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Vertical distribution of aerosol particles and NO_x close to a motorway

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Abstract

In May 2001, the large-scale field project BAB II was performed at the highly frequented motorway BAB (Bundesautobahn) A656 with two traffic lanes in each direction between the German cities Heidelberg and Mannheim. Extensive measurements of air pollutants were carried out on both sides of the motorway. In a distance of 60 m (north side) and 84 m (south side) to the traffic lanes, two 52-m-high towers were installed, at which electrically powered elevators were fixed. In these elevators, two NO_x analysers, an Electrical Low Pressure Impactor (ELPI; measurement of the particle number size distribution in the diameter range D between 30 nm and 10 μm) and a Diffusion Charger (DC; measuring the particle surface area concentration), were operated to record continuous vertical profiles from 5 to 50 m above the earth's surface. On the upwind side, particle number and surface area concentration as well as NO_x values were constant over the entire height profile. On the downwind side, increased concentrations appeared in the near-ground range: in the forenoon, a monotonous decrease in pollutant concentrations with increasing height was found, while around noon the concentration maximum of the particles was slightly shifted to 10 m above ground. This height dependence was found for two different size ranges, i.e., for particles with $D < 300$ nm (consisting of soot particles and nucleation mode particles formed by condensation as a result of cooling of the exhaust gas after emission), and for coarse particles ($D > 1$ μm, abrasion and resuspension products). In the size range between 300 and 700 nm, no height dependence was found, corroborating the fact that motor traffic emits only few particles in this size range. On the downwind side of the motorway, only background concentrations were measured above 25 m. The results of the profile measurements were confirmed by stationary measurements of particle size distributions with Scanning Mobility Particle Sizers (SMPS) at various heights. A good correlation between particle surface area and NO_x concentration was

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observed. Vehicle emission factors were determined for the particle surface area, number and volume of several size ranges.

Keywords: Vertical profile; Particle number size distribution; Surface area; Traffic emissions; Emission factor; LDV/HDV

1. Introduction

Since epidemiology studies indicated that increased concentrations of atmospheric aerosol particles cause adverse health effects in humans (Dockery et al., 1993; Peters et al., 1997; Donaldson et al., 1998; Pope et al., 2002), the respirable fraction of airborne particles with aerodynamic diameters $D<10\,\mu m$ (PM10) and their sources and sinks have received special attention. Emissions of motor vehicles are an important source of atmospheric particle concentrations (Weingartner et al., 1997; Kittelson et al., 2000; Harrison et al., 2001; Shi et al., 2001; Wehner et al., 2002). While spatial and temporal variations of aerosol number and mass concentration are well investigated for many different urban and rural sampling sites at the standard measuring height of about 2–5 m above ground level (Bukowiecki et al., 2002; Röösli et al., 2001), only very few investigations of the vertical distribution of particulate matter exist up to now. The available studies dealt primarily with the vertical profile of various PM concentrations in cities and in their street canyons with the aim to define consequences on air quality in adjacent buildings (Morawska et al., 1999) and to perform model calculations of the dispersion of pollutants in those street canyons (Colls and Micallef, 1999). Morawska et al. (1999) found no significant height dependence of the particle number concentration (3rd to 25th floor of an office building) in a distance of 80 m from the motorway in Brisbane, Australia. They concluded that the air pollutant concentration level in the close-up range of a high-rise building, which is not located directly at a motorway, is a mixture of fresh emissions by local turbulence and an aged aerosol from traffic emissions and other urban emission sources. Wu et al. (2002) observed a clear height dependence of the mass concentrations PM10, PM2.5 and PM1 measured in six different heights in Macao, China. Compared to the sampling station at road level, the different PM concentrations decreased rapidly up to 8 m above ground, whereas in the upper height range between 8 and 79 m above street level, only a slight concentration decline with increasing height was found. Väkevä et al. (1999) likewise detected significant differences in the particle number concentration between street level and rooftop (25 m) in Lahti, Finland. Another study compared mass and a large number of chemical components of the PM1 fraction at 3.5 and 20 m height at an urban site in Helsinki (Pakkanen et al., 2003).

A vertically highly resolved case study on the distribution of particulate air pollutants in the planetary boundary layer under undisturbed conditions in the vicinity of a motorway is not available until now. To investigate horizontal as well as vertical profiles of motorway traffic emissions, the field campaign BAB II (BAB = Bundesautobahn) was carried out in Germany with the participation of a number of various European research groups. The major goals of the project were the evaluation of traffic emission rates for NO_x, VOC and particles and the comparison of measured and modelled emissions. By the comprehensive set of instruments, further information should be obtained about the following (Corsmeier et al., 2005a):

- diurnal variation in NO_x, VOC and particle concentration in various distances from the motorway,
- concentration gradients in horizontal and vertical direction,
- vertical dimension of the exhaust plume as a function of the atmospheric stratification and the distance to the motorway.

This paper focuses on vertical profiles of particulate air pollutants and NO_x concentrations under upwind and downwind conditions with special consideration of the vertical dimension of the exhaust plume in surroundings without obstacles for approaching airflow.

2. Experimental techniques and procedures

2.1. Measurement site

The field campaign BAB II was conducted on both sides of the motorway A 656 between Mannheim and Heidelberg in the northern part of the flat Upper Rhine Valley in May 2001. A detailed description of the location is given in Corsmeier et al. (2005a). The investigated part of the motorway with two lanes in each direction is situated on a 1.0–1.5 m high embankment, surrounded by land of agricultural use without trees or buildings. The environment is flat and thus does not create restrictions to the approaching air flow. No

other appreciable emission sources of the investigated air pollutants were in the surroundings. The motorway shows a very small slope (0.1%). The traffic frequency is 55,000 vehicles per day, with mainly local traffic between the two cities rather than long-distance traffic; therefore, the fraction of heavy-duty vehicles (HDV) is considerably lower (6%) than on other German motorways. Traffic speed is limited to 120 km h^{-1}.

