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Aerosol and NO_x emission factors and submicron particle number size distributions in two road tunnels with different traffic regimes

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Abstract. Measurements of aerosol particle number size distributions (18–700 nm), mass concentrations (PM2.5 and PM10) and NO_x were performed in the Plabutsch tunnel, Austria, and in the Kingsway tunnel, United Kingdom. These two tunnels show different characteristics regarding the roadway gradient, the composition of the vehicle fleet and the traffic frequency. The submicron particle size distributions contained a soot mode in the diameter range D=80–100 nm and a nucleation mode in the range of D=20–40 nm. In the Kingsway tunnel with a significantly lower particle number and volume concentration level than in the Plabutsch tunnel, a clear diurnal variation of nucleation and soot mode particles correlated to the traffic density was observed. In the Plabutsch tunnel, soot mode particles also revealed a diurnal variation, whereas no substantial variation was found for the nucleation mode particles. During the night a higher number concentration of nucleation mode particles were measured than soot mode particles and vice versa during the day. In this tunnel with very high soot emissions during daytime due to the heavy-duty vehicle (HDV) share of 18% and another 40% of diesel driven light-duty vehicles (LDV) semivolatile species condense on the pre-existing soot surface area rather than forming new particles by homogeneous nucleation. With the low concentration of soot mode particles in the Kingsway tunnel, also the nucleation mode particles exhibit a diurnal variation. From the measured parameters real-world traffic emission factors were estimated for the whole vehicle fleet as well as differentiated into the two categories LDV and HDV. In the particle size range D=18–700 nm, each vehicle of the mixed fleet emits $(1.50\pm0.08)\times10^{14}$ particles km^{-1} (Plabutsch) and $(1.26\pm0.10)\times10^{14}$ particles km^{-1} (Kingsway), while particle volume emission factors of $0.209\pm0.008\,cm^3\,km^{-1}$ and $0.036\pm0.004\,cm^3\,km^{-1}$, respectively, were obtained. PM1 emission factors of $104\pm4\,mg\,km^{-1}$ (Plabutsch) and $41\pm4\,mg\,km^{-1}$ (Kingsway) were calculated. Emission factors determined in this work were in good agreement with results from other studies.

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1 Introduction

Airborne particles are known to have an important influence on global climate by absorption and scattering of radiation (Haywood and Boucher, 2000) and by acting as cloud condensation nuclei (Lohmann and Feichter, 2005). In addition, numerous studies have verified that aerosol particles can cause adverse effects on human health. Whereas initially the consequences of an exposure to an increased particle mass concentration PM10 and PM2.5 (particulate matter with an aerodynamic diameter $D<10\,\mu$m and $D<2.5\,\mu$m, respectively) were investigated (Dockery and Pope, 1994; Pope et al., 2002), in recent studies number and surface area concentration as well as the chemical composition of the ultrafine particles ($D<100$ nm) reveal a more toxic effect than the same parameters of coarser particles (Oberdörster, 2000; Donaldson et al., 1998).

Traffic emissions are one important source of submicron particles. Road tunnels serve as an appropriate instrument to investigate the large spectrum of vehicular emissions, because in a tunnel dilution and boundary conditions are better known than in open field experiments, and the influence of the varying meteorological parameters is usually negligible. Various tunnel studies mainly dealt with the gaseous

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Table 1. Tunnel specifications.

	Plabutsch	Kingsway
Location	Graz	Liverpool
Number of lanes	2	2
Traffic flow	bidirectional	unidirectional
Length [m]	9755	2480
Traffic density [d^{-1}]	23 000	20 000
HDV share [%]	18	7
Residence time of the measured aerosol in the tunnel [s]	10–400	200–350

emissions for comparing emission factors obtained by test bench measurements with the real-world values, e.g. Pierson et al. (1996), Robinson et al. (1996), McLaren et al. (1996), John et al. (1999). Results of tunnel measurements of various particulate mass concentrations were reported by Weingartner et al. (1997) and Kirchstetter et al. (1999). Only in recent years tunnel studies focused on the particle size distribution (Weingartner et al., 1997; Abu-Allaban et al., 2002; Gouriou et al., 2004) and on emission factors for various size classes (Gidhagen et al., 2003; Kristensson et al., 2004).

Typical particle number size distributions from vehicle emissions show three modes (Morawska et al., 1999; Kittelson et al., 2000). The first mode is often observed in the size range below 50 nm and is also termed nanoparticles (Kittelson, 1998). These particles are formed by condensation of the exhaust gas directly after emission, a homogeneous nucleation process and are therefore called nucleation mode particles in this paper (Baltensperger et al., 2002). They mainly consist of volatile material which contains sulphate and some organic species from unburned lubricating oil (Tobias et al., 2001). The concentration of the nucleation mode particles depends on various parameters such as ambient temperature and relative humidity (Bukowiecki et al., 2003). The second mode includes the size range between 50 and 300 nm, exhibiting a maximum concentration at $D \approx 80$–100 nm. It is composed of soot particles, forms strongly branched chain aggregates and predominantly stems from incomplete combustion. Therefore in the following, these particles are termed soot mode particles to distinguish them from the accumulation mode particles which are also present in the background aerosol and show a larger modal diameter. Coarse particles in the range between 1 μm and 10 μm comprise the third mode, which is much more pronounced in the volume or mass than in the number size distribution. It mostly contains abrasion products from break and tyre wear as well as resuspended material from the road or from adjacent surfaces (Gehrig et al., 2004).

In 2000 the EU research project Assessment and Reliability of Transport Emission Models and Inventory Systems (ARTEMIS) was started to develop a harmonised emission model for road, rail, air and ship transport to provide consistent emission estimates at the regional, national and international level. As part of the ARTEMIS project road tunnel studies were performed to validate fleet weighted gaseous emission factors based on chassis dynamometer tests. In order to gain insight into the interrelation between the gaseous and particulate components, particle size distributions and mass concentrations were measured as well, but were not the main focus in the ARTEMIS workpackage.

The goal of this paper is to present the results of particle measurements in two road tunnels with different levels of air pollution, different vehicle fleet composition and roadway gradients. First results of one tunnel study were illustrated by Sturm et al. (2003) with the objective to show similarities and differences between tunnel and open field experiments. In contrast to this previous paper, this work focuses on the behaviour of nucleation mode particles in the presence of a temporally varying soot mode and demonstrates the discrepancies observed in two road tunnels. The second aim was the determination of emission factors for several aerosol parameters such as PM10, PM1, as well as particle number and volume for several characteristic size intervals.

2 Experimental

2.1 Description of the tunnels and the measurement sites

Measurements of particle size distributions, mass concentrations and nitrogen oxides (NO_x) were conducted in two road tunnels, in the Plabutsch tunnel and in the Kingsway tunnel, with different speed limits and traffic regimes. Some specifications for each tunnel are given in Table 1 to facilitate the comparison.

The Plabutsch tunnel is located in South Eastern Austria near Graz. It is already described in Sturm et al. (2003). Briefly, the tunnel has a length of 9755 m and was opened to traffic in 1987. It serves on the one hand as bypass of the city of Graz, on the other hand as part of an important transit route through the Eastern Alps from Germany and the northern areas of Austria to the Balkan region. For this reason it carries not only local and regional commute traffic but also international long-distance traffic, especially road transport of goods. As a consequence, the heavy-duty vehicle share is relatively high with 18% of 23 000 vehicles crossing the tunnel on an average day. The speed limit is 80 km/h. During the time when the measurements took place (from Monday 5 November until Monday 12 November 2001), traffic in the Plabutsch tunnel was operated bidirectionally on one lane per direction in a single bore (the second bore was under construction).

The ventilation system of the tunnel is transverse. Fresh air is provided from two funnels into a fresh air duct situated above the tunnel ceiling. From there it is dispersed along

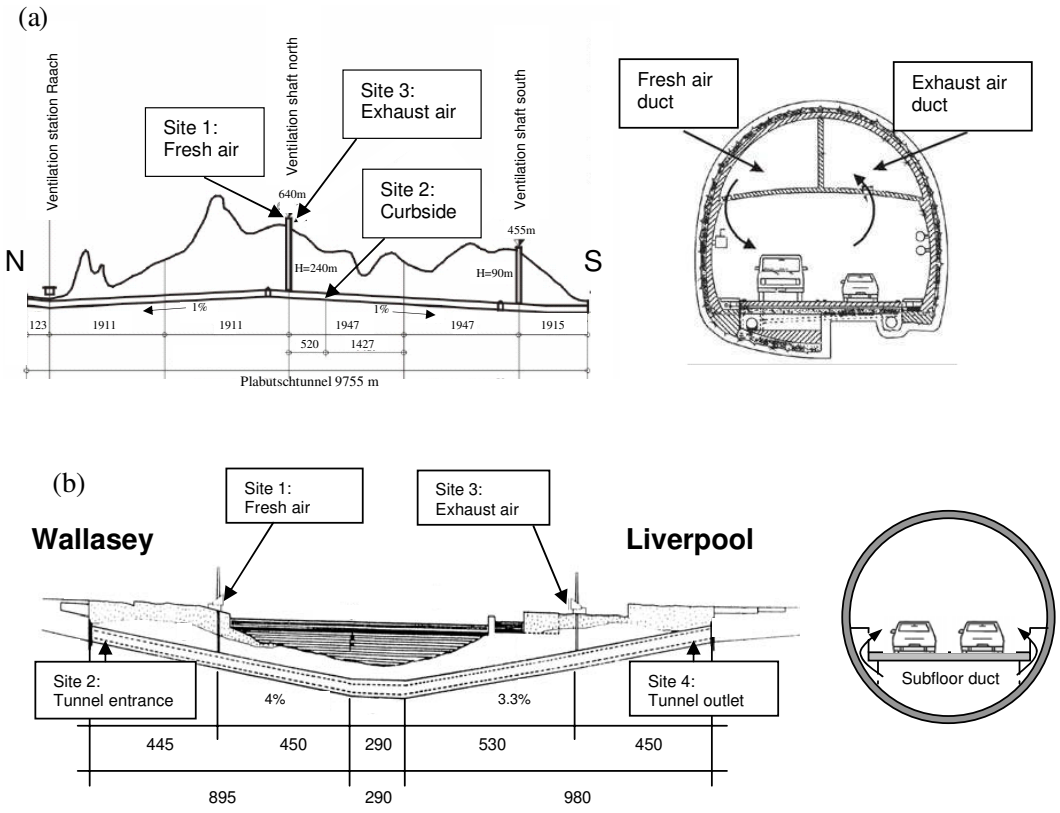

Fig. 1. Schematic diagram of the Plabutsch **(a)** and the Kingsway tunnel **(b)**. On the right cross sections of each tunnel are presented. Distances are given in metres.

the whole section into the road tunnel. Waste air is sucked off through another duct and released to the environment by the exhaust funnels. Ventilation of the Plabutsch tunnel is separated into five segments, each of which with a length of about 2 km. Air pollution in Sect. 3 was investigated during the field campaign.

