{HDV(highLDV)}$ when $n_{HDV/}$ n_{fleet} were < 25%.

Respectively, $\Delta N_{i,highHDV}$ (in #/cm^3) corresponds to situations with large contribution of HDV to total traffic counts. It is interpreted in analogy to Eq. (4), as:

$$\Delta N_{i,highHDV} = \frac{EF_{LDV,i} \times n_{LDV(highHDV)} + EF_{HDV,i} \times n_{HDV(highHDV)}}{v} \quad (5)$$

The Eqs. (4) and (5) contain two unknowns ($EF_{LDV,\,i}$ and $EF_{HDV,\,i}$) for each i which can be calculated analytically. We computed $EF_{LDV,\,i}$ and $EF_{HDV,\,i}$ for each i and each 30 min time interval and employed the averages for each size class i for further analysis.

RESULTS

Data interpretation in this study follows a two step approach. First, correlations between particle number concentration within various size classes, PM_1 and PM_{10} on the one hand, and NO_x, LDV, and HDV on the other hand, are studied. The size resolved particle emission factor for LDV and HDV are then calculated in the second step.

Particle Concentration

The particle size distribution of the downwind (traffic influenced) measurement station exhibited higher number concentration values than those of the upwind (background) station (Fig. 3) throughout all sizes and the entire data set. The median of the total number concentration (TNC) for the upwind and downwind stations are 2,700 #/cm^3 and 15,100 #/cm^3 and the mean value 3,400 #/cm^3 and 20,900 #/cm^3 respectively.

Two modes are present in the particle number size distribution at both sides, the upwind and downwind station. One mode is identified in the size range around 50 nm in diameter, and a second mode around 200 nm. At the downwind station, an additional mode for particles below the lower particle size detection limit of 15 nm is seen.

A comparison of the two size distributions by calculating the ratio of the number size distributions (Fig. 3) identifies two modes which can be attributed to the traffic: a mode below 20 nm particle size and another around 80–100 nm. The former is the one dominating the particle number concentration and the TNC at the downwind site (Fig. 3).

A high correlation between traffic counts and the measured UFP concentration is expected because vehicle emissions were the main source for UFP in the region. A Kendall-Tau-b correlation analysis investigating the correlation between emission related downwind-upwind particle number concentration with traffic counts for LDV and HDV, shows a significant correlation for particles smaller than 500 nm. The correlation of HDV was in all size ranges higher than the correlation with LDV, except particle sizes with a diameter < 20 nm. Here the LDV showed the highest correlation (R) > 0.4. LDV correlation with particles > 30 nm was always smaller than 0.4. The correlation between the particles in the size range 50–200 nm and HDV as well as overall vehicles showed highest correlation > 0.6 and the lowest correlation for LDV (Fig. 4).

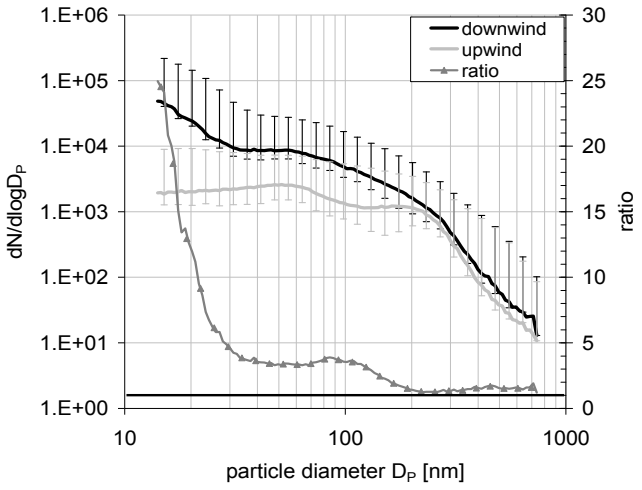

Fig. 3. Median of the particle number size distributions of the downwind station (black) and the upwind station (grey), whiskers = 90 and 10 percentiles, and the corresponding downwind/upwind ratio. The line marks a ratio of one (right y-axis).

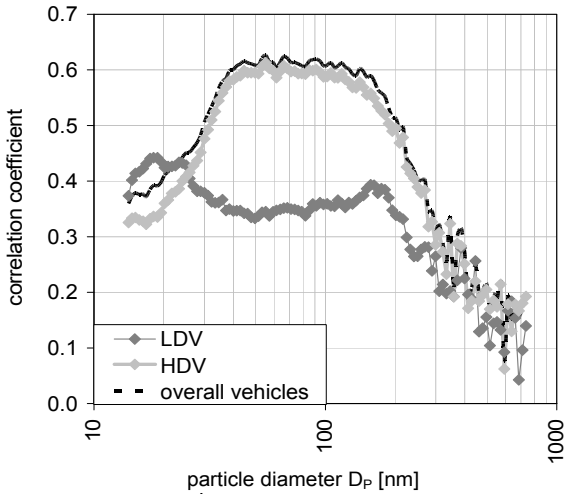

Fig. 4. Kendall-Tau-b correlation analysis of the particle number concentration difference downwind - upwind with the overall vehicles, LDV and HDV density over the entire particle size range. The correlation is significant for the whole size range shown with a level of significance of τ = 0.01; n = 341.

The average measured concentration difference downwind – upwind ΔPM_1 was 5 µg/m^3 and ΔPM_{10} 10 µg/m^3, respectively. A Kendall-Tau-b correlation analysis investigating the size dependent correlation between particle number concentrations and PM-mass concentrations showed no significant correlation (not shown). This result can be explained by the difference in the metric and corresponding atmospheric behaviour. Condensation and coagulation effects (described e.g., by Buonanno *et al.*, 2009; Hagler *et al.*, 2009; Zhu *et al.*, 2002) change the number concentration of UFP but not the mass of e.g., PM_{10}.