2.2. Instrumentation

The set-up of all instruments is described in detail in the general survey of the experimental design for the BAB II project (Corsmeier et al., 2005a). In the following, only the instrumentation relevant for this paper is described. The main components of the field experiment on each side of the motorway were two towers (52 m height), on which instruments were fixed at various heights to measure particles, gases and the meteorological parameters. One tower was installed in a distance of 84 m south of the motorway, the other was constructed 60 m from the motorway in the north (see Fig. 1). Further information about the vertical distribution of particles, NO_x and ozone was obtained by profile measurements during several special observation periods (SOPs). For this purpose, electrically powered elevators were installed at the two towers. On the elevator at the northern tower, which was arranged to be on the downwind side for a specific SOP, an Electrical Low Pressure Impactor (ELPI; Outdoor Air Electric Low Pressure Impactor, Dekati Ltd., Finland; no back-up filter stage used) and a Diffusion Charger (DC; Diffusion Charging Particle Sensor Type LQ 1-DC, Matter Engineering, Switzerland) were operated. The ELPI measures the size distribution of airborne particles in the size range of 0.03–10 μm with a potential time resolution of 1 s. The principle is based on particle charging, followed by deposition on impactor plates due to the aerodynamic inertia in a 12-stage impactor, with electrical detection (Keskinen et al., 1992; Baltensperger et al., 2001). For this measurement, aluminium foils coated with Apiezon-L vacuum grease were used to prevent particle bouncing. With the DC, the active surface area of particles is measured, again with a potential time resolution of 1 s (Baltensperger et al., 2001).

In addition, two NO_x analysers were located on the elevator: one (Bendix) for the determination of NO and NO_x using chemiluminescence with ozone and one LMA4 (Scintrex) for the measurement of NO_2, which is based on the detection of the chemiluminescence produced during the reaction of NO_2 with Luminol (liquid chemiluminescence). The NO_x analysers were operated with a time resolution of 3 s. To get NO_x profiles, the corrected NO signal (see Section 3.4) of the chemiluminescence and the NO_2 signal of the Luminox analyser were added. This procedure had two reasons: first, the NO_x analyser operates on a single channel system; NO and NO_x is measured alternately, whereby in each case the other channel is kept on the last measured value. When recording a vertical profile, where a high temporal resolution is necessary, this gives rise to a measuring artefact resulting in an unnaturally angular curve structure. Second, it is known that the chemiluminescence analysers with molybdenum

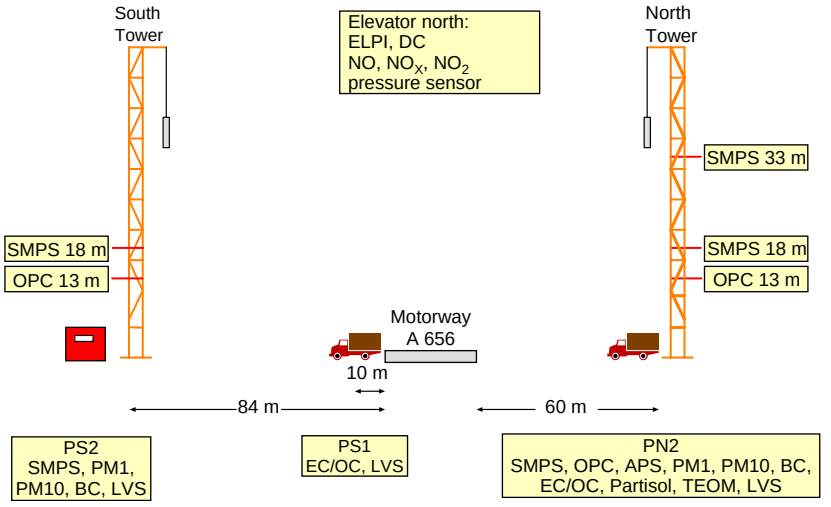

Fig. 1. Selection of instrumentation at the BAB II site (adapted from Corsmeier et al. (2005a)). The trucks and the box represent ground-level measurement points equipped with the instruments in the boxes underneath. OPC: Optical Particle Counter; APS: Aerodynamic Particle Sizer; BC: Black Carbon; EC/OC: elemental and organic carbon; TEOM: Tapered Element Oscillating Microbalance; LVS: Low Volume Sampler.

converters also detect higher oxidized nitrogen components than NO_2, resulting in a positive bias of these NO_2 measurements.

Profiles were taken from a height range between 5 and 50 m above ground, where the tower base determined the lower height limit. The elevator speed was about $0.15\,m\,s^{-1}$, and therefore each profile run took about 5 min.

At three different heights (ground level, 18 and 33 m above the earth's surface) on the northern tower, the downwind side for the prevailing wind direction in the Rhine Valley, and at two different heights (ground level and 18 m above ground) on the southern tower, Scanning Mobility Particle Sizers (SMPS, TSI Inc.) were operated to determine the number size distribution of particles in the size range of about 10–400 nm. The SMPS systems on the northern tower (results discussed in this paper) included a Differential Mobility Analyser (DMA) 3080 and a Condensation Particle Counter (CPC) 3025 at ground level, as well as a DMA 3071A and a CPC 3010 18 and 33 m above ground; all operated with an air flow rate of $0.6\,l\,min^{-1}$. The up scan time was 180 s; multiple charge correction was not applied. These three SMPS systems were running permanently throughout the entire measurement period in May 2001.