In this tunnel three sampling sites were used to characterize pollution emissions by passing vehicles (see Fig. 1a):

- Measurement of the fresh (intake) air at the top of the ventilation funnel at an altitude of 640 m above sea level in a forest on a hill, representing typical background concentration levels (site 1)

- Curbside measurement inside the tunnel on the parking area of an emergency niche, directly beside the traffic lanes (site 2)

- Measurement of the ventilation outlet (waste) air at the top of the 240 m high exhaust funnel (site 3)

At site 2 the fresh emissions of vehicles are measured, while at site 3 the aerosol has aged during a travel time of about 12 s in the ventilation shaft without further emissions or dilution. Before entering the shaft, the aerosol represents a mix of newer and older emissions collected over the whole ventilation segment.

The Kingsway tunnel is a road tunnel under the Mersey river in the United Kingdom connecting the city of Liverpool in the east to its suburbs Wallasey and Birkenhead in the west. It was opened in 1971 and incorporates two circular bores, which are 2480 m long. Each bore carries two lanes of uni-directional traffic, with the north bore carrying traffic from Wallasey to Liverpool, and the south bore carrying traffic in the opposite direction. Vehicles passing the tunnel are mainly commuter traffic between Liverpool and its suburbs on the peninsula of the Wirral. The speed limit in this toll tunnel is 40 mph (64 km/h).

The tunnel ventilation system is semi-transverse. Clean air enters the tunnel via the two ventilation shafts and via the portals. The air from the ventilation shafts is fed into a subfloor duct and permeates into the tunnel through vents along its entire length. The vents are designed to allow for an even flow of inlet air along the whole tunnel length. Exhaust air is removed via the ventilation shafts, and can also leave via the portals. However, at times of heavy, congested traffic, jet fans mounted in the tunnel crown are activated. The tunnel is fitted with equipment, at several locations, to monitor carbon monoxide levels and visibility. These two criteria are used to

Table 2. Instrumentation.

Instrument	Parameter	Time resolution	Sampling sites
Plabutsch tunnel			
Scanning Mobility Particle Sizer (SMPS)	size distribution D=16–730 nm	5 min	1, 2, 3
Tapered Element Oscillating Microbalance (TEOM)	mass concentration PM10	1 min	1, 2
Kleinfiltergerät	mass concentration PM1	1 h	1, 2
NO_x analyzer	NO_x	1 min	1, 2, 3
Kingsway tunnel			
Scanning Mobility Particle Sizer (SMPS)	size distribution D=16–730 nm	5 min	1, 2, 3
Tapered Element Oscillating Microbalance (TEOM)	mass concentration PM2.5	1 min	1, 2, 3, 4
Kleinfiltergerät	mass concentration PM1	1 h	1, 4
NO_x analyzer	NO_x	1 min	1, 2, 3, 4

adjust the speed of the ventilation system, and an alarm and manual control system is in place.

Emission measurements were performed in the north bore (traffic to Liverpool) with a traffic frequency of about 20 000 vehicles per day and a heavy-duty vehicle share of 7% on average. This bore shows different roadway gradients, a downward slope of 4% in the first 900 m after the tunnel entrance, then a section with nearly even conditions, followed by a continuous upward slope of 3.3% until the eastern portal (Fig. 1b).

During the field campaign of the Kingsway tunnel from Saturday 8 February until Friday 14 February 2003, measurements took place at four different sampling sites.

- Measurement of the fresh (intake) air in the Promenade ventilation station located in Wallasey on the left bank of the river Mersey, representing urban background concentration levels (site 1)
- Curbside measurement inside the tunnel, 50 m from the entrance of the Wallasey portal (site 2)
- Measurement of the ventilation outlet (waste) air at the Victoria ventilation station located in Liverpool (site 3)
- Curbside measurement inside the tunnel, 50 m from the outlet of the Liverpool portal (site 4)

These four sampling sites allowed a characterization of all air masses entering or leaving the tunnel. During the experiment, the ventilation was configured in a way which would encourage the longitudinal flow of air through the bore in the direction of the traffic. At the Promenade ventilation station the inlet air fan was switched on and the exhaust fan was switched off. At the Victoria ventilation station the exhaust fan was turned on and the inlet fan was deactivated. The jet fans in the tunnel bore were not used for extended periods.

2.2 Instrumentation

A large set of instruments was used in both tunnel campaigns which covered a variety of gaseous parameters that are not further discussed in this paper except for NO_x. Table 2 gives an overview of the instruments relevant for the discussion of the results of this paper and their sampling locations. During both campaigns, three Scanning Mobility Particle Sizers (SMPS, TSI Inc.) were in operation to measure the submicron particle size distribution with diameters D between 18 and 700 nm. The components of all SMPS systems consisted of the same type and model, a Differential Mobility Analyzer (DMA 3071A) and a Condensation Particle Counter (CPC 3022A). They were operated with $0.3\,l\,min^{-1}$ aerosol and $3\,l\,min^{-1}$ sheath airflow rate in a 5-min interval with an up-scan time of 220 s, a down-scan time of 20 s and a waiting time of 60 s between each spectrum. Whereas in the Plabutsch tunnel all sites were equipped with SMPS systems, in the Kingsway tunnel, only at sites 1–3 particle size spectra were recorded.

Aerosol mass concentration of PM10 (in the Plabutsch tunnel) and PM2.5 (in the Kingsway tunnel) was collected by Tapered Element Oscillating Microbalances (TEOM). The principle of the TEOM measurement consists in a frequency change of oscillation when particles are deposited on a vibrating collection medium, such as a filter or impaction plate (Baltensperger et al., 2001). The change in frequency corresponds to the mass concentration on the tapered element. All TEOM devices were operated with a sampling airflow rate of $1\,m^3\,h^{-1}$ and with a temperature of 40°C. Since only two

instruments were available for the Plabutsch campaign, one TEOM recorded the fresh air mass concentration, the other one was positioned at the curbside.

Mass concentration of PM1 was measured by so called Kleinfiltergeräte (Small filter devices, Type LVS 3.1, Derenda, Germany). Air is sucked through filters and the particles are deposited on the filter material. By means of differential weighing of the coated filters after conditioning at 25°C, the mass concentration of the aerosols is determined. These instruments were operated with a sampling airflow rate of 2.3 $m^3 h^{-1}$. The filters needed to be changed manually after approximately one hour sampling time. Because the Kleinfiltergeräte were very labour intensive, they were only during special time periods (rush hour, noon, and night) in operation and their results were mainly used for calibration purposes of other instruments. Both campaigns were performed with two instruments, one sampling the intake air and the other located at the curbside.

Nitrogen oxides (NO_x) measurements were carried out at all sampling sites with NO_x analysers (e.g. Type Monitor Labs ML 8841) applying molybdenum converters and the ozone chemiluminescence method.

The fluxes of the inlet and the exhaust air of each ventilation shaft were recorded automatically by flowmeters operated by the tunnel operation centres. Flow measurements in the Plabutsch tunnel (divided into five ventilation sections) took place in section No. 3, only supplied by ventilation shaft north (Fig. 1a). Since the traffic in the tunnel bore is operated bidirectionally, horizontal air exchange between section 3 and the two adjacent ventilation segments was neglected. Actual volumetric flow rates at the entrance and at the outlet of the Kingsway tunnel were measured by Ultrasonic Gas Flowmeters (FLOWSIC 600). Data sets were available on the basis of one-minute values, which were then aggregated to 0.5-h and 1-h mean values. Time periods with uncertain or low flow rates were not used for the calculation of emission factors.

Traffic densities, differentiated in light-duty (LDV) and heavy-duty vehicles (HDV), were recorded by loop detectors in the tunnel. Motor vehicles were categorised according to their length. Vehicles with a length L<6 m were defined as LDV, i.e. motor cycles, passenger cars and delivery vans. Those with a length L>6 m were assigned to HDV, for example trucks and busses which are predominantly driven by diesel motors. These data were provided by the Plabutsch tunnel operator with a time resolution of 1 min and by the Merseyside passenger transport authority for the Kingsway tunnel with a time resolution of 1 h.

2.3 Quality assurance

Since all instruments mentioned above were used in double, triple or quadruple implementation, a careful calibration procedure and several intercomparison measurements were indispensable to obtain comparable data during the pollution experiments in the tunnels. Quality assurance measurements were performed before and after the campaign in each case.

The four NO_x analysers were calibrated with test gas of a well-known concentration and parallel measurements with ambient air concentrations. Further calibrations took place during the field campaigns.

The quality assurance procedure of the three SMPS systems done before the tunnel experiments included several steps. First the Condensation Particle Counters were tested by parallel measurements in order to guarantee a consistent counting of the particles. Second the shape of the size distribution, especially the correct sizing of the SMPS systems, was investigated using a monodisperse aerosol. This procedure is very important because only in case of a correct sizing, a later transformation of the recorded size distributions by a correction factor is applicable. It was carried out by a tandem DMA configuration where a monodisperse aerosol with a defined particle diameter was selected in a first DMA. These monodisperse particles with diameters D=20 nm, 40 nm, 80 nm, 150 nm and 300 nm were fed into the DMAs of the SMPS systems which recorded the size spectrum. Differences were found to be around ±5%, with the lowest standard deviations in the size range D<100 nm.

