Atmospheric Environment 96 (2014) 37–49

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Measuring and modelling the local-scale spatio-temporal variation of urban particle number size distributions and black carbon

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HIGHLIGHTS

- Local-scale urban measurements of particles and black carbon.
- Measurements were categorised into different outdoor microenvironments.
- Particle number size distributions were modelled by a multiple regression approach.
- Two models with different input parameters.
- Best agreement for near-traffic microenvironments.

ARTICLE INFO

Article history:
Received 22 January 2014
Received in revised form
3 July 2014
Accepted 7 July 2014
Available online 8 July 2014

Keywords:
Aerosol
Ultrafine particles
Microenvironment
Modelling
Multiple regression
Mobile measurement
Lung deposited surface area

ABSTRACT

Mobile measurements were performed to study the spatio-temporal variation of particle number size distributions (NSD) in the range $11 < D_p < 365$ nm as well as total particle number and black carbon concentrations in Braunschweig, Germany during the winter and summer period 2012/2013. The study area of about 1 km^2 consisted of six different outdoor microenvironments (ME) that were classified according to different traffic intensities and dominant land use types along the measurement route.

Highest averaged total number concentrations measured at roadside (RO) were 2.5×10^4 pt cm^{-3} (with a maximum of 7.6×10^4 pt cm^{-3}) during winter and about 1.2×10^4 pt cm^{-3} on average during the summer campaign. Measurement spots which are more distant to traffic were characterised by lower concentrations of 1.6×10^4 pt cm^{-3} and 9.0×10^3 pt cm^{-3} during winter and summer, respectively. Black carbon (BC) concentrations were also clearly related to traffic emissions and resulted in concentrations of 2.8 μg m^{-3} on average (absolute maximum of 6.2 μg m^{-3}) at RO-sites. The concentrations of particles and BC in the different ME (aggregated from the single measurement spots) documented the concentration of both metrics to be a function of distance of the measurement to fresh traffic emissions.

A multiple regression based model was established to identify significant parameters which can be used to model the microscale variation of particle NSD in the outdoor ME. Two models with different numbers of input parameters were calculated. The first contained all measured parameters as input, the second only a reduced number consisting of TNC, BC and wind speed. Both models worked convincingly, even the approach with the limited number of input parameters. The average size integrated (TNC) deviation to observed data in all ME during both seasons was <13%. The best agreement between model and observations is given for the near-traffic ME.

1. Introduction

Urban areas are hot spots of particulate air pollution due to the large number of stationary and mobile emission sources (Kumar et al., 2010; Weber et al., 2013). A significant fraction of the total urban particle number concentration, e.g. more than 80%–90%, is observed in the ultrafine size range with diameters <100 nm (Morawska et al., 2008). These particles are reported to be particularly relevant for human health due to their ability to penetrate deep into the airways after inhalation (Oberdörster and Utell, 2002; Knibbs et al., 2011). Road traffic is one of the major sources of ultrafine particles in urban air (Morawska et al., 2008). However, the

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http://dx.doi.org/10.1016/j.atmosenv.2014.07.020
1352-2310/

exposure of the urban population to airborne pollutants is characterised by a high spatio-temporal variation on the urban scale (e.g. Dons et al., 2014; von Bismarck-Osten et al., 2013).

Particles in urban environments are emitted as primary particles mainly from combustion processes (road traffic, district heating) or they are formed as secondary particles from precursor gases. Atmospheric transformation of particles by condensation, coagulation or dilution plays an important role in altering the aerosol number size distribution (NSD) between the point of emission and the receptor site. Again this is particularly evident in the ultrafine size range of the size spectrum since these particles are strongly related to fresh emission from traffic and their concentration rapidly decreases with increasing distance from roads (e.g. Hagler et al., 2009; Weber, 2009). Krudysz et al. (2009) and Moore et al. (2009) observed a homogeneous behaviour of the intra-urban number concentration of particles >70–80 nm in the size spectrum since these mainly consist of aged and long-range transported aerosol, while the ultrafine size fraction was characterised by high spatial heterogeneity on the urban scale due to their close local relation to fresh traffic emission on roads.

Due to the significant spatial gradients of urban particle number concentrations a single measurement site is non-representative to assess the exposure of a larger group of urban residents. This especially holds, the more the particle type of emission and the source strength varies across a study area. In exposure and epidemiological research the concept of 'microenvironments' (ME) is a possibility to cope with the spatial gradients of size-dependent particle concentrations. ME are defined as microscale areas with quasi-homogeneous concentrations which can be used to derive characteristic average concentrations or to assess the exposure of people due to different time–activity profiles (e.g. Ott et al., 2007).