Measurements of the meteorological parameters, i.e. wind, temperature and humidity, were performed at six different heights of both towers. Pressure sensors were installed on the elevators to measure the actual pressure, from which the effective height of the elevator was determined. Traffic census was performed including speed measurements of cars. Highly resolved video observations during the SOPs allowed to evaluate licence plate numbers for determining type of cars, engine type, cubic capacity and registration date.

SOPs with profile measurements were performed during favourable weather conditions (i.e., approaching air flow perpendicular to the motorway and no precipitation).

Convenient time periods for detailed evaluation were determined when the conditions for the estimation of emissions using downwind and upwind concentration differences were well fulfilled. Criteria were wind from sectors 0–90° or 180–270°, CO mixing ratio differences >20 ppb between the lowest and the uppermost sampling level at the downwind side, NO_x differences >6 ppb and a traffic density >1000 vehicles per hour (Corsmeier et al., 2005a).

2.3. Quality assurance

Parallel measurements were performed before and after the field campaign with all instruments used for particle number size distributions (SMPS, ELPI, OPC). The comparison was conducted by measuring ambient air concentrations at ground level at the same location where the field experiment took place. Concerning the five SMPS systems, the SMPS located at 18 m above ground on the northern tower was taken as reference instrument. By calculating a size-dependent correction factor for the other SMPS systems, all spectra were corrected based on this calibration. These correction factors were found to be around 5–30% in the diameter size ranges $20<D<400$ nm. In order to facilitate a comparison between the size distributions obtained by the different instruments, correction factors were computed for ELPI and OPC as well (for details see Baumbach and Vogt (2005)).

Quality assurance of the two NO_x analysers was conducted by calibration with test gas of a well-known concentration and parallel measurements with ambient air concentrations (Baumbach and Vogt, 2005).

3. Results and discussion

All results shown in this chapter were obtained in the fourth SOP lasting from 13 May 2001, 20:00, to 14 May 2001, 18:00 Central European Summer Time (CEST). This SOP was particularly favourable because of a change in the wind direction of 180° in the course of the time period so that the instruments were first on the upwind and afterwards during the day on the downwind side of the motorway. Because of the different meteorological conditions during this SOP, three time periods were selected and further investigated:

- Upwind: 13.05.01, from 22:55 to 23:40 CEST,
- Downwind: 14.05.01, from 07:00 to 10:00 CEST,
- Downwind: 14.05.01, from 11:00 to 14:30 CEST.

The downwind period was divided into two parts with respect to the different ground temperatures and solar irradiation as well as the associated thermal effects which cause a rising of the air masses near the ground and consequently an altered level of the vehicle exhaust gas plume. A detailed description of the meteorological conditions at different heights during the analysed time periods is given in Table 1, while the traffic data are reported in Table 2.

3.1. Vertical profile of the particle active surface area concentration

The temporal variation of the particle active surface area concentration measured by the DC on 14 May 2001 is illustrated in Fig. 2. The arrows indicate wind direction and speed recorded in four different heights. During southern and southwestern approaching airflow, the devices were located on the downwind side of the

Table 1
Meteorological conditions during the analysed time periods

Number of analysed profile measurements	Upwind 22:50–23:30 8		Downwind 07:00–10:00 32		Downwind 11:00–14:30 40	
	Mean	Std. dev.	Mean	Std. dev.	Mean	Std. dev.
Wind speed (6 m) ($m\,s^{-1}$)	2.7	0.3	2.2	0.4	2.6	0.9
Wind speed (48 m) ($m\,s^{-1}$)	6.0	0.5	3.1	0.7	3.6	1.2
Wind direction (6 m) (deg)	106.3	7.3	167.6	19.2	197.0	28.9
Wind direction (52 m) (deg)	95.5	4.4	189.5	11.1	208.1	20.7
Temperature (6 m) (°C)	21.0	0.2	18.3	0.9	22.6	1.2
Relative humidity (6 m) (%)	34.5	0.5	62.9	1.4	54.6	5.2
Specific humidity (6 m) ($g\,kg^{-1}$)	5.4	0.0	8.3	0.6	9.4	0.3
Global radiation ($W\,m^{-2}$)	−0.8	0.2	290.6	153.7	683.4	141.2

Table 2
Traffic counts during the analysed time periods (in vehicles per hour)

	Upwind (23:00–24:00)		Downwind (07:00–10:00)		Downwind (11:00–15:00)	
	Counts h^{-1}	Fraction (%)	Counts h^{-1}	Fraction (%)	Counts h^{-1}	Fraction (%)
Total	839	100.0	4695	100.0	3320	100.0
LDV[a]	812	96.8	4379	93.3	2981	89.8
HDV[b]	12	1.4	284	6.0	310	9.3
Motorcycles	15	1.8	32	0.7	29	0.9

[a]Light-duty vehicles (passenger cars and delivery vans).
[b]Heavy-duty vehicles (including busses).