In contrast, the various metrics of nitrogen oxides (ΔNO_x, ΔNO and ΔNO_2 – average of the measured ΔNO_x concentration 54 ppb, ΔNO 38 ppb and ΔNO_2 16 ppb, respectively) correlate significantly (at a significance level of τ = 0.01) with the downwind-upwind differences of size-dependent particle concentrations, especially for the two emission modes around 20 nm and around 100 nm particle diameters (Fig. 5).

For particles smaller than 20 nm, a decreasing correlation, mainly for ΔNO_2, was detected. The correlation for particles around 20 nm (first particle mode) shows a slightly better correlation with ΔNO, whereas particles in the size range 50–200 nm (second particle mode) show a better correlation with ΔNO_2 and the highest correlation coefficient (> 0.6). Similar results were found by Wang *et al.* (2010), although they found the minima of correlation coefficients at somewhat larger particles sizes, around 50 nm in diameter.

Separate workday and weekend analyses with the calculation of the contribution of different particle size classes with the traffic counts were conducted (Fig. 6).

All size classes show significant differences between workday and weekend (Mann-Whitney-U-Test; significance level of p = 0.05). The dominant fraction was the UFP with a high contribution of 98% on workdays and a lower

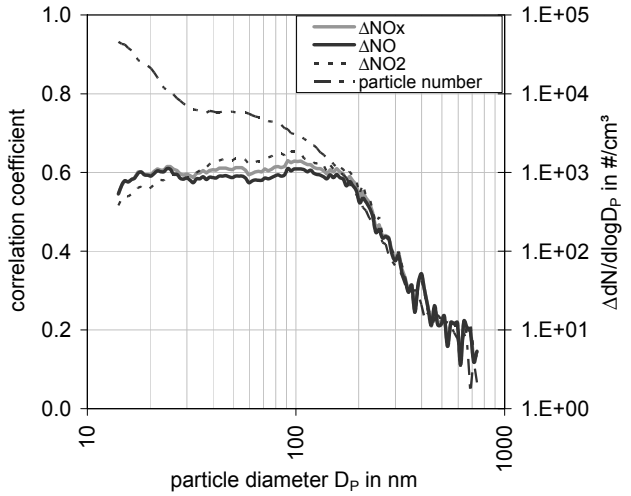

Fig. 5. Kendall-Tau-b correlation of particle diameter and particle number concentration with ΔNO_x, ΔNO and ΔNO_2 over the entire particle size range. The correlation is significant throughout with a level of significance of τ = 0.01. The dotted line shows the median particle number size distribution; n = 341.

contribution of 88% on weekends. Particles between 50–200 nm showed a contribution of 13% on workdays and 21% on weekends. To get more information about the UFP variation, an analysis of the diurnal variation of UFP, HDV and LDV downwind-upwind differences (normalised to the respective maximum values) was conducted (Fig. 6). On workdays, HDV, LDV and UFP exhibit similar patterns, with a steep increase around 0600 hrs and a steady increase of both LDV and HDV traffic until 1700 hrs. The UFP showed the highest values around 1100 hrs, and relatively little variability between 1100 hrs and 1900 hrs. After 1900 hrs, a steep decrease of all parameters occurs. At midday the UFP concentration pattern does not follow the still increasing traffic flow, probably due to better dispersion conditions. On weekends, the UFP concentration and the HDV values show a similar pattern. In contrast, the LDV is similar to that of workdays and does not correlate with HDV values and UFP concentrations. On weekends a steep LDV increase occurs at a later hour compared with workdays, with a maximum at 1400 hrs. A considerable coherence between UFP number concentrations and the HDV traffic volume is apparent from Fig. 6. On Sunday around 2200 hrs, a steep increase of the HDV number counts occurs due to the end of the weekend HDV traffic ban, whereas although the increase in UFP is extenuated due to the mixture of Saturday and Sunday values.

Overall, the differences of the diurnal variations had shown (Fig. 6) that HDV significantly influences the particle number concentrations. Total number concentrations are lower when HDV traffic is lower while this influence is not seen for LDV traffic.

For the diurnal variations, not differentiated between workday and weekend, the HDV and LDV showed a comparable correlation with UFP of τ = 0.53 and τ = 0.58, respectively. For the separate workday and weekend analysis the correlation of LDV with UFP was higher (workdays τ = 0.64 and weekends τ = 0.55) compared to the correlation with HDV and UFP (workdays τ = 0.57 and weekends τ = 0.29).

Emission Factors

Emission factors for various size classes for overall vehicles, and differentiated for HDV and LDV, as well as emission factor size distributions for overall vehicles, HDV and LDV are derived in this section. These are further summarised into size dependent particle number emission factors for four different particle size classes for easier comparison and discussion (Table 1).

NO_x emission factors from HBEFA 3.1 (2010) were used in Eq. (1) and Eq. (3) as described above. The HBEFA NO_x emission factors are based on a modelled mixture of different fuel types and emission concepts of a traffic fleet in Germany for the year 2006. This mixture changes from year to year and depends on the country and traffic situation.

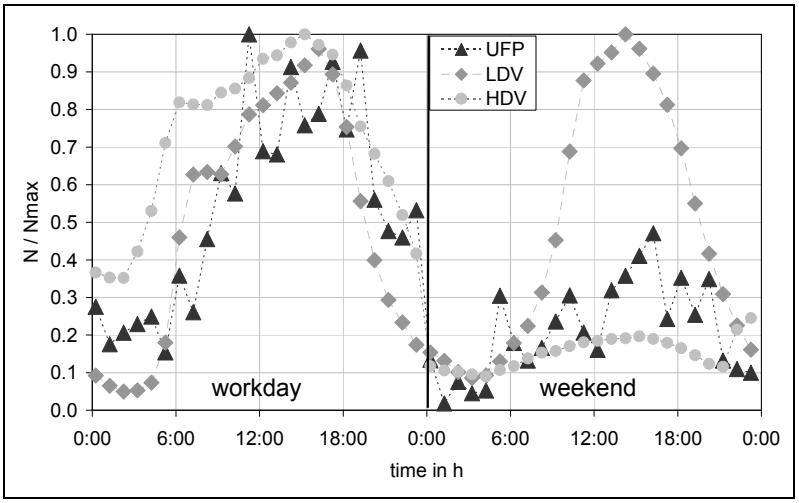

Fig. 6. Diurnal normalised UFP (triangles) concentrations and traffic counts, for workday and weekends – based on one-hour averages, differentiate between light duty vehicles (diamonds) and heavy duty vehicles (circles).