The third step was parallel measurements with a polydisperse aerosol having a concentration and size distribution similar to the aerosol which should be sampled during the campaign. These measurements were performed in the ventilation station at the outlet air during a time period of one day. As an example, the results of the intercomparison subsequent to the Plabutsch campaign are presented in Fig. 2. All SMPS systems measured the same aerosol with copper sampling lines of equal length to avoid different diffusion or coagulation losses. The instruments show a very similar shape of the size spectra. Nevertheless discrepancies of 30% were found in the nanoparticle size range (D<50 nm), approximately 25% at D=60–120 nm and 10% at D>200 nm. The reasons for these discrepancies are not clear.

The correction factor for each SMPS instrument was calculated as follows: First 2-h mean values of the size distributions were formed and then the average of these mean size spectra was computed. This resulted in an "ideal" spectrum to which the spectra of all SMPS systems were transformed by a certain size dependent correction factor. The permissibility of this procedure was proven by the fact that during the one-day measurement period of the exhaust air, the size distributions changed significantly. Therefore the size-dependent correction curve as well as the standard deviation from all 2-h periods was calculated and plotted in Fig. 2 for each SMPS system. The best agreement with the mean curve was obtained by SMPS No. 2 which had a correction factor of nearly 1 in the size range D=30–200 nm. SMPS No. 3 showed a consistent correction factor over a wide size range. SMPS 1 exhibited a continuous increase of the correction factor from 0.7 at 16 nm to 1.4 at 400 nm, but the error bars from the various 2-h periods were very small (mostly less

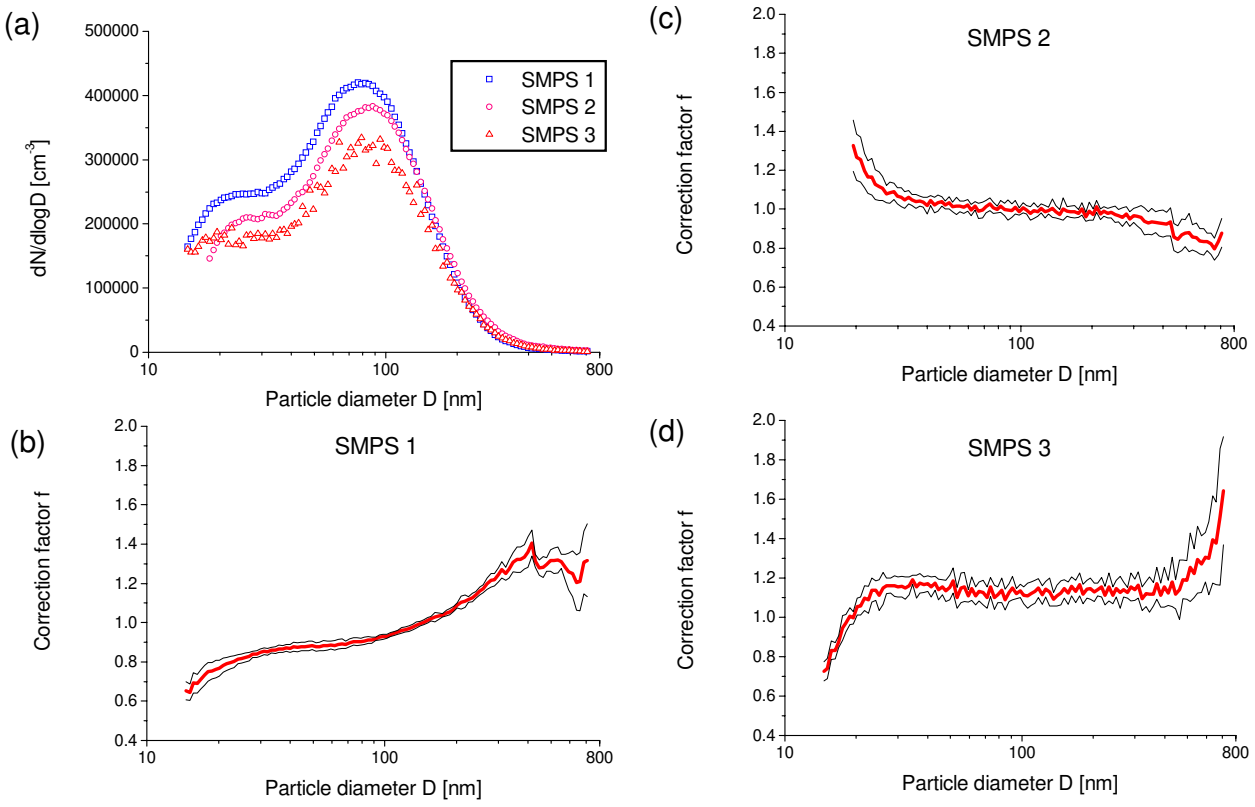

Fig. 2. Intercomparison measurement of three SMPS systems (mean value of 12 size spectra, **a**) and the calculated size-dependent correction factor for each instrument (**b** to **d**). The thin black lines indicate the standard deviation of the correction factor derived from eight 2-h mean values with different concentrations and different size distribution structure.

than 2%). Also in the margin ranges, the standard deviations did not exceed 10%. This consistency even for totally different size distributions confirms the applicability of correcting the size spectra in this way.

In addition, all SMPS size spectra were corrected for diffusion losses in the sampling line, because due to the amount of space at the sites, lines of different length had to be used.

2.4 Data processing

For the Kingsway tunnel, mean values of one hour were calculated, according to the availability of the traffic data. Data from the Plabutsch campaign were aggregated to both 30-min and 1-h averages. In order to obtain further information about the nucleation and the soot mode, the 1-h size spectra were parameterized by a bimodal fit function with log-normal distribution using the formula described by Heintzenberg (1994). By this procedure, the geometric standard deviation, the number median diameter and the integrated number concentration of each mode is defined. The boundary conditions for the median of the nucleation mode were set at D=10–40 nm, those for the soot mode at D=40–200 nm. Assuming a spherical particle shape, particle volume size distributions were then calculated from the number size distributions.

3 Results and discussion

In this section, the results of the particle size distribution measurements at selected sampling sites are first characterised for both tunnels, and then the method of determining emission factors followed by the vehicle emission factors for a number of aerosol parameters is discussed.

3.1 Characterization of the submicrometer size spectra at the different sampling sites

Contour plots of the time series of the corrected particle size distributions measured in the Plabutsch and the Kingsway tunnel are depicted in Fig. 3, in each case for the fresh air provided to the tunnel by the ventilation inlet and for the exhaust air at the ventilation outlet. The fresh air measurement at the Plabutsch tunnel represents values of a typical background site (Ketzel et al., 2004) with a slight diurnal variation and a maximum in the accumulation mode at diameters of around 100–120 nm. An increased concentration

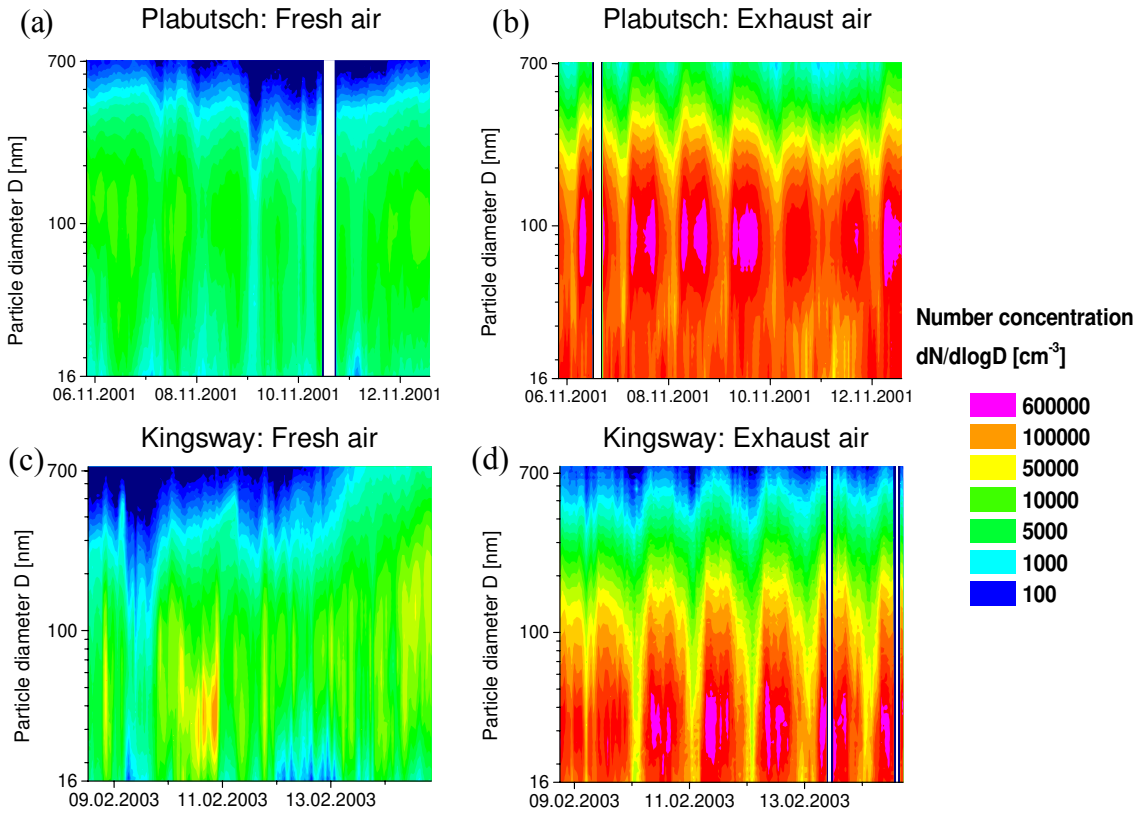

Fig. 3. Time series of the particle size distribution in the Plabutsch (**a** and **b**) and in the Kingsway tunnel (**c** and **d**). White bars represent measurement failures.

of nucleation particles was observed during the day which might be attributed to secondary aerosol formation. In contrast, the fresh air sampled during the Kingsway campaign is characterized by an urban background where the concentrations primarily depend on the wind direction and the weather conditions. There are a lot of fluctuations depending on the air mass trajectory (e.g. on 10 February), but no evident diurnal variation is visible.