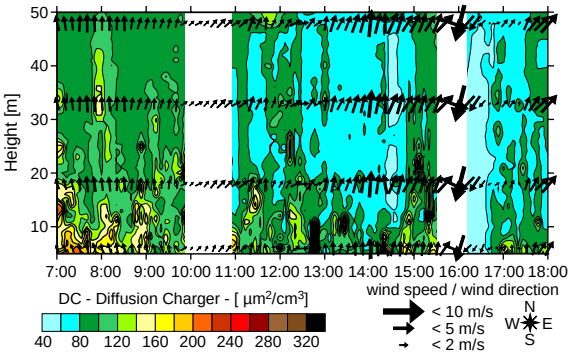

Fig. 2. Isopleth diagram for 14 May from 07:00 to 18:00 CEST for the particle surface area concentration, determined by profile measurements with the DC, combined with wind arrows obtained by measurements at a height of 6, 18, 33 and 48 m above ground level. White areas represent calibration periods.

motorway, and with wind from northern directions they were on the upwind side. As shown in Fig. 2, one recognizes the low and over the entire height homogeneously distributed concentration during the upwind situation after 16:00 CEST. In the remaining time, the DC detected the exhaust gas plume of the motorway traffic, characterized by a high inhomogeneity. Near the ground the highest concentrations were measured, while peaks occasionally reached heights close to the upper limit of the measurements. The variability of the plume strongly depended on the current wind condition, the atmospheric layering and the traffic composition.

Fig. 3 depicts mean vertical profiles (together with single profile measurements) of the surface area concentration for the three chosen time periods. All profiles start at 5 m above ground at the height of the tower base. During a time period of 45 min in the night with constant wind direction and speed, almost no vertical changes in the particle surface area concentration were found at the upwind side (average: $38\,\mu m^2\,cm^{-3}$), even though it was not always a clear upwind situation.

In the morning with high traffic intensities, a high variability of the single profile runs on the downwind side can be seen especially near the ground, but occasionally also at a height of more than 25 m above the earth's surface. Note that the measured background concentration was about $95\,\mu m^2\,cm^{-3}$, i.e., about 2.5 times higher than during the night, stressing the

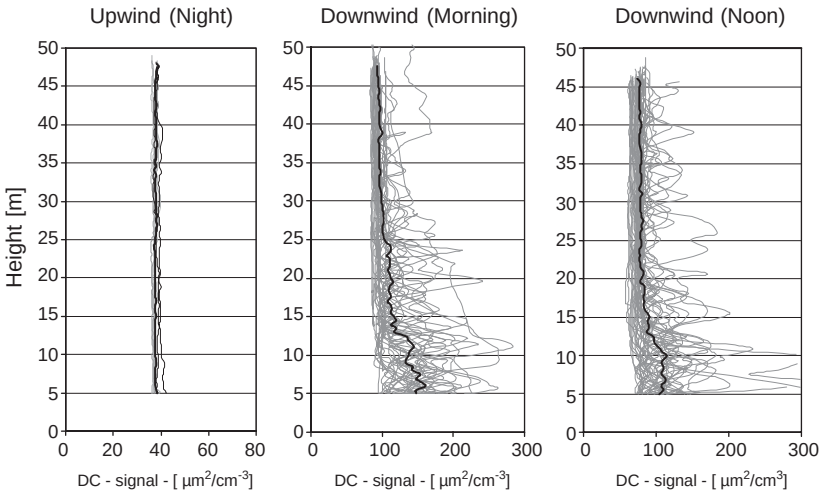

Fig. 3. Vertical profiles of the particles active surface area (DC signal) on the upwind side (23:00–24:00 CEST) of the motorway, as well as on the downwind side in the morning (07:00–10:00 CEST) and around noon (11:00–14:30 CEST). Thick black line: average of all profile measurements in the respective period; dotted grey lines: single profile runs.

variability of the background and the necessity to measure this background on a continuous basis. As a result of the particle emissions of the heavy morning traffic, which culminated between 07:00 and 08:00 on that day, the highest surface area concentrations were found near the ground. With increasing height the mean concentrations decreased more or less continuously.

Around noon, the average vertical profile looks slightly different: the maximum concentration appears to be slightly shifted to a height of about 10 m. Above this level, the surface area concentration decreases and reaches the background value at a height of about 20 m above the earth's surface. Averaging over a longer time period results in quite a smooth curve in the upper height range.

3.2. Vertical profile of particle size distributions

A highly time-resolved pattern of the particle number size distribution was determined by the ELPI. The results are shown as contour plots of the average number size distributions as a function of the height in Fig. 4. For the calculation of the averages, the same profiles were used as for the DC data provided in Fig. 3. On the upwind side of the motorway, the particle sizes in the entire height range showed nearly identical number concentrations in the night. As expected, the highest number concentration of particles was measured in the ultrafine size range ($D < 100$ nm).

On the downwind side of the motorway during the day, the average size distribution profiles showed a high similarity to the mean surface area concentration profiles. With the morning traffic, the concentrations increased very rapidly near the ground, and the highest concentrations were found in the level between 5 and 10 m above the ground. Around noon, a slight increase was observed between 5 and 10 m in the particle size range with $D < 50$ nm, as was already seen in the DC profiles. It is interesting to note that both, the smallest ($D < 300$ nm) as well as the largest particles ($D > 1$ µm), show a profile very similar to the DC signal. Both size ranges can be attributed to the same source, i.e. traffic emissions. The particles with $D < 300$ nm stem from the exhaust pipe, while the coarse mode particles ($D > 1$ µm) are formed by tyre and brake abrasion and resuspension of soil dust. In contrast, nearly no height dependence was observed in the intermediate size range ($D = 300-700$ nm). This can be explained by the fact that car traffic causes only a small number density in this size range.