Table 1. Calculated emission factor (particles per kilometer) for various size classes, LDV and HDV. Results for case scenario A (LDV EURO 4 and HDV EURO 5) and scenario B (LDV and HDV EURO 2) are given in brackets.

emission factor [10^{14}/km]	TNC	< 50 nm	UFP	50–200 nm	> 23 nm* (non volatile particles EU limit)
overall vehicles	3.7 (1.5; 6.7)	3.1 (1.3; 5.6)	3.5 (1.5; 6.3)	0.6 (0.2; 1.1)	1.1 (0.5; 1.2)
LDV	2.1 (0.4; 4.7)	1.6 (0.8; 3.7)	1.9 (0.9; 4.4)	0.5 (0.2; 1.0)	1.0 (0.5; 1.0)
HDV	11.8 (4.2; 16.3)	10.9 (3.9; 15.3)	11.4 (4.1; 15.8)	0.7 (0.3; 0.8)	1.4 (0.5; 1.4)

* The EF for the particle > 23 nm was calculated after the size distribution was fitted. Therefore, the size distribution > 40 nm was fitted by using a lognormal function (R^2 0.98–0.99). Afterwards the new size distribution was used for the calculation of the size class > 23 nm.

The assumption that the HBEFA NO_x emission factors employed in this study correspond to real vehicle fleet at the measuring site has an uncertainty significantly influencing the derived particle number emission factors. To estimate the importance of this uncertainty, two scenarios were additionally considered and also presented in Table 1: Case scenario A assuming better than average vehicle composition with all LDV being EURO 4 and HDV Euro 5 vehicles, and the other case scenario B assuming all vehicles, LDV and HDV, being EURO 2 vehicles.

Based on the results of the different size classes (Table 1) it was shown that mainly ultrafine particles (UFP) and especially particles < 50 nm dominate the traffic emissions.

For HDV, UFP account for 96% of TNC and 90% for LDV, respectively. Compared with the emission factor of HDV, the LDV emission factor showed 83% lower values, with the highest differences for UFP and here mainly the size classes < 50 nm. More details of the size dependent emission factors are shown in Fig. 7.

For both traffic fleet fractions, HDV and LDV, the highest emission factor were detected for the smallest particle sizes, close to the lower size range end of the used instruments (< 30 nm). In addition, the ratios between LDV and HDV emissions, ($EF_{LDV,i}/EF_{HDV,i}$) are the largest for the smallest particles.

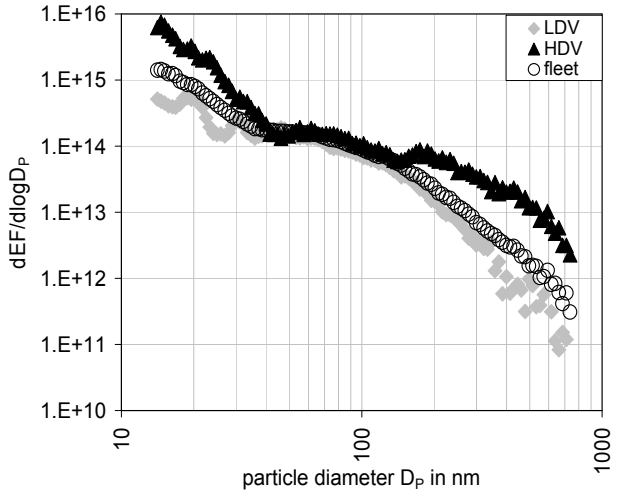

Fig. 7. Emission factor for different particle diameter and fleet (circles), HDV ($EF_{HDV,i,}$ black triangle) and LDV ($EF_{LDV,i}$, grey diamonds).

For particle sizes > 50 nm, the emission factors for LDV and HDV decrease monotonically with increasing particles size. In the size range 50–150 nm the emission factor for HDV and LDV become similar and the differences are virtually zero. Only for HDV another maximum in the size range just around 200 nm is apparent, beyond which the emission factor decreases again with increasing particle size.

DISCUSSION

Particle Number Concentrations

The mean and average measured particle number concentrations were calculated to facilitate the comparison with results of other studies (Table 2).

The main concentration differences between the downwind and the upwind station were detected for UFP and especially particles < 50 nm (factor 9), and minor differences for particles in the size range of 100–200 nm.

Three particle size modes were identified at the downwind station. The first mode is below the lower detectable particle size with the SMPS (< 14 nm), the second mode around 50–70 nm, and the third mode around 200 nm. Comparable modes were found by Jamriska and Morawska (2001), Zhu *et al.* (2002), Rosenbohm *et al.* (2005), Birmili *et al.* (2009) and Wang *et al.* (2010).

Zhu *et al.* (2002) measured the particle size distribution in the size range 6–220 nm at different distances (from 17 m to 300 m) to the motorway Interstate 710 in Downey, California. At a comparable distance of 17 m from the motorway they detected three particle modes, the first mode around 10 nm, the second mode around 20 nm and the third around 70 nm. Zhu *et al.* (2002) also found that with increasing distance from the street the UFP concentration decreased (dilution, coagulation) and the particle modes were displaced to larger particle sizes. They detected up to 90% higher particle concentrations, 200,000 #/cm^3, compared to our study (Table 3). It is conceivable that the different traffic fleet and higher traffic flow (up to 12,200 vehicles per hour with a HDV fraction of 25% as compared to 3,300 vehicles per hour in our study) lead to higher particle concentrations. A comparison of particle number concentrations measured at different highway stations are

Table 2. Median and average of the measured particle concentration at the traffic influenced downwind station and the upwind (background) station.