Comparing the exhaust air measurements of both tunnels, some similar but also some different features become apparent. The clear diurnal variation of the particle number size distribution is obvious for both tunnels. During the night, especially in the Kingsway tunnel low concentrations were measured whereas during daytime, even the emission peaks of the morning and the evening rush hour were perceptible. Weekends, i.e., 10 and 11 November 2001 as well as 8 and 9 February 2003, are characterised by lower daytime maxima compared to weekdays. A similar pattern was observed by Ketzel et al. (2003) in a street canyon in Copenhagen.

The most obvious difference between the two tunnels is the shape of the particle size spectra. These show the maximum concentration for the Plabutsch tunnel in the size range D=80–100 nm, and for the Kingsway tunnel in the range D=30–40 nm. In the size ranges at the upper end of the SMPS detection, the number concentrations are considerably lower in the exhaust air of the Kingsway than of the Plabutsch tunnel. Another disparity appears in the explicitly lower number concentrations measured in the Kingsway tunnel during nighttime. This circumstance can be explained by the fact that the Kingsway is a toll tunnel of mainly local significance carrying only very few vehicles after midnight (during the weekday hours from midnight to 05:00 a.m., a mean traffic frequency of only 68 vehicles per hour was recorded, with a HDV share of 12%). In contrast, the Plabutsch tunnel exhibits besides local traffic also long-distance goods traffic carried out by HDV, clearly visible in higher emission rates. Traffic frequency is with 178 vehicles per nighttime hour approximately 2.5 times higher than in the Kingsway tunnel, and the HDV share is 38%.

Another important distinguishing factor in the exhaust air of the Plabutsch tunnel is the detection of a substantial amount of nucleation mode particles especially during nighttime. Although the maximum spectral particle number concentrations recorded in both tunnels were in the same range, a much higher average total number concentration was measured in the Plabutsch tunnel.

A further characterization of the two tunnels can be done by a closer examination of the particle size spectra for three

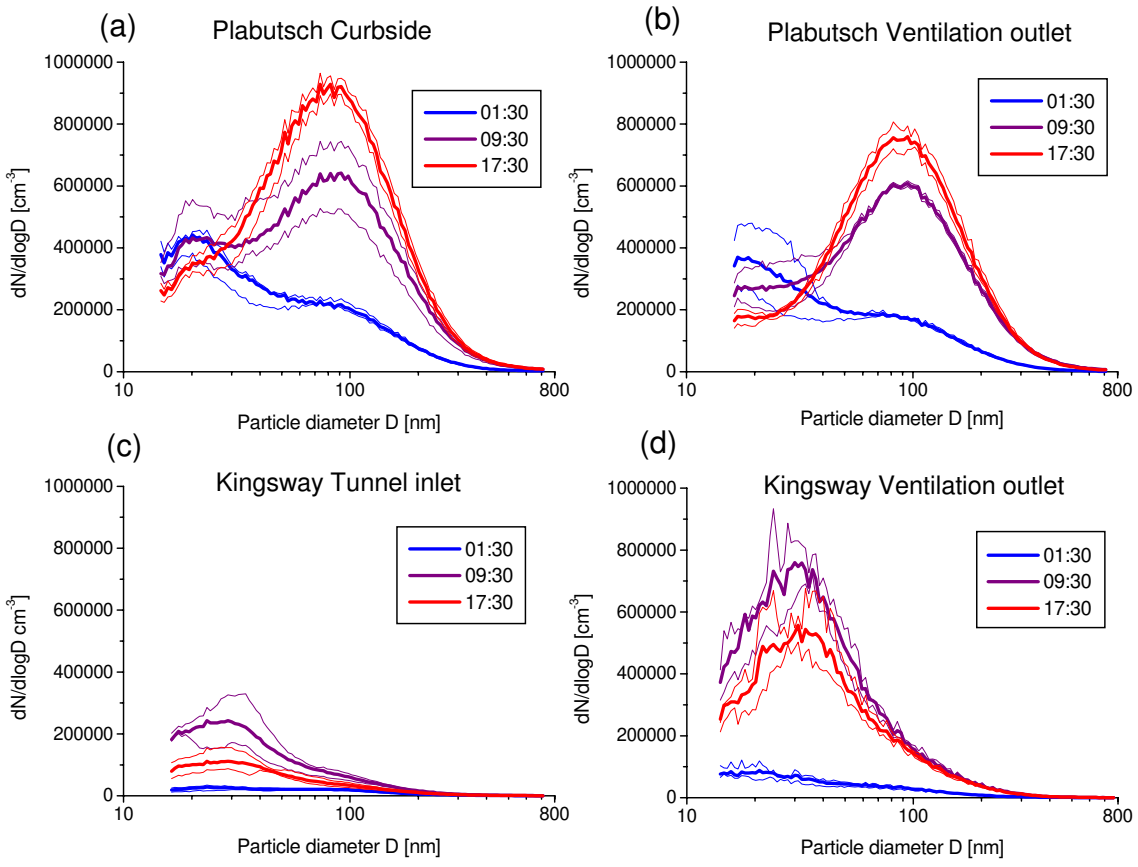

Fig. 4. Particle size distribution as a function of time of day for the Plabutsch (**a** and **b**) and Kingsway tunnel (**c** and **d**). Thick lines are mean values of all weekday measurements in the respective time period, thin lines represent 25 and 75 percentile values.

specific time periods (Fig. 4). For this purpose, one-hour mean values together with the 25 and 75 percentiles were calculated from all available weekdays for the following time periods: A night period from 01:00 to 02:00 h with very low traffic frequency, a morning period from 09:00 to 10:00 h after the morning rush hour with a typical daytime vehicle frequency and an evening period from 17:00 to 18:00 h during the evening rush hour with the highest traffic frequency.

In Figs. 4a and b the size spectra of the Plabutsch tunnel at the curbside and at the outlet of the ventilation shaft are compared to each other. Number size distributions measured at the ventilation outlet show a slightly lower concentration, due to coagulation in the ventilation stack. All spectra exhibit a clear bimodal structure. The nucleation mode particles have a maximum concentration at diameters of around 20 nm, while the soot mode particles were observed in the diameter range of 70 to 100 nm. During daytime, the size distributions in the Plabutsch tunnel reveal a distinct soot mode, whereas during the night it is still present but much smaller. In contrast, the nucleation mode behaves inversely, with lower concentrations during the day than during the night. This is explained by the fact that during very high traffic density semivolatile material condenses on the large surface area of the pre-existing soot particles. If there is only a small surface area available, which is the case during low traffic frequencies in the night, new particles are formed by homogeneous nucleation. The high volatility of these particles was verified by Baltensperger et al. (2002), while the behaviour of non-volatile traffic-related particles was investigated by Wehner et al. (2004).

For the Kingsway tunnel, number size distributions for the equivalent time periods recorded at the tunnel entrance and at the ventilation outlet are depicted in Figs. 4c and d. The sampling site 1 is located near the entrance and therefore shows the initial pollution level after 50 m driving in the tunnel being strongly increased compared to the concentration of the ventilation inlet (not shown). Maximum concentrations were found in the nucleation mode at diameters $D \approx 30$ nm. The soot mode is clearly seen at the tunnel entrance but almost not recognizable in the exhaust air, because there it is hidden by the large amount of nucleation mode particles, resulting in an almost unimodal distribution. In this tunnel, the concentration of the evening period is lower than that in the morning because measurements took place in the bore with traffic in the direction to Liverpool, which is not the main track of the evening rush hour traffic. The relatively high

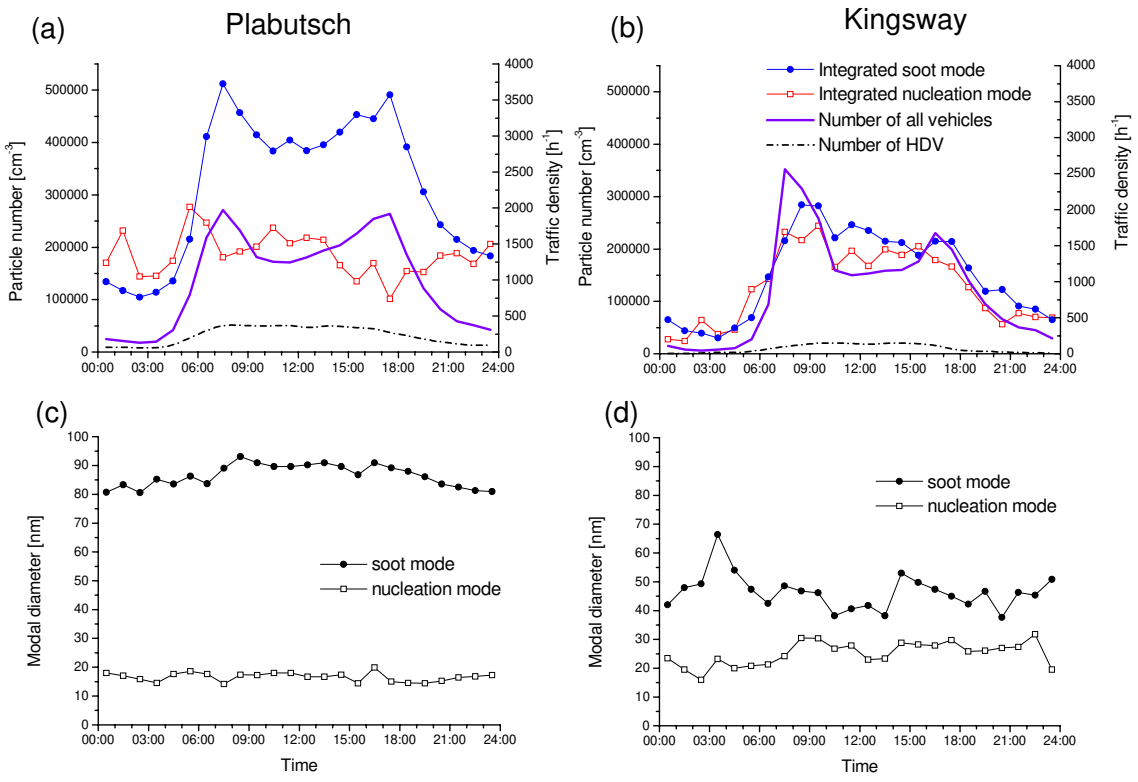

Fig. 5. Average diurnal variation of the number concentrations of nucleation mode and soot mode particles at the ventilation outlet of the Plabutsch **(a)** and Kingsway **(b)** tunnel. The diurnal variation of the median diameter of these two modes is displayed in **(c)** and **(d)**.

percentile range demonstrates a large fluctuation of these nucleation mode particles, in particular at the tunnel entrance. However, at nighttime a behaviour analogous to that of the Plabutsch tunnel was observed, although to a smaller extent. In case of very low soot particle concentrations, the modal diameter of the nucleation particles is shifted to smaller diameters around 20 nm.