A couple of studies to derive characteristic concentrations in different ME were conducted recently, measuring average concentrations in transport microenvironments, i.e. walking, cycling, car (Kaur et al., 2006, 2007; Weichenthal et al., 2008; Boogaard et al., 2009), in different urban microenvironments (Pirjola et al., 2012) or within urban street canyons (Weber, 2009; Buonanno et al., 2011).

Moreover, efforts were taken recently to evaluate the most adequate pollutant metrics that accurately characterise traffic-related particle emissions in urban areas. There is evidence that the combination of particle number and black carbon concentrations is a promising approach to assess the spatio-temporal behaviour of traffic related particle (Rodriguez and Cuevas, 2007; Dall'Osto et al., 2013). Reche et al. (2011) argue that the estimation of particle number concentrations alone is not sufficient to accurately point to traffic related emission, since high number concentrations of particles can also be related to events of new particle formation (cf. Wehner et al., 2007; von Bismarck-Osten et al., 2013). For exposure assessment it is of general importance to analyse how particulate pollutants are influenced by atmospheric conditions. von Bismarck-Osten et al. (2013) recently quantified the influence of meteorological conditions on particle NSD at different types of urban measurement sites. They pointed to the importance of wind (speed and direction), temperature and solar radiation as the most important parameters on average concentrations.

The present study aims to combine simultaneous mobile measurements of particle number and black carbon concentrations on the urban local-scale in order to study the spatio-temporal variation of both metrics and analyse their relation to traffic-related particle emissions. Aerosol number size distributions were measured simultaneously since spatial differences in the particle size spectrum and its statistical properties (modal diameters, concentration) can offer information about the type, the origin and atmospheric transformation processes of particles (cf. Wegner et al., 2012; von Bismarck-Osten et al., 2013; Hussein et al., 2014). A regression based model incorporating pollutant and meteorological input data is developed to calculate particle number size distributions in different urban ME.

2. Materials and methods

2.1. Study area

To study the spatio-temporal variation of particles in an urban area NSD, total particle number and black carbon concentrations were measured between January and August 2013 in the city of Braunschweig, Germany.

The study area had a size of 1 km^2 and was divided into 35 regular grid cells whose midpoints represented the sampling sites (Fig. 1). The cells close to the main road were subclassified in order to have additional spatial information about concentrations with increasing distance to traffic. To achieve a compromise between a high spatial resolution on a regular grid while keeping the measurement time of a specific mobile measurement tour as short as possible (to minimise trends of particle concentration on the diurnal course) the amount of sampling spots was reduced to 31 (excluded cells are shown as white boxes marked with 'x', Fig. 1). The setup of grid-cell based measurements was established to realise equidistant measurement spots, e.g. to study concentrations with respect to distance to roads and other sampling sites. However, this specific aspect will be subject to future studies.

The sampling sites were established in different local outdoor ME which were classified as a function of traffic intensity, road type and type of land use of the surrounding area. Hence, the study area was represented by six outdoor ME (Table 1). The ME are useful to combine single measurements to characteristic urban concentrations and number size distributions (cf. Section 1).

Roadside measurement spots (RO), side road spots with medium and low traffic intensity (SM, SL) are situated within urban street canyons with average traffic intensities between <10,000 veh. day^{-1} (SL) and >30,000 veh. day^{-1} (RO). Residential areas (RE) with limited traffic intensity are located within neighbourhoods (east and west of the main road 'Hagenring', Fig. 1) which are dominated by a distinct fraction of green within the streets. Backyard sites (BY) are situated within residential areas and are characterised by low to very limited traffic activity. These sites are normally sheltered from direct traffic influence by buildings and were reported to be characterised by significantly lower particle concentrations in comparison to main roads, even when only separated by a short horizontal distance of about 50 m (Weber and Weber, 2008). Additionally, urban parks (PA) are characterised as ME with very limited to virtually none traffic activity situated in vegetated areas.

2.2. Instrumentation and data handling

The mobile measurements were performed with a bicycle connected to a 2-wheel trailer which contained the instruments in a weather protected aluminium box, the 'AerosolBox'. The AerosolBox contained three instruments which measured different metrics of air pollution. The sampling air inlet was situated at the top of the trailer at a height of 1.6 m above ground level. Inside the AerosolBox three separate instrument inlets (stainless steel, inner diameter 4 mm) were connected to the main inlet.