#/cm^3	TNC		< 50 nm		UFP		50–200 nm	
	down-wind	up-wind	down-wind	up-wind	down-wind	up-wind	down-wind	up-wind
Median	15,100	2,700	11,800	1,300	13,600	2,000	3,100	1,000
Mean	20,900	3,400	17,300	1,900	19,500	2,600	3,500	1,300

Table 3. Comparison of different study results of particle number concentrations as measured at highways.

Measurement range in nm	Particle number concentration in #/cm^3	Measuring site	Ref.	Comments
13.8–749.5	20,900	Motorway A61 Meckenheim	This study	Background station a factor 6 smaller values – measuring period 3 weeks – December-January
10–700	24,900	Motorway Copenhagen	Wang *et al.*, 2010	measuring period 4 weeks – March and April
10–500	28,000	Motorway A100 Berlin	Birmili *et al.*, 2009	measuring period 10 weeks - July-September
10–1,000	25,000	Motorway A656 near Heidelberg	Rosenbohm *et al.*, 2005	background station factor 5 smaller values – measuring period 3 weeks – May
7–960	220,000	Motorway Helsinki	Yli-Tuomia *et al.*, 2005	Direct measurement using a measuring vehicle – measuring period 3 days at the morning and evening rush hour period – October
6–220	200,000	Motorway Downey	Zhu *et al.*, 2002	background station: factor 4 smaller values – measuring period 7 days – April and July
17–890	50,000	Motorway, Queensland	Jamriska and Morawska, 2001	A factor 2 to 6 higher particle concentration at the traffic influenced site compared to the background site – one sunny day

shown in Table 3. Jamriska and Morawska (2001) measured twofold higher average particle concentration at the downwind site compared to our study. This can be explained by the different measuring site, a street canyon type with rock walls with a height of approximately 5 m, and different meteorological conditions. Yli-Tuomi *et al.* (2005) measured an almost fresh aerosol. They used a measuring vehicle to detect the particle concentration close to the exhaust pipe during the drive. The measurements were conducted during the morning and evening rush hour and particle number concentrations around 220,000 #/cm^3, (7 nm–960 nm diameters) were measured. In our study the measured particle concentration at the downwind station show on average about 90% lower values and 70% lower values if only "rush hour" data from our study are considered. It was shown that with increasing distance to the emission source the number of small particles decreased by dilution and agglomeration. This change of distance from the source was also investigated by Hagler *et al.* (2009) showing an 8% drop in UFP concentrations per 10 m distance. Most of the differences in the presented results above can be explained taking all these influencing factors of traffic composition, distance to the point of emission, variance in meteorology and variability in the used measurement equipment into account.

Emission Factors

Only two other studies (Birmili *et al.*, 2009; Wang *et al.*, 2010) conducted similar analyses of emissions factors of HDV and LDV at motorways as conducted in this study. For HDV a steep increase of the emission factor for particles < 50 nm was detected with the highest emission factor at the lowest particle diameter around 15 nm. This increase was also shown but alleviated for LDV. Around 170 nm a second mode for the HDV EF occurred which was assumed to be due to agglomerated soot particles emitted mainly by diesel engines. For LDV the highest values were detected around 20 nm, a second peak at 30 nm. Between 20 nm and 30 nm and for particle sizes > 30 nm the particles reach a comparable value and decreased with increasing particle diameter. The differences between HDV and LDV emission factors were largest for nucleation mode particles (< 50 nm) and for particle sizes > 150 nm. For the size class 50–150 nm, the LDV and HDV emission factors were in the same range. One explanation is that LDV were defined as a mixture of different vehicle categories (passenger cars, light commercial vehicles and motorbikes). Therefore it is conceivable that this could have affected the values of the emission factor since e.g., light commercial vehicles, which are mainly dominated by diesel engines, could have

increased the emission factor in this size range.

For both classes HDV and LDV with increasing particle diameter the emission factor decreased. For the whole vehicle fleet two modes were detected one below 30 nm and another around 100 nm particle diameters shown in Fig. 7. These results are similar to those presented by Birmili *et al.* (2009) and Wang *et al.* (2010). The prior detected a bimodal distribution of emission factors with maximums of the geometric means around 17 nm and 71 nm for the entire vehicle fleet, respectively. Wang *et al.* (2010) also detected a bimodal distribution with one peak at about 10–30 nm and the second at about 70–100 nm.

Further studies determining emission factors for vehicles at motorways did not present any size dependent particle emission factors (Table 4) and the EF are reported for the whole size range determined. Average EF calculated over the whole particle size range in this study resulted in 11.8 $\times 10^{14}$ particle/km for HDV and 2.1 $\times 10^{14}$ particles/km for LDV emission factors. The latter are 83% lower than the prior. Using particle size classes from 30–10,000 nm and 3–3000 nm Corsmeier *et al.* (2005) and Ban-Weiss *et al.* (2010) found comparable results with around 85% and 88% lower EF for LDV compared to the HDV. In other studies much higher differences between LDV and HDV emission factors were detected, ranging from a factor 10 by Imhof *et al.* (2005) up to a factor 149 by Birmili *et al.* (2009).