Results of the bimodal fitting of the particle size distributions for the ventilation outlet are presented in Fig. 5, aggregated to average diurnal variations of all measured weekdays. Owing to the above mentioned clear bimodal distribution, the fit uncertainties for the Plabutsch tunnel are small. In this tunnel, the integrated particle number concentration exactly follows the traffic density curve. In contrast, the nucleation mode number concentration does not show a distinct diurnal cycle. During time periods with very high concentrations of soot mode particles (i.e. around 07:00 to 08:00 a.m. and around 06:00 p.m.), the nucleation mode number concentration reveals a slight decrease compared to the adjacent time periods. The modal diameter of the nucleation mode remains constant at $D \approx 15$–20 nm, whereas the soot mode diameter displays a slight diurnal variation, with smaller diameters ($D \approx 80$ nm) during the night than during daytime ($D \approx 90$ nm). This behaviour is a further indication for the growing of freshly emitted soot particles by coagulation during high soot concentration periods. The σ values expressing the geometric standard deviation of the log-normal distribution were in the same range for both modes: 1.81 ± 0.17 for the nucleation mode and 1.85 ± 0.02 for the soot mode.

As expected from the size spectra, the bimodal fit results of the soot mode number concentrations had a relatively high statistical uncertainty in the Kingsway tunnel. Therefore we fitted the particle surface area distribution where the soot mode was more pronounced and calculated the number concentration and modal diameter from the surface distribution parameters. The same procedure was done for the Plabutsch tunnel, but there the results showed no significant difference compared to those obtained by fitting the number size distributions. In the Kingsway tunnel, the number concentrations of the soot mode and nucleation mode particles correlate rather well with the traffic counts, and both modes are comprised of a similar amount of particles. Though much fewer soot mode particles are observed here than in the Plabutsch tunnel, during daytime the concentration of nucleation mode particles is comparable in both tunnels. In the Kingsway tunnel the median diameters of the two modes did not show a diurnal variation. The average diameter was around 25 nm of the nucleation mode and approximately 45 nm for the soot mode. Here the σ values for the two modes were different: 1.46 ± 0.09 for the nucleation mode and 2.22 ± 0.05 for the

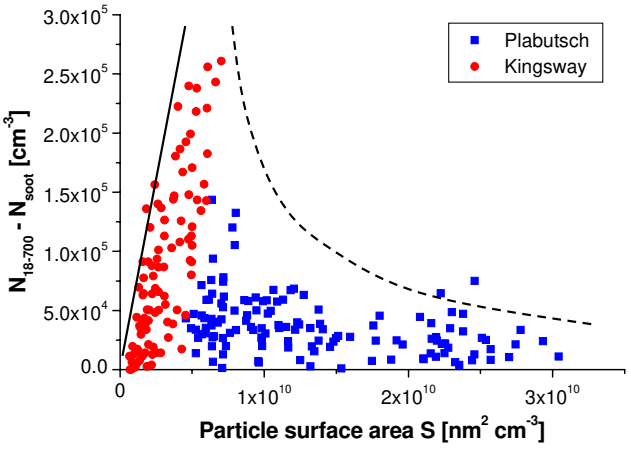

Fig. 6. Scatter plot of the aerosol surface area concentration versus the number concentration of particles in the size range D=18–700 nm subtracted by the number concentration of the soot particles (N_{18-700}–N_{soot}) observed in the two tunnels.

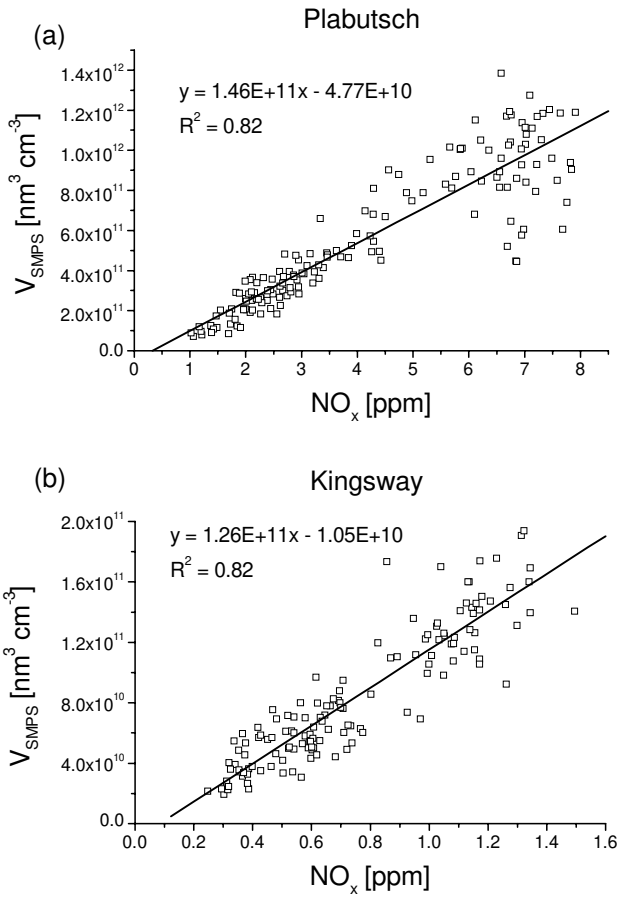

Fig. 7. Correlation between NO_x and the particle volume (V_{SMPS}) in the Plabutsch **(a)** and the Kingsway **(b)** tunnel.

soot mode, indicating a comparatively narrow nucleation and a broad soot mode.

An attempt to clarify the connections between the soot mode particles and the nucleation mode particles is shown in Fig. 6, where the number concentration of particles in the size range D=18–700 nm subtracted by the number concentration of the soot mode particles (N_{18-700}–N_{soot}) is plotted against the surface area concentration of the soot mode particles. This number was preferred over the fitted nucleation mode number concentration because the latter exhibited high uncertainty when the mode was close to the lower limit of the SMPS. The scatter plot of Fig. 6 depicts the different features in the tunnels at the ventilation outlet. In the Kingsway tunnel, much lower soot concentrations were observed than in the Plabutsch tunnel, and in the former the concentration of N_{18-700}–N_{soot} was clearly higher. Two diverse trends are visible: Nucleation mode particle concentrations (D>18 nm) seem to increase linearly with an increase of the surface area of soot particles in the concentration regime of the Kingsway tunnel. In contrast, with a further enhancement of the surface area the number of nucleation mode particles decreases. This phenomenon can be explained by the following facts. Traffic is known to emit both, soot and nucleation mode particles. On the left hand side of the diagram, the slope of the solid line marks empirically the number of condensation particles which is possible together with a certain amount of soot surface area. In this regime, a tunnel study can be expected to yield number emission factors that are representative of most of the real world conditions. At a soot particle surface area concentration of around 5×10^9 nm^2 cm^{-3}, the relationship reaches the ratio of nucleation particles versus aerosol surface area without an influence of the particle surface area on the nucleation mode. A further increase of the soot surface area leads to a preferential condensation of the semivolatile organic material on the already existing surfaces, indicated by the enveloping dashed line. In this regime, tunnel studies cannot be expected to reveal true number emission factors. Furthermore, it needs to be stated that the observed threshold value for the two investigated tunnels may not be valid for tunnel situations in general. Different temperatures and different fuel sulphur content in other tunnels would most probably lead to another threshold value.

3.2 Correlation between NO_x and the total particle volume

Since particle emission factors are sometimes computed using the known NO_x emission factors (Imhof et al., 2005a; Gehrig et al., 2004), the relationship between these two parameters was investigated at the exhaust air of both tunnels. Scatter plots of the total volume of particles in the size range between 18 and 700 nm against the NO_x mixing ratio are depicted in Fig. 7. A significant correlation between these two parameters was observed, the determination coefficient R^2 was 0.82 in both cases. The larger the recorded NO_x mixing ratio was, the higher the submicron particle volume concentration was. Similar correlations were also found between

NO_x and PM10 in the Plabutsch tunnel (R^2=0.87) and between NO_x and PM2.5 in the Kingsway tunnel (R^2=0.81). From the strong relationship between NO_x and the different particle parameters the above approach of calculating particle emission factors based on known NO_x emission factors appears to be justified.

Findings in this paper are consistent with the results from previous studies. Ketzel et al. (2003) report an R^2=0.81 for the particle number concentration (10–700 nm) and the NO_x concentration. Very high correlations between NO_x mixing ratio and the particle surface area concentration (R^2=0.9) were obtained for a traffic aerosol in an open field experiment (Imhof et al., 2005b). Quite high correlations between PM2.5 and NO_x (R^2=0.62) as well as PM10 and NO_x (R^2=0.43) were also reported (Harrison et al., 2001).

3.3 Determination of emission factors

Mean emission factors were determined for particle number, volume and mass as well as for the nitrogen oxides. Particle number and volume, measured by the SMPS systems in the size range between 18 and 700 nm were split into various sections. The terminology was selected as follows: N_x comprises the number of all particles in the size range from 0.018 μm to x μm, and V_x correspondingly includes the volume of the emitted particles in the size range from 0.018 μm to x μm. These values were chosen in analogy to Imhof et al. (2005a).