Particle NSD were measured with a mobile scanning mobility particle sizer (SMPS), the NanoScan (TSI 3910, TSI Inc., cf. Tritscher et al., 2013). It measures the NSD over the size range $11 < D_p < 365$ nm with a resolution of 13 size bins. The NanoScan

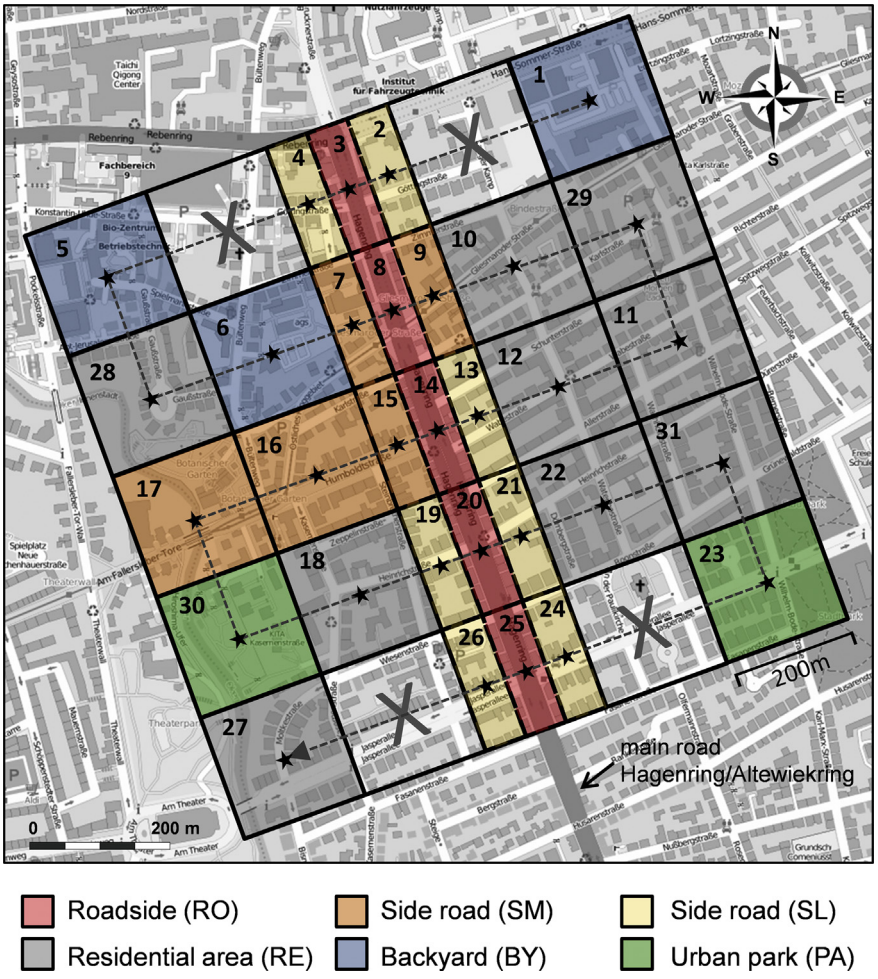

Fig. 1. Overview of the study area and the measurement sites in different urban outdoor microenvironments (coloured boxes). The numbers refer to specific measurement sites (sectors which are marked with 'x' were not considered). The mobile measurement routes started in the uppermost right gridcell (1) and ended in the bottommost left cell (27). The star symbols indicate the midpoint (sampling point) of each ME. Map base: OpenStreetMap – Germany, 2013. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

device is fully battery powered and operates with a fixed 1-min scan time for a full size distribution. A diffusion dryer was used in the sheath air loop of the Nanoscan-SPMS in order to minimize the influence of particle growth under conditions of high relative humidity.

Total number concentrations (TNC), mean size of the particle distribution and alveolar lung deposited surface area (LDSA) of particles were measured with an Electrical Diffusion Size Classifier (DiSCmini, MatterAerosol). LDSA is a very promising metric in aerosol health effects studies, since particle surface area is reported to be highly associated to different respiratory effects (Asbach et al., 2009; Fierz et al., 2011). The DiSCmini is a battery operated hand-held sensor detecting particles in the size range of about $10 < D_p < 500$ nm according to the manufacturer. The DiSCmini sampled with a flow rate of 1.0 L min^{-1} at a time resolution of 1 s. The sensor was tested in several studies and showed reproducible results and sufficient agreement (e.g. <30% deviation in number concentration measurement) with condensation particle counters and SMPS-systems (Meier et al., 2013; Kaminski et al., 2013).