At this stage, it should be mentioned that it is a known problem of the ELPI that ultrafine particles can deposit by diffusion on the coarse particle stages and cause a false electrometer signal in the size range between 3 and 10 µm (Virtanen et al., 2001). Although there exists a correction algorithm in the ELPI software, it does not seem to correct the error accurately. In laboratory experiments, we put a PM1 impactor in front of the ELPI inlet system to remove large particles. The results showed nevertheless an interrelationship between the increasing number concentrations of ultrafine particles and coarse mode particles. However, the different course of the number concentrations of fine and coarse mode particles in a height level of about 10 m during noon argues only for a weak correlation. Therefore, no correction concerning this fact was applied for the ELPI data.

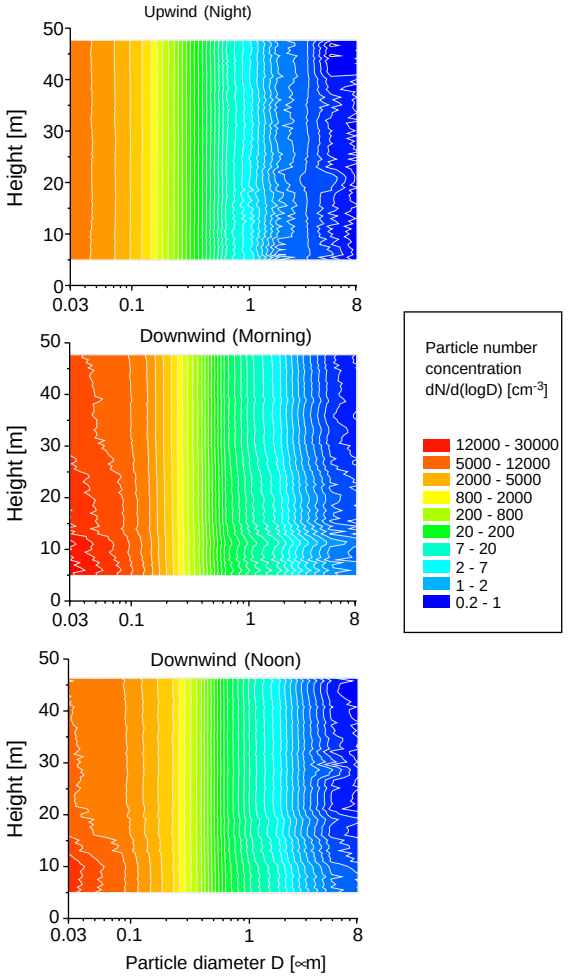

Fig. 4. Vertical profiles of mean particle size concentrations (ELPI) on the upwind side (23:00–24:00 CEST) of the motorway, as well as on the downwind side in the morning (07:00–10:00 CEST) and around noon (11:00–14:30 CEST).

3.3. Size distribution of particles with $D < 300$ nm

The number size distributions measured by SMPS systems at three different heights are presented in Fig. 5 for one upwind and two downwind situations. All size distributions show a typical bimodal structure with a concentration maximum in the particle size range of 20–30 nm, followed by a secondary peak around 100 nm. The highest variation between nighttime and daytime concentration was measured by the SMPS near the ground, whereas the instrument at a height of 33 m above the earth's surface registered comparatively little variations. Particles with $D < 50$ nm are formed by supersaturation of semivolatile species due to a rapid cooling of traffic exhaust gas resulting in a homogeneous nucleation (Bukowiecki et al., 2002; Kittelson et al., 2000). They are termed as primary nucleation mode particles, in contrast to secondary nucleation mode particles formed after atmospheric oxidation processes in the atmosphere (Baltensperger et al., 2002). Particles larger than 50 nm show much less variation in the given time periods. This second mode referred to as accumulation mode results from incomplete combustion and consists of soot particles emitted especially from diesel vehicles. Because of the lower size resolution of the ELPI and higher detection limit ($D > 29$ nm), the nucleation mode cannot be resolved in the ELPI size spectra.

During the night (upwind conditions), the lowest number concentrations were measured, and the accumulation mode, which has a strong traffic component, but is also transported over long distances, appears more strongly. Ultrafine particles with a concentration maximum around 30 nm were also observed. These particles derive from an aged aerosol, where the even smaller particles had enough time to coagulate. More information on the SMPS spectra is found in Rosenbohm et al. (2005).

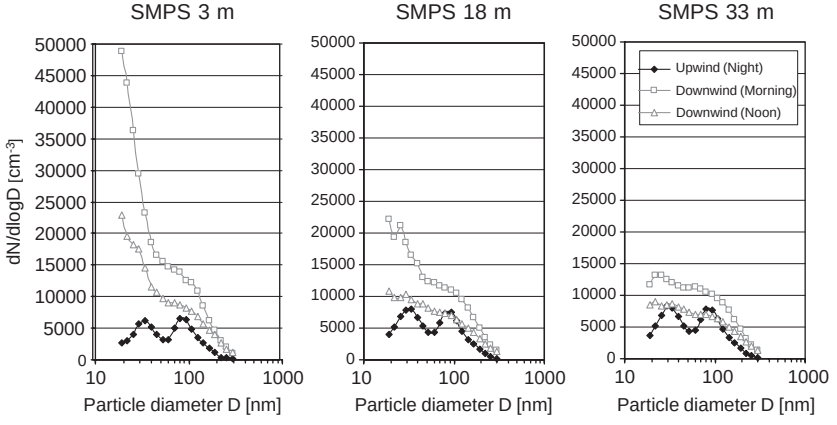

Fig. 5. Particle number size distributions measured by SMPS systems at three different heights.