3.3.1 Method

With the aim to calculate emission factors for different vehicle categories, first the total traffic emissions of particles and NO_x in the investigated tunnel section were computed. Since the tunnels dispose of diverse ventilation systems and traffic conditions, different formulas were applied. For the Kingsway tunnel with unidirectional traffic and passive ventilation which means air moving in the same direction as the vehicles, the following formula was used:

$$EF = \frac{(c_o \cdot v_o + c_{vv} \cdot v_{vv} - c_i \cdot v_i - c_{pv} \cdot v_{pv}) \cdot \Delta t}{n_{\text{tot}} \cdot d} \qquad (1)$$

where: EF: Emission factor per vehicle
c: Concentration of the compound at the measuring points (indices o for outlet, vv for Victoria ventilation station with exhaust air, i for tunnel inlet and pv for Promenade ventilation station with intake air)
v: ventilation rate
Δt : Length of the time interval
n_{tot}: Number of vehicles driving through the tunnel during the time interval Δt
d: Distance between the measuring points

Since no SMPS system was operated at the tunnel outlet, vehicle emissions of particle number and volume represent only the 1670 m long section from the tunnel entrance to Victoria ventilation station with the following longitudinal section (see also Fig. 1): first a downward gradient of around 4%, then an almost even part and at the end an upward gradient of 3.3%. An estimation of the emissions in the last 450 m of the tunnel is given by the two PM1 emission factors in Table 3. PM1 (section) denotes the emissions on the first 1670 m of the tunnel, whereas PM1 (whole) comprises the emissions of the vehicles in the entire tunnel.

For the Plabutsch tunnel, the calculation of the emission factor per vehicle was more complex. There the influence of the already existing air pollution level in the tunnel had to be considered, because polluted air stays longer in the tunnel due to the bidirectional traffic flow and only the additional pollutant contribution is emitted by the current passing vehicle fleet. Therefore the following formula was used (Rodler, 2000):

$$EF = \frac{1}{n_{\text{tot}} \cdot l} \cdot \left(\frac{V_T}{\Delta t} + Q_o \right) \cdot c_t - \frac{V_T}{\Delta t} \cdot c_{t-\Delta t} - Q_i \cdot c_i \qquad (2)$$

where l: length of the ventilation section (1947 m)
V_T: Volume of the tunnel in the ventilation section 3
Q_o: ventilation rate of exhaust air
Q_i: ventilation rate of intake air
c_i: concentration of the intake air
c_t: concentration of the tunnel air
$c_{t-\Delta t}$: concentration at the previous time step

In this equation, the second term ($[V_T/\Delta t + Q_o]c_t$) describes the actual air pollutant concentration in the tunnel, while the third term is the concentration one time step before and the fourth term defines the background concentration fed into the tunnel by the ventilation systems. Air exchange between the ventilation segments is possible, but can be neglected here, because pollutant concentrations of the adjacent tunnel sections are similar to those in the investigated part.

Emission factors were computed according to Eqs. (1) and (2) for the Kingsway and Plabutsch tunnel, respectively, on the basis of one hour values for the British and of half-hour values for the Austrian tunnel. Time periods in which ventilation rates were considerably lower than the normal value were excluded in order to eliminate periods with poor air exchange and insufficient mixing. From the remaining dataset, mean emission factors per vehicle were calculated, and the results are given in Table 3.

By means of statistical regression, mean emission factors for two different vehicle categories were estimated. As a result of rigorous analyses the following regression models were found to give the best accuracy for each tunnel:

For the Kingsway tunnel:

$$EF = a \cdot nLDV + b \cdot nHDV + \varepsilon \qquad (3)$$

where *nLDV* is the number of light-duty vehicles, *nHDV* the number of heavy-duty vehicles, a the emission factor for LDV, b the emission factor for HDV, and ε a random error.

Table 3. Emission factors per vehicle for different aerosol parameters as well as for NO_x. CI: Confidence interval. The confidence intervals calculated for the emission factors of the total fleet in the Plabutsch tunnel were not corrected for the effect of the serial correlation and are thus somewhat underestimated.

Plabutsch							
Parameter	Unit	Total fleet	(95%CI)	LDV	(95%CI)	HDV	(95%CI)
N0.05	$10^{12}\,km^{-1}$	59	7	13	6	155	22
N0.1	$10^{12}\,km^{-1}$	106	8	37	8	278	30
N0.3	$10^{12}\,km^{-1}$	148	8	59	9	386	31
N0.7	$10^{12}\,km^{-1}$	150	8	59	9	394	31
V0.05	$10^{-3}\,cm^3\,km^{-1}$	1.1	0.1	0.3	0.1	3.1	0.4
V0.1	$10^{-3}\,cm^3\,km^{-1}$	12.0	0.4	6.7	0.6	27	2
V0.3	$10^{-3}\,cm^3\,km^{-1}$	116	3	58	6	285	21
V0.7	$10^{-3}\,cm^3\,km^{-1}$	209	8	72	13	612	48
PM1	$mg\,km^{-1}$	104	4	36	6	306	24
PM10	$mg\,km^{-1}$	233	12	–	–	944	75
NO_x	$mg\,km^{-1}$	3260	150	215	81	12 600	400
Kingsway							
Parameter	Unit	Total fleet	(95%CI)	LDV	(95%CI)	HDV	(95%CI)
N0.05	$10^{12}\,km^{-1}$	95	8	39	9	544	105
N0.1	$10^{12}\,km^{-1}$	118	9	53	10	652	119
N0.3	$10^{12}\,km^{-1}$	126	10	59	11	684	124
N0.7	$10^{12}\,km^{-1}$	126	10	59	11	684	124
V0.05	$10^{-3}\,cm^3\,km^{-1}$	1.7	0.1	0.8	0.2	9.6	1.8
V0.1	$10^{-3}\,cm^3\,km^{-1}$	6.0	0.4	3.5	0.4	29.6	5.2
V0.3	$10^{-3}\,cm^3\,km^{-1}$	25	3	14	2	127	24
V0.7	$10^{-3}\,cm^3\,km^{-1}$	36	4	19	3	200	38
PM1 section	$mg\,km^{-1}$	18	2	10	2	100	19
PM1 whole	$mg\,km^{-1}$	41	4	21	3	310	39
PM2.5	$mg\,km^{-1}$	49	5	19	6	381	63
NO_x	$mg\,km^{-1}$	1520	300	540	60	10 700	800

And for the Plabutsch tunnel:

$$\log(EF) = \log(\alpha + \beta \cdot pHDV) + \gamma \cdot v + \varepsilon \quad (4)$$

where *pHDV*: proportion of heavy-duty vehicles
$\gamma \cdot v$: description of the influence of the total number of vehicles passing the tunnel
$\alpha \cdot e^{\gamma \cdot v}$: emission factor for light-duty vehicles
$(\alpha+\beta) \cdot e^{\gamma \cdot v}$: emission factor for heavy-duty vehicles
ε: random error

A very similar model was also used in earlier studies treating the emission factors for the gaseous compounds in the Plabutsch tunnel (Colberg et al., 2005; Catone, 2002).

As the air pollutant time series usually exhibit non-stationary behaviour, the effect of the serial correlation of the measurements on the estimation of the standard errors, the degrees of freedom and thus the confidence intervals for the different parameters in the regression models was taken into account. More precisely, the residuals were fitted to autoregressive moving-average models (ARMA) (Wilks, 1995; Von Storch and Zwiers, 1999). While the dataset from the Kingsway tunnel did not show any significant serial correlation, third-order autoregressive models, AR(3), had to be applied to the Plabutsch tunnel. The reasons for these different results are most probably the tunnel characteristics and the shorter time interval selected for the Plabutsch tunnel. Due to the bidirectional traffic flow in this tunnel and consequently a longer residence time of the air in the tunnel, a correlation in the measurements from one time step to the other had to be expected.

Unfortunately PM1 was not collected continuously but only for some time periods. Since PM1 is a commonly known aerosol parameter, we computed this quantity from the volume of the measured SMPS size spectra (18–700 nm) according to the procedure described below. Although there are a large number of particles with $D<18$ nm, these small particles can be neglected when considering the aerosol volume (Imhof et al., 2005a; Morawska et al., 1999). The hourly data of the total volume were then correlated to the available

PM1 values. The Plabutsch and Kingsway tunnel show substantially different concentration levels, as depicted in Fig. 8, but nevertheless the points lie along a linear regression line, with an excellent determination coefficient R^2 of 0.93. The conversion factor corresponds to an effective particle density of 0.5 g cm^{-3}, which is consistent with the values observed by Park et al. (2003) for freshly emitted soot particles. Thus we conclude that it is appropriate to calculate PM1 for the entire measurement period of the tunnel campaigns from the SMPS volume.

3.3.2 Emission factors estimated for the two tunnels

The vehicle emission factors for LDV and HDV estimated by the described procedure and the 95% confidence intervals are listed in Table 3. In the Kingsway tunnel the emission factors for the parameters PM1 (whole), PM2.5 and NO_x represent the whole tunnel length, whereas the number and volume emission factors are valid for the first 1670 m.

A slightly higher emission factor is observed for PM2.5 than for the (calculated) PM1 in the Kingsway tunnel concerning the total vehicle fleet and the HDV, while for the LDV the uncertainty is higher than the difference. These results seem to be reasonable, since the particle mass of vehicle emissions in the size range from 1 μm to 2.5 μm is relatively small. For an urban environment, Gomišček et al. (2004) report a PM1/PM2.5 ratio of 0.83 which is exactly the same value found in this study. It is also important to mention that the different measurement techniques may influence the PM1/PM2.5 ratio. The TEOM instruments recording PM2.5 were operated at a temperature of 40°C, while the PM1 measurements were conducted within the tunnels at ambient temperatures of about 18 to 23°C. The resulting influence on the PM1/PM2.5 ratio is difficult to quantify, but is expected to be small, since the temperature difference between the two mass concentration measurements is not particularly high.