In detail it becomes evident that the highest emission factors for total number concentrations were determined in studies which used instruments with lower particle size detection limits (smaller than in our study, < 14 nm). If the emission factor for a similar size range as in this study is calculated, based on e.g., data provided by Imhof *et al.* (2005) for the size range 18–300 nm, their emission factor was only a factor two higher. In contrast, Corsmeier *et al.* (2005) detected lower emission factor for both HDV and LDV by using instruments which started at particle sizes > 30 nm. By not capturing the particle concentrations in the 14–30 nm size range, they missed a portion of particles that made up about 88% for HDV and 59% for LDV, respectively, in our data set. In any case, this result indicates that the largest differences between data sets are often caused by differences of the lower size detection limits of the employed instrumentation. It may also be noted that the measurement techniques used in all studies made no difference between volatile and the non-volatile particles as it is done for the EU regulation.

Since 2011 the EU directive Euro V is obligatory for all new vehicles in the EU. In this directive particle number emission limits for diesel passenger cars were established with a limit value of 6 $\times 10^{11}$ particles/km. This value is defined for particles > 23 nm. The EU limit refers to solid/non-volatile particles > 23 nm, after removing volatile particles, by the standardised measurement procedure (dynamometer test system).

With the separation of the volatile particles and the chosen lower particle size detection limit of 23 nm, an important part of the aerosol, possibly relevant for adverse health effects, is neglected (Giechaskiel *et al.*, 2012). Studies indicate that both the solid particle core and/or adsorbed chemical molecules are responsible for adverse health effects (Lovik *et al.*, 1997; Yang *et al.*, 1999; US EPA, 2002). Recent studies have shown that the volatile fraction, like PAHs, might be more important than the non-volatile fraction

Table 4. Comparison of the calculated and specified (from other studies) particle number emission factor (EF) differentiated for LDV and HDV for different traffic situations.

Measurement range in nm	EF LDV [10^{14}]	EF HDV [10^{14}]	method dilution calculation	Ref.	Comments
14–750	2.1	11.8	NO_x	This study	Motorway, Meckenheim Based on HBEFA 3.1
7–3,000	6.9	73	NO_x	Imhof *et al.*, 2005	Motorway (speed limit 120 km/h), Birrhard
7–3,000	3.2	69	NO_x	Imhof *et al.*, 2005	Motorway (100 km/h), Humlikon
10–700	0.8	17.5	NO_x	Wang *et al.*, 2010	Motorway, Copenhagen
10–3,000*	0.4	25	Tunnel ventilation	Kirchstetter *et al.*, 1999	Caldecott Tunnel, San Francisco
3–900	1.1–5.9	58	Tunnel ventilation	Gidhagen *et al.*, 2003	Tunnel, Stockholm
3–900	0.3–4	4–50	Tunnel ventilation	Kristensson *et al.*, 2004	Tunnel (speed of 85 km/h), Stockholm
7–270	2.2	32.3	Tunnel ventilation	Geller *et al.*, 2005	Caldecott Tunnel, San Francisco
3–3,000*	3.9	33	Tunnel ventilation	Ban-Weiss *et al.*, 2010	Motorway tunnel, San Francisco
17–890	1.75[1]	-	box model	Jamriska and Morawska, 2001	No differentiating between LDV and HDV, Brisbane
30–10,000	1.2	7.8	box model	Corsmeier *et al.*, 2005	Rural motorway, Heidelberg
10–500	0.2	29.6	CFD	Birmili *et al.*, 2009	Motorway, Berlin
7–400	0.019	0.9	CFD	Wehner *et al.*, 2009	Motorway - Dif. between gasoline and diesel cars (on road tailpipe measurement), Leipzig

* # * kg/fuel total vehicle fleet.

(Biswas *et al.*, 2009) and different PAHs can easily condense on other particles (Chow and Watson, 2007). In diesel exhaust emissions of HDV around 560–760 μg/m^3 PAH were detected (Tsai *et al.*, 2011). Considering the fact that adverse health effects can be affected by small volatile particles and also by adsorbed volatile compounds on the particle surface, the detection of the ambient aerosol without separation of the volatile particles is very important, though the comparability between different studies and situations is poor. To enable some comparison between the EU limit and this study, the measured size distributions > 40 nm were fitted with a lognormal function (R^2 0.98–0.99). For fitting it was assumed that all measured particles < 23 nm were volatile particles and that a stable soot mode was determining accumulation mode (40–200 nm) for LDV and HDV emissions. Afterwards the EF for LDV and HDV for the size range 23–750 nm were re-calculated.

The differences between the EF for HDV and LDV decreased significantly if only the fitted size distribution and values above 23 nm were considered (LDV only 27% lower than HDV compared to 83%). For LDV an EF of 1.0×10^{14} particles/km and for HDV an EF of 1.4×10^{14} particles/km was derived. The emission factor for LDV is still exceeding the EU emission limit of 6×10^{11} particle/km.

The presented emission factors for LDV and HDV represents the particle size emissions for a typical traffic fleet at a motorway in Germany in 2006/2007. Based on the introduction of different new EU emission concepts for cars or the particle filter for diesel engines it is conceivable that the emissions decreased during the last years. This needs to be verified in further studies.

CONCLUSION

Measurements of particle number concentration and size distributions together with NO_x, PM_{10}, PM_1 and meteorological parameters were conducted at a motorway in Germany in December 2006 and January 2007. Applying the NO_x tracer method and using the NO_x emission factors (EF) of the HBEFA 3.1 (2010), an EF of 11.9×10^{14} particles/km were derived for heavy duty vehicles (HDV) and 2.1×10^{14} particles/km for light duty vehicles (LDV).

It was shown that UFP and mainly particles in the size class < 50 nm dominated the emission of both LDV and HDV. The differences between the HDV and LDV emission factors were largest for particles < 50 nm and > 150 nm. For the size class 50–150 nm, the LDV and HDV emission factors were in the same range.