3.4. Vertical profiles of the NO_x mixing ratio

Data processing of NO_x was performed in two steps. At first, the measured values were quantitatively corrected according to the calibration curves of the intercomparison measurements. Furthermore, the NO_x analyser did not show the same response time on concentration changes as the NO_2 analyser. Comparing both NO_2 curves, the NO_2 analyser was characterized by an efficient time response (less than 10 s), while the NO_x analyser recorded concentration changes with a delay. Thus, ascending and descending profiles showed a different shape. Therefore, separate mean values of NO_x mixing ratio of the ascents and descents were formed over a time period of 3 h. Then the averaged ascents and descents were moved against each other on the vertical axis until the best correlation was reached. This was the case with a vertical shift by about 8 m (variation coefficient $R^2 = 0.88$). On the time axis, this represents a shift by 27 s for the entire NO_x data set of the chemiluminescence analyser.

As seen in Fig. 6, the corrected vertical profiles of the NO_x mixing ratio show a similar behaviour at different daytimes as the profiles recorded by the DC. On the upwind side of the motorway, a nearly constant NO_x value was measured over the full profile. Because of its low mixing ratio of 4 ppb NO_x and the relatively short time period (45 min) with only 8 profiles, the mean value displays some fluctuations but no significant height dependence. In the course of the morning on the downwind side, the maximum mixing ratios with more than 50 ppb NO_x were measured near ground level. With increasing height, the values diminished rapidly and reached the background value of 20–25 ppb already at 15 m. This value seems to be relatively high for a background situation, but contemporaneously on the upwind side of the motorway likewise mean NO_x values of around 20 ppb were registered. Around noon, the NO_x mixing ratios were significantly lower than during the forenoon. In contrast to the DC measurements, the maximum of the NO_x mean values was observed already near the ground and decreased continuously with increasing height, and the background value (with 5–6 ppb now only somewhat higher than during the night) was attained at a height of approximately 20 m above ground, similarly to the particle data. Due to the high pollutant production of the morning traffic and the still existing stable nocturnal boundary layer in the early forenoon, the air pollutants accumulate near the ground, and a steep decrease with increasing height is observed. The air pollution declines from forenoon to noon by more than 50%; the gradient near the ground is substantially less developed. This phenomenon can be explained by the lower traffic density and the higher wind speeds during the afternoon provoking lower concentrations because the emissions are dispersed in a larger air volume as nicely seen in Fig. 2 around 14:00 CEST. Also, the vertical transport of the air mass near the ground and dispersion in a much higher air volume results in a reduction in the pollutant concentration. The average NO_x values obtained with profile measurements were confirmed by the ground monitoring stations (see Fig. 6). In the upwind case, these stations show the expected extrapolation of the vertical profile to the

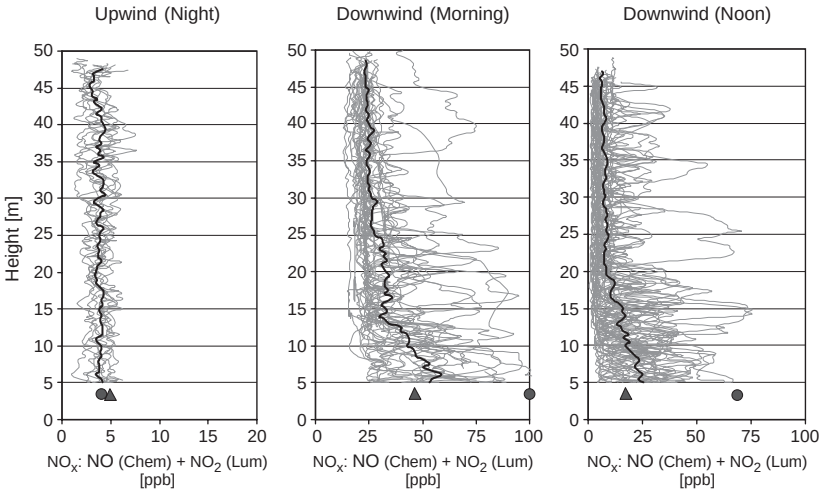

Fig. 6. Vertical profiles of the NO_x mixing ratio on the upwind side (23:00–24:00 CEST) of the motorway, as well as on the downwind side in the morning (07:00–10:00 CEST) and around noon (11:00–14:30 CEST). Thick black line: average of all profiles in the respective period; dotted grey lines: single profile runs. Filled circles: ground monitoring station beside motorway lanes; filled triangles: ground monitoring station 80 m from motorway.

concentration at ground level. During the downwind situations, the monitoring station next to the motorway lanes shows higher concentrations than the 5 m-value of the tower (60 m from the motorway). On the other hand, lower NO_x mixing ratios were measured at the sampling station located in a distance of 80 m from the motorway. This also refers to the dilution with increasing distance to the highway.

A more detailed discussion of the vertical profiles of the components NO and NO_2 and their partitioning as a function of the height is presented in Kohler et al. (2005).