In contrast, PM1 and PM10 emission factors at the Plabutsch tunnel show a difference of about factor 2.3 to 3 for the total vehicle fleet as well as for the HDV. A large difference between PM1 and PM10 was also found in other studies, for example in Gehrig et al. (2004). As mentioned there, PM1 almost exclusively contains vehicle exhaust emissions while the supermicron particle mass comprises abrasion products from tyres and clutches and from braking events. Also abrasion of the road surface and the continuous resuspension of all these particles by the vehicles play an important role (Garg et al., 2000; Abu-Allaban et al., 2003). No PM10 emission factor for LDV in the Plabutsch tunnel is reported in Table 3, since both a low value and a high uncertainty resulted from the regression model (4) in the case of this parameter. Possible reasons for this high uncertainty are the relatively high detection limit of the TEOM combined with the relatively low contribution of LDV to PM10 emissions. In contrast, the PM10 emission factor for HDV was

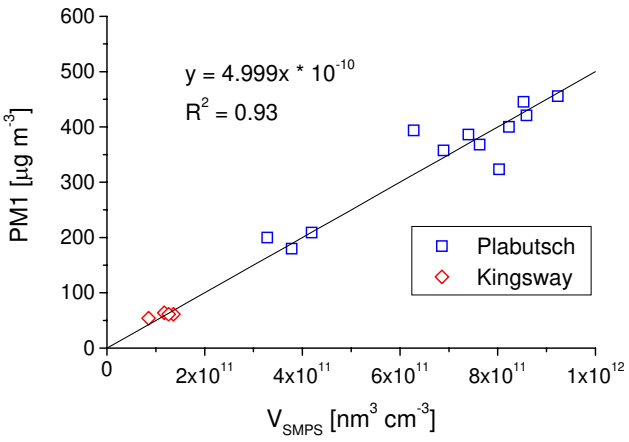

Fig. 8. Correlation between the SMPS particle volume (V_{SMPS}) and PM1 (filter sampling).

obtained with a relatively low 95% confidence interval, i.e. a robust value with low uncertainty.

A significant difference in the PM1 emission factors (parameters "PM1" for Plabutsch and "PM1 whole" for Kingsway) was found for LDV and the total vehicle fleet. Since HDV show nearly the same emission factors regarding PM1, a higher proportion of Diesel passenger cars in the Plabutsch tunnel compared to the Kingsway tunnel might be responsible for the different values found for LDV. In addition, the HDV/LDV ratio in Plabutsch was on average 3–4 times higher than in the Kingsway tunnel. The higher proportion of HDV at Plabutsch can therefore explain the higher emission factor found for the total vehicle fleet in this tunnel.

Particle volume emission factors exhibit a different picture. In the Kingsway tunnel, distinctly higher values were found for the smallest fraction V0.05 than in the Plabutsch tunnel, whereas all the other particle volume fractions were emitted in a higher amount in the Austrian tunnel. Even if a factor of around 2.5 for the last tunnel section is applied, which is permissible as shown in Fig. 8, the values are lower than in the Plabutsch. The reason for this behaviour pattern is most probably the much higher HDV share combined with the approximately 39% of LDV operated by diesel motors in Austria. These two factors overcompensate for the increased emissions by the steeper roadway gradient in the Kingsway tunnel.

The particle number emission factors showed higher values in the Kingsway tunnel than in the Plabutsch, especially for the ultrafine particles with $D<100$ nm. These findings are consistent with the mean size distributions depicted in Fig. 4, where it is displayed that during the day much more particles are emitted in the size range below 50 nm in the Kingsway tunnel. As explained in Sect. 3.1 the number of nucleation mode particles depends on the existing soot surface and on the traffic volume passing the tunnel. Thus a comparatively high uncertainty is expected for the N0.05 emission factors.

In fact, the uncertainty was almost 45% for the LDV and 15% for the HDV regarding this parameter which is considerably higher than for the other number emission factors. Recapitulating, in the presence of an abundant soot surface area the determination of an emission factor for particles with $D<50$ nm proves to be difficult and uncertain.

Results obtained in this study show a fairly good agreement with previous investigations. Average PM10 emission factors in the Plabutsch tunnel for the total vehicle fleet of 233 mg km^{-1} correspond quite well to 285 mg km^{-1} found in the Lundby tunnel (Sweden) with a comparable fleet composition and traffic speed (Sternbeck et al., 2002). The authors also report a PM1 emission factor of 89 mg km^{-1} which is slightly less than observed in the Plabutsch (104 mg km^{-1}). PM2.5 emission factors for different vehicle categories are stated by Kirchstetter et al. (1999) for the Caldecott tunnel (California, vehicle speed 70–90 km h^{-1}). The values of 43 mg km^{-1} for LDV and 987 mg km^{-1} for HDV are higher than the findings in this study, probably due to the steep roadway gradient of 4.2%. Weingartner et al. (1997) give a PM3 emission factor for the Gubrist tunnel (Switzerland) of 8.5 mg km^{-1} (LDV) and 384 mg km^{-1} (HDV). These values are consistent with the PM2.5 emission factors in the Kingsway tunnel and the PM1 emission factors in the Plabutsch tunnel. Even with results obtained by open land experiments and a different calculation method (Gehrig et al., 2004), a good consistency was found.

Only very few studies so far dealt with emission factors of particle numbers and volumes in various size ranges under real world conditions. For urban driving conditions, particle number emission factors of 1.75 to 2.8×10^{14} particles km^{-1} are known for the whole vehicle fleet (Ketzel et al., 2003; Gramotnev et al., 2003; Jamriska et al., 2001). Regarding higher traffic speeds, Kristensson et al. (2004) found 4.6×10^{14} particles km^{-1} for an average speed of 70–90 km h^{-1} in the Swedish Söderleds tunnel. Compared to the N0.7 emission factors of this work, these values are about 1.5 to 4 times higher, but one has to bear in mind that the other studies use a lower cut-off size ($D_{\mathrm{cut-off}}$=10 nm, and even $D_{\mathrm{cut-off}}$=3 nm in the Söderleds tunnel). Therefore a significantly higher particle number can be expected for the mentioned studies. A separation in two vehicle categories was again performed by Kirchstetter et al. (1999). The authors estimated particle number emission factors ($D_{\mathrm{cut-off}}$=10 nm) of 0.41×10^{14} particles km^{-1} for LDV and 25×10^{14} particles km^{-1} for HDV. A good correspondence between these values and those for the Plabutsch and Kingsway tunnel was found for LDV, but the emission factors of the HDV in the Caldecott tunnel were significantly higher. Even higher are the number emission factors ($D_{\mathrm{cut-off}}$=10 nm) derived by Gidhagen et al. (2003): 1.1–5.9×10^{14} particles km^{-1} for LDV and 58×10^{14} particles km^{-1} for HDV.

The differences between these studies can be attributed to various cut-off sizes, to different vehicle fleet composition, traffic speeds and fuel compounds and also to the driving conditions which are specific for each individual study.

4 Conclusions

Emission factors were determined for two highly different tunnels. In the Plabutsch tunnel, the observed concentrations were generally higher than in the Kingsway tunnel. During nighttime a particle number concentration in the size range between 18 and 700 nm of 2.45×10^5 particles cm^{-3} was found in the Austrian tunnel whereas in the Kingsway tunnel only 50 000 particles cm^{-3} were measured. In contrast to that the differences were smaller during the rush hours with 5.4×10^5 particles cm^{-3} (Plabutsch) and 4.4×10^5 particles cm^{-3} (Kingsway) but still evident. The size distributions likewise displayed an obvious discrepancy. While during daytime, in the Plabutsch a distinct soot mode with maximum concentrations around 80–100 nm was observed, the Kingsway tunnel showed high particle concentrations in the nucleation mode with a median modal diameter in the size range D=25–35 nm and comparatively low concentrations in the soot mode. Results from the log-normal bimodal fit revealed that in the Kingsway tunnel, the number of nucleation mode particles increased with the surface area of the soot mode particles. In the Plabutsch the opposite effect was found, an increasing surface area was combined with a decreasing amount of nucleation mode particles. This behaviour occurs at a significantly higher concentration level. The explanation for this feature is the condensation of the semivolatile organic species (forming new particles under relatively low air pollution, i.e. Kingsway tunnel), on the preexisting large surface area in the Plabutsch tunnel.

The differences between the concentrations of soot mode particles in the two tunnels can principally be attributed to the fleet composition and the traffic density. Diesel engines emit a large fraction of soot and nucleation mode particles. In the Plabutsch, a HDV share of 18% combined with another 39% of the LDV also driven by diesel motors are responsible for very high emissions of soot mode particles. In contrast, only 7% of the vehicle fleet which frequented the Kingsway tunnel are HDV. Thus a clearly lower soot particle concentration can be expected.

From the particle and NO_x measurements emission factors were calculated. The findings are consistent with results of other studies. The higher emission factors in the Plabutsch tunnel are explained by the higher traffic speed (Kristensson et al., 2004; Kittelson et al., 2004). The meaningfulness of number emission factors in this range also needs to be assessed. The formation of nucleation mode particles depends on the temperature, the relative humidity and – as shown in this work – on the aerosol surface area concentration. Therefore a strong dependency on the sampling location is expected especially for nanoparticle emission factors.

Concluding, emission factors are always in the same order of magnitude, but they vary from one country to the other due to the share of passenger cars with diesel engines. Furthermore they depend on the conditions at the sampling sites, e.g. on the traffic frequency, the share of HDV, the roadway gradients and on meteorological influences.

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References

Abu-Allaban, M., Coulomb, W., Gertler, W. A., Gillies, J., Pierson, W. R., Rogers, C. F., Sagebiel, J. C., and Tarnay, L.: Exhaust particle size distribution measurements at the Tuscarora Mountain tunnel, Aerosol Sci. Technol., 36, 771–789, 2002.

Abu-Allaban, M., Gillies, J. A., Gertler, A. W., Clayton, R., and Proffitt, D.: Tailpipe resuspended road dust, and brake-wear emission factors from on-road vehicles, Atmos. Environ., 37, 5283–5293, 2003.

Baltensperger, U., Weingartner, E., Burtscher, H., and Keskinen, J.: Dynamic Mass and Surface Area Measurements, in: Aerosol Measurement – Principles, Techniques and Applications, edited by: Baron, P. A. and Willeke, K., 387–418, 2001.