Black carbon concentrations (BC) in the PM_1 size fraction were measured by the single wavelength Aethalometer AE51 (Magee Scientific), which uses optical analysis to determine the mass concentration of BC particles collected from the air stream passing through a PTFE-coated borosilicate-fibreglass filter. The wavelength is 880 nm in the IR range. The device is small and portable and fully battery operated. The flow rate and time resolution was set to 0.05 L m^{-1} and 1 min respectively, which is suited for mobile measurements (Hagler et al., 2011). The AE51 was successfully used in several studies to quantify BC concentrations in different urban environments (Buonanno et al., 2011; Dekoninck et al., 2013; Dons et al., 2013; Ning et al., 2013; Van Poppel et al., 2013).

Meteorological parameters i.e. air temperature (Ta), relative humidity (RH), wind speed (WS) and wind direction (WD) were taken from a meteorological station of the German weather service (DWD) that is situated at a distance of 3 km NW off the study area. The data was used to characterise ambient weather conditions. Ta and WD were measured at 2 m and 10 m height above ground level, respectively. All parameters were available as 1-h averages.

To ensure a high accuracy and data quality all instruments were synchronised before each measurement tour. Due to the different time resolutions of the instruments a 3-min measurement was performed at every sampling spot and the data were aggregated to 3-min averages for subsequent analysis.

Additionally, all datasets passed checks to identify unreasonable concentrations and missing data. Suspicious data (non-plausible values, i.e. >5 standard deviations of single values compared to the spatial average of a measurement day) were then excluded from subsequent analysis. The data availability at the sites was 93% for winter and 99% for summer. To characterise the spatial variation of air pollution parameters the measurements were performed during

Table 1
Overview of different sampling site types and microenvironments. 'Percentage ME' indicates the frequency of occurrence of each ME in the study area.

ME		Road type	ADT[a] [veh. d^{-1}]	Dominant land use	Percentage ME [%]
Roadside	RO	4-lanes	>30,000	Commercial, residential	16
Side road (medium traffic)	SM	2-lanes	>10,000	Commercial, residential	16
Side road (low traffic)	SL	2-lanes	<10,000	Residential	23
Residential area	RE	local street	<2000	Residential	29
Backyard	BY	–	–	Residential	10
Urban park	PA	–	–	Green space	6

[a] Average daily traffic based on city of Braunschweig authorities.

similar weather conditions, i.e. on clear and calm days with wind speeds $\leq$4 m s^{-1} at 10 m above ground level. A total of 16 measurements were conducted, 9 in winter and 7 during the summer period. Each measurement tour took about 2 h to minimize trends of particle concentrations on the diurnal course.

2.3. Data quality

To check for comparability and reproducibility of the mobile devices they were subject to comparison measurements against laboratory reference systems prior to the first measurement campaign in winter 2012/2013 at the Leibniz Institute for Tropospheric Research (TROPOS) in Leipzig, Germany. The NanoScan SMPS was compared to a commercial SMPS System (Classifier 3080, DMA 3081 and CPC 3789, TSI, Inc) and a custom-built differential mobility particle sizer (TROPOS) over a couple of days. Simultaneously the AE51 was compared to a MAAP (Multi Angle Absorption Spectrometer, Thermo Electro Corporation) over one week in February 2013 in ambient air. The filter strips of the AE51 were changed regularly every 2 days as is recommended for this setup in order to ensure a high data quality.

The results of AE51 compared to the MAAP showed an underestimation of about 22% on average of BC mass concentrations measured with the AE51 ($y = 0.78x + 53.3$, $R^2 = 0.83$, BC in ng m^{-3}). The NanoScan SMPS and the SMPS (3969, TSI, Inc.) showed sufficient agreement for TNC ($y = 0.94x + 274.8$, $R^2 = 0.93$, TNC in pt cm^{-3}).

Additional comparison measurements in our Braunschweig laboratory were performed before and during the measurement campaign to ensure the instruments, especially the number concentration measurements of the NanoScan SMPS and DiSCmini, to work properly. The measurements were performed with indoor and ambient air (data not shown here) over at least 16 h. Deviations between both instruments for TNC were in the order of about 10%–15%, respectively (Fig. 2). The comparison measurements indicated sufficient comparability and reproducibility of the mobile instrumentation. Since observed deviations between mobile and reference devices are in a range than can usually be observed in particle sizing and counting instrumentation (e.g. Asbach et al., 2009, 2012) we decided to not calculate any correction factors to adjust the mobile data to laboratory instruments.