4. Correlation between particle surface area and NO_x

For the calculation of vehicle emission factors, a good correlation between particles and NO_x is of great importance since often particle emission factors are computed using the respective known NO_x values. As already mentioned above, the pattern of the NO_x curves strongly resembles the vertical profiles of the DC for the same time period. The arithmetic mean of the concentration in the height segment from 35 to 45 m above ground level was assumed as background value for the "active" surface area (SA) as well as for the NO_x values, and was subtracted from the amount in the lower height range. In this way only the traffic-related emissions were correlated yielding the following linear equations:

$SA_t = 1.93NO_{xt} - 0.038$ with a correlation coefficient $R^2 = 0.96$ for the downwind morning period,

$SA_t = 1.98NO_{xt} - 0.626$ with a correlation coefficient $R^2 = 0.89$ for the downwind noon period,

where SA_t is the surface area caused by traffic emissions and NO_{xt} the nitrogen oxide mixing ratio contributed by traffic sources. The slopes of the two regression lines do not show a significant difference. These findings do not only point to the fact that there exists a common source for both air pollutants, but also confirm the assumption that the pollutants experience a similar dilution during the transfer from the source to the sampling station (a distance of 60 m, which corresponds to a transportation time of at least 30 s). Looking at the ratio of the surface area to the NO_x mixing ratio emitted by the motorway traffic as a function of the height (Fig. 7), this curve exhibits interesting features. At 5–10 m, a lower surface area concentration was found compared to NO_x than above 10 m. An explanation for this fact is that many HDV in Germany have their tail pipe at a height of about 4 m. HDV in general show a higher

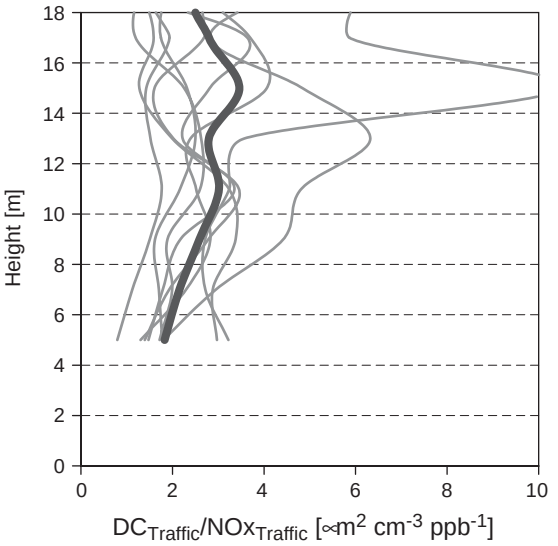

Fig. 7. Hourly ratios (thin lines) and mean values (thick black line) of the surface area (DC) to the NO_x mixing ratio for pure traffic emissions as a function of the height on 14 May 2001, 07:00–18:00.

emission ratio surface area/NO_x than light-duty vehicles (LDV). The investigated clear day was characterized by a high solar irradiation and temperatures of about 25 °C in the afternoon. Thus, during the forenoon, the tarmac of the motorway warms up faster than the surrounding agricultural areas. As a consequence, the thermal gradient induces a vertical rise of the warmer air above the motorway, resulting in a vertical shift of the traffic emissions. Another process to remove particles more effectively than NO_x molecules from the atmosphere would be dry deposition on grassland, which would lead to a depletion of the surface area compared to NO_x in the air masses near the ground.

High correlations between particulate air pollutants and NO_x were observed also in other studies on traffic emissions. The "active" particle surface area can best be compared with the calculated geometric surface area assuming spherical particles (for $D < \sim 100$ nm these two values are identical) from measurements with differential mobility particle sizer (DMPS) systems, as accomplished by Ketzel et al. (2003). In this study, likewise, a high correlation between geometric particle surface and NO_x with a correlation coefficient $R^2 = 0.78$ was found for traffic emissions. For the particle number concentration (10–700 nm) and the NO_x concentration, an $R^2 = 0.81$ was reported in the above-mentioned study. For a traffic aerosol also quite high correlations between PM2.5 and NO_x ($R^2 = 0.62$) as well as between PM10 and NO_x ($R^2 = 0.43$) are known (Harrison et al., 2001).

5. Determination of emission factors

From the results of the profile measurements, we determined the source strength of the motorway traffic emissions for the particle surface area and for various particle size ranges. We calculated specific emission factors for the driving mode in the investigated motorway segment, where LDV cruise with an average speed of 113 km h^{-1}, HDV with 86 km h^{-1} and the share of LDV equipped with diesel engines is about 20% (Kühlwein and Friedrich, 2005). The source strength was evaluated by the difference between the downwind and the background concentration, integrated over the height of the plume, as expressed in

$$Q_i = \int_0^h v_\perp(z)(c_i(x_d, z) - c_i(x_u, z))\,dz, \quad (1)$$

where Q_i is the source strength of the substance i (units: y/(km h), where y stands for 1, cm^2, cm^3 regarding particle number, surface or volume). c_i is the concentration of the substance i, $v_\perp$ is the wind velocity perpendicular to the motorway, h is the height of the traffic exhaust gas plume at the upwind and downwind position x_u and x_d, respectively. Since no upwind measurements were available, the mean concentration in the height interval from 35 to 45 m was taken as background value (Fig. 8a). The accuracy of this assumption was verified by the NO_x profiles, which showed equal concentrations on the upwind side as in the upper height level of the downwind side (Kohler et al., 2005). To compute the source intensities for various hours, the vertical concentration profiles were aggregated to 1 hourly means and height segments of 2 m. It was assumed that the mean value of 5 m above ground was constant in the gap between the ground and the beginning of the profile measurements.