The presented results are in general agreement with those published in earlier studies from other countries and different traffic situations and experimental approaches. The largest variations between the different studies were found for HDV emission factors. Higher total emission factors compared to this study were found for those studies which use measuring instruments detecting smaller particle sizes (< 14 nm) because the particles at the lower end of the detected size range are the ones with the highest emission factors. Therefore, the size range analyzed is an important factor to be taken into account when comparing total particle emission factors.

According to this, a comparison of total particle emission factors derived from different countries and different traffic fleets as well as measurements with different size classes must be done with great care.

The EURO V directive sets an EU limit value of 6×10^{11} particles/km for diesel passenger cars referring to solid/non volatile particles > 23 nm according to the standard measurement procedure at dynamometer stands. This limit value is hence not directly comparable to field studies because different size ranges as well as all particles, including the volatile fraction are normally measured.

For a better comparability of our data with the EU limit, the measured size distribution > 40 nm were fitted with a lognormal function and the new size distribution was used for the calculation of the particle number concentration for the size range 23–750 nm. The derived emission factors were still higher than the EU emission limit. An EF of 1.0×10^{14} particles/km was calculated for LDV, here representing diesel passenger cars. Comparable or even higher EF maybe expected at highly frequented roads in urban areas with stop and go traffic directly affecting the urban population. The importance of traffic emission in general and the number concentration limit values are both specifically stressed further taking the recommendation of the IARC working group of the WHO in June 2012 into account, which classified diesel engine exhaust as carcinogenic to humans and gasoline engine exhaust as "possibly carcinogenic to humans".

The results from this study indicate that the highest differences of the calculated emission factor were detected with decreasing detection limit of the used instrument. Therefore, size resolved measurements are necessary for a comprehensive interpretation of the results of the calculated emission factor under real world conditions.

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REFERENCES

Ban-Weiss, G.A., Lunden, M.M., Kirchstetter, T.W. and Harley, R.A. (2010). Size-resolved Particle Number and Volume Emission Factors for On-road Gasoline and Diesel Motor Vehicles. *J. Aerosol Sci.* 41: 5–12.

Baulig, A., Singh, S., Marchand, A., Schins, R., Barouki, R., Garlatti, M., Marano, F. and Baeza-Squiban, A. (2009). Role of Paris $PM_{2.5}$ Components in the Pro-inflammatory Response Induced in Airway Epithelial Cells. *Toxicology* 261: 126–135.

Benbrahim-Tallaa, L, Baan, R.A., Grosse, Y., Lauby-Secretan, B., Ghissassi, F.E., Bouvard, V., Guha, N., Loomis, D. and Straif, K. (2012). on Behalf of the International Agency for Research on Cancer Monograph Working Group. Carcinogenicity of diesel-Engine and

Gasoline-engine Exhausts and Some Nitroarenes. *Lancet Oncol.* 13: 663–664.

Birmili, W. and Wiedensohler, A. (2000). New Particle Formation in the Continental Boundary Layer: Meteorological and Gas Phase Parameter Influence. *Geophys. Res. Lett.* 27: 3325–3328.

Birmili, W., Alaviippola, B., Hinneburg, D., Knoth, O., Tuch, T., Borken-Kleefeld, J. and Schacht, A. (2009). Dispersion of Traffic-related Exhaust Particles near the Berlin Urban Motorway – Estimation of Fleet Emission Factors. *Atmos. Chem. Phys.* 9: 2355–2374.

Biswas, S., Verma, V., Schauer, J., Cho, A. and Sioutas, K. (2009). Oxidative Potential of Semi-volatile and Non Volatile Particulate Matter (PM) from Heavy-duty Vehicles Retrofitted with Emission Control Technologies. *Environ. Sci. Technol.* 43: 3905–3912.

Buonanno, G., Lall, A.A. and Stabile, L. (2009). Temporal Size Distribution and Concentration of Particles near a Major Highway. *Atmos. Environ.* 43: 1100–1105.

Chow, J.C. and Watson, J.G. (2007). Review of Measurement Methods and Compositions for Ultrafine Particles. *Aerosol Air Qual. Res.* 7: 121–173.

Corsmeier, U., Imhof, D., Kohler, M., Kuhlwein, J., Kurtenbach, R., Petrea, M., Rosenbohm, E., Vogel, B. and Vogt, U. (2005). Comparison of Measured and Model-calculated Real-world Traffic Emissions. *Atmos. Environ.* 39: 5760–5775.

Daigle, C.C., Chalupa, D.C., Gibb, F.R., Morrow, P.E., Oberdörster, G., Utell, M.J. and Frampton, M.W. (2003). Ultrafine Particle Deposition in Humans during Rest and Exercise. *Inhal. Toxicol.* 15: 539–552.

Dockery, D.W., Pope, C.A., Xu, X., Spengler, J.D., Warem, J.H., Faym, M.E., Ferrism, B.G. and Speizer, F.E. (1993). An Association between Air Pollution and Mortality in Six U.S. Cities. *New Engl. J. Med.* 329:1753–1759.

Donaldson, K., Li, X.Y. and MacNee, W. (1998). Ultrafine (Nanometre) Particle Mediated Lung Injury. *J. Aerosol Sci.* 29: 553–560.

Gehrig, R., Hill, M., Buchmann, B., Imhof, D., Weingartner, E.H. and Baltensperger, U. (2004). Separate Determination of PM_{10} Emission Factors of Road Traffic for Tailpipe Emissions and Emissions from Abrasion and Resuspension Processes. *Int. J. Environ. Pollut.* 22: 312–325.

Geller, M.D., Brata Sardar, S., Phuleria, H., Fine, P.M. and Sioutas, C. (2005). Measurements of Particle Number and Mass Concentration and Size Distributions in a Tunnel Environment. *Environ. Sci. Technol.* 39: 8653–8663.