2.4. Modelling particle number size distributions – multiple linear regression approach

A multiple linear regression analysis was used to model the particle NSD from a combination of pollutant and meteorological input parameters in the outdoor ME. In comparable statistical modelling approaches only total number concentrations or specific

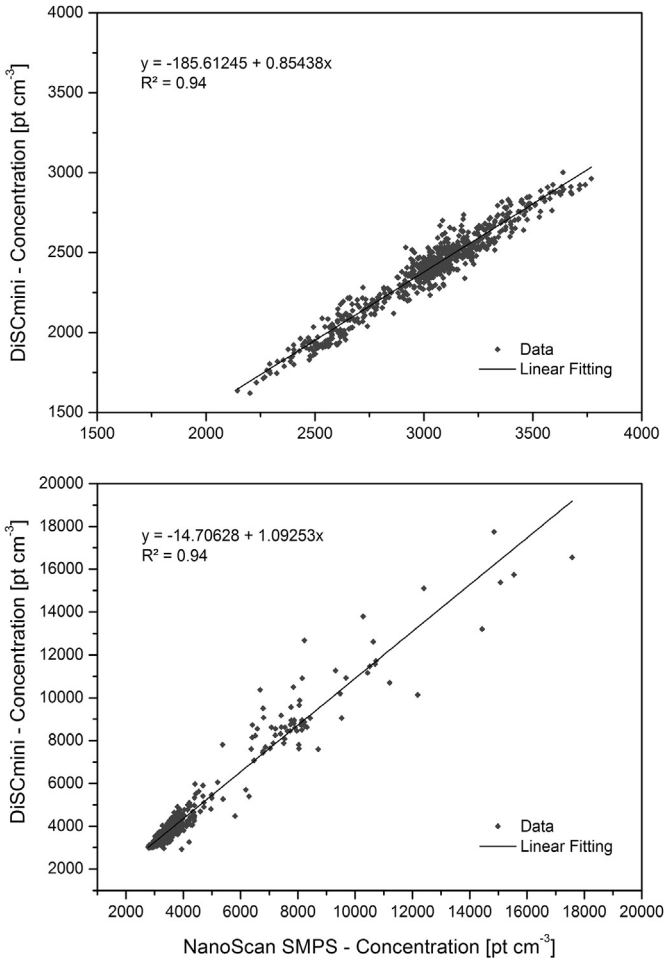

Fig. 2. Laboratory comparison measurement between DiSCmini and NanoScan SMPS.Top: indoor air over a period of 16 h ($n = 980$) Bottom: ambient air over a period of 12 h ($n = 705$).

size integrated parts of the size spectrum (e.g. ultrafine particles, accumulation mode particles) where estimated to date (Clifford et al., 2011; Mølgaard et al., 2012, 2013). The present approach is based on the assumption that the input parameters are linearly related to the NSD. The NSD can then be deduced from

$$\sqrt{y}_{i,j} = ß1_j P1_{i,j} + ß2_j P2_{i,j} \ldots + ßn_j Pn_{i,j}$$

with $\sqrt{y}$ the predicted square rooted particle concentrations in the respective size bin j at point in time i and n is the number of total input parameters. $ß1$–$ßn$ are the regression coefficients for the respective input parameter P1–Pn, derived by the multiple linear regression approach. In this context a constant regression term $ß0$ was not included as the resulting values were tending to zero. The calculation of the regression coefficients was applied to the square root of the measured number concentrations in each size bin to achieve an almost normally distributed variance of the predictands which is one of the desired precondition when performing linear regression in general (Atkinson, 1985). For further details on multiple linear regression the reader is referred to Wilks (2006).

The model was calculated with the full data set from the 16 mobile measurements. To quantify the relationship of each parameter with the NSD the relative influence was calculated on the basis of standardised regression coefficients according to the procedure reported in von Bismarck-Osten et al. (2013).

To study the contribution of different meteorological and air pollution parameters two models were established. The first (Model 1) contained all available air pollution and meteorological parameters (TNC, BC, Size, LDSA, Ta, WS, RH and WD) whereas the second (Model 2) contained a reduced amount of the three most significant parameters (TNC, BC and WS), i.e. the parameters with the largest standardised regression coefficients. Model 2 does not cover information about the particle size of the distribution as is given by the parameters Size and LDSA in Model 1. The aim of Model 2 was to assess the performance of a reduced and easily measurable number of model input parameters to estimate the NSD for each ME. The three selected parameters cover the spatio-temporal relation between TNC and BC while the wind speed data should provide information about the dilution capability of the urban atmosphere.