Baltensperger, U., Streit, N., Weingartner, E., Nyeki, S., Prévôt, A. S. H., Van Dingenen, R., Virkkula, A., Putaud, J. P., Even, A., ten Brink, H., Blatter, A., Neftel, A., and Gäggeler, H. W.: Urban and rural aerosol characterization of summer smog events during the PIPAPO field campaign in Milan, Italy, J. Geophys. Res., 107, doi:10.1029/2001JD001292, 2002.

Bukowiecki, N., Dommen, J., Prévôt, A. S. H., Weingartner, E., and Baltensperger, U.: Fine and ultrafine particles in the Zürich (Switzerland) area measured with a mobile laboratory: an assessment of the seasonal and regional variation throughout a year, Atmos. Chem. Phys., 3, 1477–1494, 2003, http://www.atmos-chem-phys.net/3/1477/2003/.

Catone, G.: Schätzung von Emissionsfaktoren für verschiedene Fahrzeugkategorien (Estimation of emission factors for different vehicle categories), Diploma Thesis, Department of Mathematics, ETH Zurich, 2002.

Colberg, C. A., Tona, B., Catone, G., Sangiorgio, C., Stahel, W. A., Sturm, P., and Staehelin, J.: Statistical analysis of the vehicle pollutant emissions derived from several European road tunnel studies, Atmos. Environ., 39, 2499–2511, 2005.

Dockery, D. W. and Pope, C. A.: Acute Respiratory Effects of Particulate Air-Pollution, Annual Review of Public Health, 15, 107–132, 1994.

Donaldson, K., Beswick, P. H., and Gilmour, P. S.: Free radical activity associated with the surface of particles: a unifying factor in determining biological activity?, Toxicology Letters, 88, 293–298, 1998.

Garg, B. D., Cadle, S. H., Mulawa, P. A., Groblicki, P. J., Laroo, C., and Parr, G. A.: Brake wear particulate matter emissions, Environ. Sci. Technol., 34, 4463–4469, 2000.

Gehrig, R., Hill, M., Buchmann, B., Imhof, D., Weingartner, E., and Baltensperger, U.: Separate determination of PM10 emission factors of road traffic for tailpipe emissions and emissions from abrasion and resuspension processes, Int. J. Environ. Poll., 22, 312–325, 2004.

Gidhagen, L., Johansson, C., Ström, J., Kristensson, A., Swietlicki, E., Pirjola, L., and Hansson, H.-C.: Model simulation of ultrafine particles inside a road tunnel, Atmos. Environ., 37, 2023–2036, 2003.

Gomišček, B., Hauck, H., Stopper, S., and Preining, O.: Spatial and temporal variations of PM1, PM2.5, PM10 and particle number concentrations during the AUPHEP-project, Atmos. Environ., 38, 3917–3934, 2004.

Gouriou, F., Morin, J.-P., and Weill, M.-E.: On-road measurements of particle number concentrations and size distributions in urban and tunnel environments, Atmos. Environ., 38, 2831–2840, 2004.

Gramotnev, G., Brown, R., Ristovski, Z., Hitchins, J., and Morawska, L.: Determination of average emission factors for vehicles on a busy road, Atmos. Environ., 37, 465–474, 2003.

Harrison, R. M., Yin, J., Mark, D., Stedman, J., Appleby, R. S., Booker, J., and Moorcroft, S.: Studies of the coarse particle (2.5–10 μm) component in UK urban atmospheres, Atmos. Environ., 35, 3667–3679, 2001.

Haywood, J. M. and Boucher, O.: Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: A review, Rev. Geophys., 38, 513–543, 2000.

Heintzenberg, J.: Properties of the log-normal particle size distribution, Aerosol Sci. Technol., 21, 46–48, 1994.

Imhof, D., Weingartner, E., Ordóñez, C., Gehrig, R., Hill, M., Buchmann, B., and Baltensperger, U.: Real-world emission factors of fine and ultrafine aerosol particles for different traffic situations in Switzerland, Environ. Sci. Technol., 39, 8341–8350, 2005a.

Imhof, D., Weingartner, E., Vogt, U., Dreiseidler, A., Rosenbohm, E., Scheer, V., Vogt, R., Nielsen, O. J., Kurtenbach, R., Corsmeier, U., Kohler, M., and Baltensperger, U.: Vertical distribution of aerosol particles and NO_x close to a motorway, Atmos. Environ., 39, 5710–5721, 2005b.

Jamriska, M. and Morawska, L.: A model for determination of motor vehicle emission factors from on-road measurements with a focus on submicrometer particles, The Science of the Total Environment, 264, 241–255, 2001.

John, C., Friedrich, R., Staehelin, J., Schläpfer, K., and Stahel, W. A.: Comparison of emission factors for road traffic from a tunnel study (Gubrist tunnel, Switzerland) and from emission modeling, Atmos. Environ., 33, 3367–3376, 1999.

Ketzel, M., Wåhlin, P., Berkowicz, R., and Palmgren, F.: Particle and trace gas emission factors under urban driving conditions in Copenhagen based on street and roof-level observations, Atmos. Environ., 37, 2735–2749, 2003.

Ketzel, M., Wåhlin, P., Kristensson, A., Swietlicki, E., Berkowicz, R., Nielsen, O. J., and Palmgren, F.: Particle size distribution and particle mass measurements at urban, near-city and rural level in the Copenhagen area and Southern Sweden, Atmos. Chem.

Phys., 4, 281–292, 2004, http://www.atmos-chem-phys.net/4/281/2004/.

Kirchstetter, T. W., Harley, R. A., Kreisberg, N. M., Stolzenburg, M. R., and Hering, S. V.: On-road measurement of fine particle and nitrogen oxide emissions from light- and heavy-duty motor vehicles, Atmos. Environ., 33, 2955–2968, 1999.

Kittelson, D. B.: Engine and nanoparticles: a review, J. Aerosol Sci., 6, 443–451, 1998.

Kittelson, D. B., Johnson, J., Watts, W., Wei, Q., Drayton, M., Paulsen, D., and Bukowiecki, N.: Diesel aerosol sampling in the atmosphere, SAE Paper No. 2000-01-2122, 2000.

Kittelson, D. B., Watts, W. F., and Johnson, J. P.: Nanoparticle emissions on Minnesota highways, Atmos. Environ., 38, 9–19, 2004.

Kristensson, A., Johansson, C., Westerholm, R., Swietlicki, E., Gidhagen, L., Wideqvist, U., and Vesely, V.: Real-world traffic emission factors of gases and particles measured in a road tunnel in Stockholm, Sweden, Atmos. Environ., 38, 657–673, 2004.

Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, Atmos. Chem. Phys., 5, 715–737, 2005, http://www.atmos-chem-phys.net/5/715/2005/.

McLaren, R., Gertler, A. W., Wittorf, D. N., Belzer, W., Dan, T., and Singleton, D. L.: Real-world measurements of exhaust and evaporative emissions in the Cassiar tunnel predicted by chemical mass balance modeling, Environ. Sci. Technol., 30, 3001–3009, 1996.

Morawska, L., Thomas, S., Jamriska, M., and Johnson, G.: The modality of particle size distributions of environmental aerosols, Atmos. Environ., 33, 4401–4411, 1999.

Oberdörster, G.: Toxicology of ultrafine particles: in vivo studies, Philosophical Transactions of the Royal Society of London A, 358, 2719–2740, 2000.

Park, K., Cao, F., Kittelson, D. B., and McMurray, P. H.: Relationship between particle mass and mobility for diesel exhaust particles, Environ. Sci. Technol., 37, 577–583, 2003.

Pierson, W. R., Gertler, A. W., Robinson, N. F., Sagebiel, J. C., Zielinska, B., Bishop, G. A., Stedman, D. H., Zweidinger, R. B., and Ray, W. D.: Real-world automotive emissions – Summary of studies in the Fort McHenry and Tuscarora Mountain tunnels, Atmos. Environ., 30, 2233–2256, 1996.

Pope, C. A., Burnett, R. T., Thun, M. J., Calle, E. E., Krewski, D., Ito, K., and Thurston, G. D.: Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution, Jama – Journal of the American Medical Association, 287, 1132–1141, 2002.

Robinson, N. F., Pierson, W. R., Gertler, A. W., and Sagebiel, J. C.: Comparison of MOBILE4.1 and MOBILE5 predictions with measurements of vehicle emission factors in Fort McHenry and Tuscarora Mountain tunnels, Atmos. Environ., 30, 2257–2267, 1996.

Rodler, J.: Luftschadstoffuntersuchungen mit unterschiedlichen Messsystemen zur Validierung von Emissions- und Ausbreitungsmodellen, Dissertation, Technical University of Graz, Graz, Austria, 2000.

Sternbeck, J., Sjödin, Å., and Andréasson, K.: Metal emissions from road traffic and the influence of resuspension – results from two tunnel studies, Atmos. Environ., 36, 4735–4744, 2002.

Sturm, P. J., Baltensperger, U., Bacher, M., Lechner, B., Hausberger, S., Heiden, B., Imhof, D., Weingartner, E., Prévôt, A. S. H., Kurtenbach, R., and Wiesen, P.: Roadside measurements of particulate matter size distribution, Atmos. Environ., 37, 5273–5281, 2003.

Tobias, H. J., Beving, D. E., Ziemann, P. J., Sakurai, H., Zuk, M., McMurry, P. H., Zarling, D., Waytulonis, R., and Kittelson, D. B.: Chemical analysis of diesel engine nanoparticles using a nano-DMA/thermal desorption particle beam mass spectrometer, Environ. Sci. Technol., 35, 2233–2243, 2001.

Von Storch, H. and Zwiers, F. W.: Statistical Analysis in Climate Research, Cambridge University Press, 1999.

Wehner, B., Philippin, S., Wiedensohler, A., Scheer, V., and Vogt, R.: Variability of non-volatile fractions of atmospheric aerosol particles with traffic influence, Atmos. Environ., 38, 6081–6090, 2004.

Weingartner, E., Keller, C., Stahel, W. A., Burtscher, H., and Baltensperger, U.: Aerosol emission in a road tunnel, Atmos. Environ., 31, 451–462, 1997.

Wilks, D. S.: Statistical Methods in the Atmospheric Sciences, Academic Press, San Diego, 1995.