Giechaskiel, B., Mamakos, A., Andersson, J., Dilara, P., Martini, G., Schindler, W. and Bergmann, A. (2012). Measurement of Automotive Nonvolatile Particle Number Emission within the European Legislative Framework: A Review. *Aerosol Sci. Technol.* 46: 719–749.

Gidhagen, L., Johansson, C., Ström, J., Kristensson, A., Swietlicki, E., Pirjola, L. and Hansson, H.C. (2003). Model Simulation of Ultrafine Particles Inside a Road Tunnel. *Atmos. Environ.* 37: 2023–2036.

Hagler, G.S.W., Baldauf, R.W., Thoma, E.D., Long, T.R., Snow, R.F., Kinsey, J.S., Oudejans, L. and Gullett, B.K. (2009). Ultrafine Particles near a Major Roadway in Raleigh, North Carolina: Downwind Attenuation and Correlation with Traffic-related Pollutants. *Atmos. Environ.* 43: 1229–1234.

HBEFA (2010). Handbook Emission Factors for Road Transport, Version 3.1.

Hinds, W.C. (1999). *Aerosol-Technology*, 2nd Edition, J. Wiley & Sons, New York.

Hoek, G., Brunekreef, B., Goldbohm, Fischer, P. and Brandt, P.A. (2002). Association between Mortality and Indicators of Traffic Related Air Pollution in the Netherlands: A Cohort Study. *Lancet* 360: 1203–1209.

IARC/WHO press release 12 June 2012.

ICRP Publication 66 (1995). Human Respiratory Tract Model for Radiological Protection. *Annals of the ICRP* 24: 1–3.

Imhof, D., Weingartner, E., Ordonez, C., Gehrig, R., Hill, M., Buchmann, B. and Baltensperger, U. (2005). Real World Emission Factors of Fine and Ultrafine Aerosol Particles for Different Traffic Situations in Switzerland. *Environ. Sci. Technol.* 39: 8341–8350.

Jamriska, M. and Morawska, L. (2001). A Model for Determination of Motor Vehicle Emission Factors from On-road Measurements with a Focus on Submicrometer Particles. *Sci. Total Environ.* 264: 241–255.

JRC Report (2011). EUR 24866 EN- 2011. http://publications.jrc.ec.europa.eu/repository/bitstream/111111111/22040/1/jrc_report_particle_emissions_from_a_euro5a_diesel_vehicle.pdf.

Kirchstetter, T.W., Harley, R.A., Kreisberg, N.M., Stolzenburg, M.R. and Hering, S.V. (1999). On-road Measurements of Fine Particle and Nitrogen Oxide Emission from Light- and Heavy-duty Motor Vehicles. *Atmos. Environ.* 33: 2955–2968.

Kittelson, D.B. (1998). Engines and Nanoparticles: A Review. *J. Aerosol Sci.* 29: 575–588.

Kreyling, W.G., Semmler-Behnke, M., Seitz, J., Scymczak, W., Wenk, A., Mayer, P., Takenaka, S. and Oberdörster, G. (2009). Size Dependence of the Translocation of Inhaled Iridium and Carbon Nanoparticle Aggregates from the Lung of Rats to the Blood and Secondary Target Organs. *Inhal. Toxicol.* 21: 55–60.

Kristensson, A., Johansson, Ch., Westerholm, R., Swietlicki, E., Gidhagen, L., Wideqvist, U. and Vesely, V. (2004). Real-world Emission Factors of Gases and Particles Measured in a Road Tunnel in Stockholm, Sweden. *Atmos. Environ.* 38: 657–673.

Löhndal, J., Massling, A., Pagels, J., Swietlicki, E., Vaclavik, E. and Loft, S. (2007). Size-Resolved Respiratory-Tract Deposition of Fine and Ultrafine Hydrophobic and Hygroscopic Aerosol Particles during Rest and Exercise. *Inhal. Toxicol.* 19: 109–116.

Lovik, M., Hogseth, A.K., Gaarder, P.I., Hagemann, R. and Eide, I. (1997). Diesel Exhaust Particles and Carbon Black have Adjuvant Activity on the Local Lymph Node Response and Systemic IgE Production to Ovalbumin. *Toxicology* 121: 165–178.

Maynard, D., Coull, B.A., Gryparis, A. and Schwartz, J. (2007). Mortality Risk Associated with Short Term Exposure to Traffic Particles and Sulfates. *Environ.*

Health Perspect. 115: 751–755.

Monn, C. and Becker, S. (1999). Cytotoxicity and Induction of Proinflammatory Cytokines from Human Monocytes Exposed to Fine ($PM_{2.5}$) and Coarse Particles ($PM_{10-2.5}$) in Outdoor and Indoor Air. *Toxicol. Appl. Pharmacol.* 155: 245–252.

Oberdörster G (2001). Pulmonary Effects of Inhaled Ultrafine Particles. *Int. Arch. Occup. Environ. Health* 74: 1–8.

Oberdörster, G., Finkelstein, J.N., Johnston, C., Gelein, R., Cox, C., Baggs, R. and Elder, A.C.P. (2000). Acute Pulmonary Effects of Ultrafine Particles in Rats and Mice, HEI Research Report Number 96, Health Effect Institute (HEI), Boston, Massachusetts, p. 75–86.

Oberdörster, G., Sharp, Z., Atudorei, V., Elder, A., Gelein, R., Kreyling, W. and Cox, C. (2002). Extra Pulmonary Translocation of Ultrafine Carbon Particles Following Whole-body Inhalation Exposure of Rats. *J. Toxicol. Environ. Health* 65: 1531–1543.

Oberdörster, G., Sharp, Z., Atudorei, V., Elder, A., Gelein, R., Kreyling, W. and Cox, C. (2004). Translocation of Inhaled Ultrafine Particles to the Brain. *Inhal. Toxicol.* 16: 437–445.