For this calculation, all time periods with constant wind direction across the motorway in the defined sectors (see Section 2.2) were used, i.e. 18 1-h values of different days. We analysed the particle surface area concentration measured by the DC and the following quantities of the ELPI: the total number concentration of particles with $D < 10\,\mu m$ (N10), as well as the volume of particles smaller than 0.25, 0.64 and 1 μm (termed as V0.25, V0.64, V1, respectively) assuming spherical particle shape. The hourly source strength divided by the total number of vehicles (n_{Total}) which passed the sampling site within this hour gives the emission factor per vehicle of the substance i (EF$_i$): $EF_i = Q_i/n_{Total}$. As an example for the surface area, the result is depicted in Fig. 8b, where an evident correlation between the share of HDV and the emission factor per vehicle can be seen. Mean emission factors per vehicle as well as those separated for LDV (EF$_{i(LDV)}$) and HDV (EF$_{i(HDV)}$) derived using a multiple linear regression model according to Eq. (2) are given in Table 3:

$$EF_i = \frac{n_{LDV}}{n_{Total}} EF_{i(LDV)} + \frac{n_{HDV}}{n_{Total}} EF_{i(HDV)}. \quad (2)$$

The uncertainty represents the standard error of the regression, which is relatively large as a consequence of the small data set. It has to be considered that the emission factor for N10 determined here with the ELPI cannot be directly compared with commonly published total particle number emission factors (by CPC measurements), because N10 does not contain the nucleation mode particles with $D < 29$ nm. However, this size range has a negligible contribution to the volume emission factors. Mass balances and emission factors of the

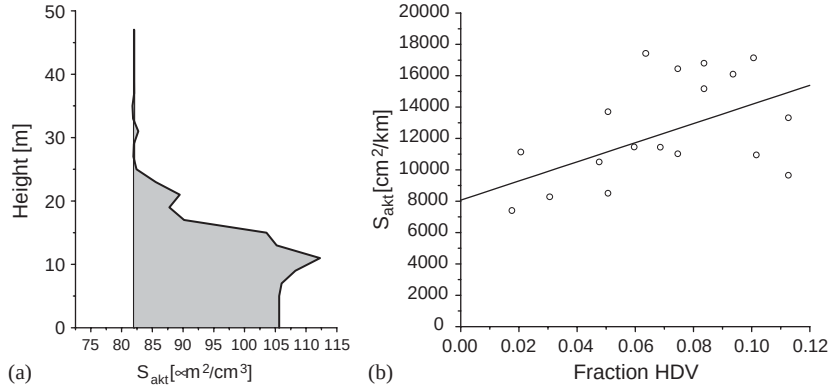

Fig. 8. Determination of emission factors for the surface area by extracting the traffic contribution (grey filled area), obtained from the measured downwind profile and the calculated upwind profile from the mean value of the background concentration in the height level between 35 and 45 m (a). Surface area emission factor per vehicle (1-h averages) as a function of the share of HDV (b). The regression line was used to calculate emission factors for LDV and HDV (see Table 3).

Table 3
Emission factors (EF) and uncertainty for various parameters

Parameter	Vehicle category	Unit	EF	Std. err.
Surface area	All	$cm^2\,km^{-1}$	12500	±3300
	LDV	$cm^2\,km^{-1}$	8100	±1700
	HDV	$cm^2\,km^{-1}$	69000	±23700
N10	All	particles km^{-1}	1.80E+14	±0.42E+14
	LDV	particles km^{-1}	1.22E+14	±0.49E+14
	HDV	particles km^{-1}	7.79E+14	±6.32E+14
V0.25	All	$cm^3\,km^{-1}$	0.06	±0.02
	LDV	$cm^3\,km^{-1}$	0.03	±0.02
	HDV	$cm^3\,km^{-1}$	0.41	±0.26
V0.64[a]	All	$cm^3\,km^{-1}$	0.07	±0.02
V1[a]	All	$cm^3\,km^{-1}$	0.09	±0.02

The surface area was measured by a DC, and the number and volume by an ELPI (lower cut-off size 29 nm). N10 indicates the number of particles with diameters D in the size range $0.029 < D < 10\,\mu m$, V_x is the volume of particles $0.029 < D < x\,\mu m$.

[a]Because of the big spread of V0.64 and V1, it was not possible to determine emission factors for LDV and HDV.

gaseous compounds are found in Kohler et al. (2005) and Corsmeier et al. (2005b).

6. Conclusions

For the first time, highly time-resolved vertical profiles of nitrogen oxides and particulate air pollutants on both sides of a motorway were recorded by a large set of instruments which were installed on electrically operated elevators fixed at two 52-m-high towers. On the upwind side of the motorway, no height-dependent difference in the particle number and surface area concentrations was observed. On the downwind side, substantially higher concentrations were measured near the ground than in the upper part of the investigated height profile. Around noon the particle number concentrations even increased slightly with height up to 10 m, which can be explained by more intense thermal vertical air transport above more strongly light-absorbing road surfaces than above the surrounding agricultural or grass areas. The results of height-dependent concentration differences were supported by the stationary SMPS measurements. Particularly, particles generated by homogeneous nucleation during the cooling of the exhaust gas ($D < 50$ nm) were found in high concentrations only on the lowest height level.

The vertical distribution of the NO_x mixing ratios showed a similar behaviour as the particle number concentrations: no height dependence on the upwind side as well as a continuous decrease with increasing height from 5 to 30 m above the earth's surface on the downwind side of the motorway. Thus, the results in this study confirmed a good correlation between particles (number and surface concentration) and the NO_x values, as observed also in former case studies, although the ratio of surface area to NO_x of traffic emissions was not exactly constant with height.

At around 30 m above ground the background value was attained. By subtracting the respective background values (above 35 m) from the concentrations measured in the near-surface air layers the direct emissions of the road could be assessed, even though contemporaneous upwind and downwind measurements were missing. Together with the parallel wind and traffic measurements, absolute emission factors for the traffic situation on the investigated motorway section were determined.

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