Particle Instruments. Model 3936 Scanning Mobility Particle Sizer (SMPS) Spectrometer, Instruction Manual, P/N 1933796, Revision J, TSI Inc., February 2005

Peters, A., Wichmann, H.E., Tuch, T., Heinrich, J. and Heyder, J. (1997a). Comparison of the Number of Ultrafine Particles and the Mass of Fine Particles with Respiratory Symptoms in Asthmatics. *Ann. Occup. Hyg.* 41: 19–23.

Peters, A., Wichmann, H.E., Tuch, T., Heinrich, J. and Heyder, J. (1997b). Respiratory Effects are Associated with the Number of ultrafine particles. *Am. J. Respir. Crit. Care Med.* 155: 1376–1383.

Peters, A., Breitner, S., Cyrys, J., Stölzel, M., Pitz, M., Wölke, G., Heinrich, J., Kreyling, W., Küchenhoff, H. and Wichmann, H.E. (2009). The Influence of Improved Air Quality on Mortality Risks in Erfurt, Germany, Health Effects Institute (HEI) (Ed.), Research report, 137, Boston, Massachusetts, p. 1–112.

Rosenbohm, E., Vogt, R., Scheer, V., Nielsen, O.J., Dreiseidler, A., Baumbach, G., Imhof, D., Baltensperger, U., Fuchs, J. and Jaeschke, W. (2005). Particulate Size Distribution and Mass Measured at a Motorway during the BABII Campaign. *Atmos. Environ.* 39: 5696–5709.

Schaumann, F., Borm, R.J., Herbich, A., Knoch, J., Pitz, M., Schins, R.P., Luettig, B., Hohlfeld, J.M., Heinrich, J. and Krug, N. (2004). Metal-rich Ambient Particles (Particulate Matter 2.5) Cause Airway Inflammation in Healthy Subjects. *Am. J. Respir. Crit. Care Med.* 170: 898–903.

Schwartz, J., Litonjua, A., Suh, H., Verrier, M., Zanobetti, A., Syring, M., Nearing, B., Verrier, R., Stone, P., MacCallum, G., Speizer, F.E. and Gold, D.R. (2005). Traffic Related Pollution and Heart Rate Variability in a Panel of Elderly Subjects. *Thorax* 60: 455–461.

Stölzel, M., Breitner, S., Cyrys, J., Pirz, M., Wölke, G., Kreyling, W., Heinrich, J., Wichmann, H.E. and Peters, A. (2007). Daily Mortality and Particulate Matter in Different Size Classes in Erfurt, Germany. *J. Exposure Sci. Environ. Epidemiol.* 17: 458–467.

Sydbom, A., Blomberg, A., Parnia, R., Stenfors, N., Sabdstrom, T. and Dahlen, S.E. (2001). Health Effects of Diesel Exhaust Emissions. *Eur. Respir. J.* 17: 733–746.

Tsai, Y.I., Yang, H.H., Wang, L.C., Huan, J.L., Young, L.H., Cheng, M.T. and Chiang, P.C. (2011). The Influence of Diesel Particulate Filter Installation on Air Pollution Emissions for Used Vehicles. *Aerosol Air Qual. Res.* 11: 578–583.

Tsang, H., Kwok, R. and Miguel, A.H. (2008). Pedestrian Exposure to Ultrafine Particle in Hong Kong under Heavy Traffic Conditions. *Aerosol Air Qual. Res.* 8: 19–27.

U.S. Environmental Protection Agency (EPA) (2002). Health Assessment Document for Diesel Engine Exhaust. Prepared by the National Center for Environmental Assessment, Washington, DC, for the Office of Transportation and Air Quality; EPA/600/8-90/057F. Available from: National Technical Information Service, Springfield, VA; PB2002-107661, and <http://www.epa.gov/ncea>.

VDI Guideline 3786 (2004). Meteorologische Messungen für Fragen der Luftreinhaltung, Blatt 1 - 5, 7.

Verordnung (EG) Nr. 692/2008 der Kommission vom 18. Juli 2008 zur Durchführung und Änderung der Verordnung (EG) Nr. 715/2007 des Europäischen Parlaments und des Rates über die Typgenehmigung von Kraftfahrzeugen Hinsichtlich der Emissionen von Leichten Personenkraftwagen und Nutzfahrzeugen (Euro 5 und Euro 6) und über den Zugang zu Reparatur- und Wartungsinformationen für Fahrzeuge.

Wang, F., Ketzel, M., Ellermann, T., Wahlin, P., Jensen, S.S., Fang, D. and Massling, A. (2010). Particle Number Particle Mass and NO_x Emission Factors at a Highway and an Urban Street in Copenhagen. *Atmos. Chem. Phys.* 10: 2745–2764.

Wehner, B., Uhrner, U., von Löwis, S., Zallinger, M. and Wiedensohler, A. (2009). Aerosol Number Size Distributions within the Exhaust Plume of a Diesel and a Gasoline Passenger Car under On-road Conditions and Determination of Emission Factors. *Atmos. Environ.* 43: 1235–1245.

Yang, H.M., Barger, M.W., Castranova, V., Ma, J.K., Yang, J.J. and Ma, J.Y. (1999). Effects of Diesel Exhaust Particles (DEP), Carbon Black, and Silica on Macrophage Responses to Lipopolysaccharide: Evidence of DEP Suppression of Macrophage Activity. *J. Toxicol. Environ. Health* 58: 261–278.

Yli-Tuomi, T., Aarino, P., Pirjola, L., Mäkelä, T., Hillamo, R. and Jantunen, M. (2005). Emission of Fine Particles, NO_x, and CO from On-road Vehicles in Finland. *Atmos. Environ.* 39: 6696–6706.

Zhu, Y., Hinds, W.C., Kim, S., Shen, S. and Sioutas, C. (2002). Study of Ultrafine Particles near a Major Highway with Heavy-duty Diesel Traffic. *Atmos. Environ.* 36: 4323–4335.

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