3. Results and discussion

3.1. Spatio-temporal variation of particle number and black carbon concentrations

In order to analyse the spatio-temporal variation of TNC and BC between specific mobile measurements along the measurement route normalised concentrations were calculated. The normalised concentrations were estimated by dividing the concentration of a pollutant at a specific sampling point by the spatial average of the pollutant concentration for the measurement trip. A general overview of the average ambient meteorological conditions and pollutant concentrations during the measurement trips is presented in Table 2. The measurements were performed during morning and afternoon hours between 09:30–11:00 and 13:00–16:30 local time, respectively (only the morning measurement on 15 January 2013 was conducted partly during the morning rush hour). The aim was to perform measurements mainly outside rush-hours to minimize the impact of high local pollutant concentrations during this time span.

The spatial pattern of TNC along the measurement route is similar during the winter and summer period. Significant peaks occurred at RO whereas RE, BY and PA sites are characterised by lower particle concentrations, e.g. reduced by about 50% on average (Fig. 3). The variation of normalised concentrations of TNC along the measurement route is more distinct during winter than during summer and varies between 0.5–1.6 (winter) and 0.75–1.4 (summer), respectively. The particle concentration decreases with increasing distance of the site to traffic hot spots even in a short distance to the kerbside. Those findings correspond to observations by others (Weber, 2009; Hagler et al., 2009). Highest average particle concentrations close to the main road were 2.5×10^4 pt cm^{-3} in winter. Lowest average particle concentrations were measured at an urban park site with 5.0×10^3 pt cm^{-3} (Fig. 4).

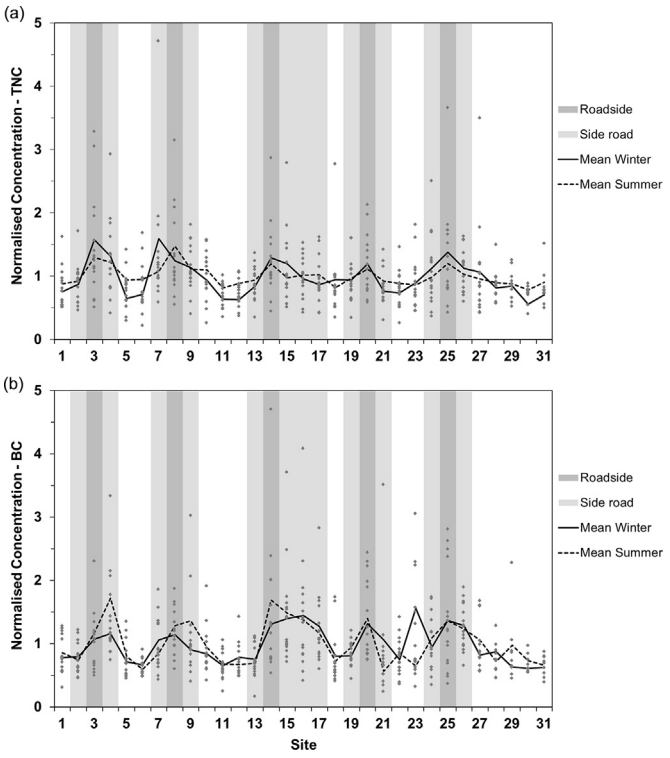

Fig. 3. Variation of normalised TNC (a) and BC (b) concentrations during each measurement tour during the winter and summer campaign in the study area Braunschweig, Germany. Roadside spots are highlighted by grey shaded bars (RO – dark grey, SM and SL – light grey).

Table 2
Measurement tours with start and end time and averages of temperature [°C], relative humidity [%], wind speed [m s^{-1}], wind direction [°], TNC [pt cm^{-3}] and BC [μg m^{-3}].

Date	Measuring period	T [°C]	RH [%]	WS [m s^{-1}]	WD [°]	TNC [pt cm^{-3}]	BC [μg m^{-3}]
11-01-13	10:40–12:50	−0.7	88	3.8	302	12,366	1.0
15-01-13	07:20–09:30	−8.8	92	1.5	164	24,913	4.9
15-01-13	13:00–15:20	−2.5	68	3.7	143	27,558	1.8
27-02-13	13:30–15:40	1.2	90	2.8	66	6814	1.2
28-02-13	09:40–11:40	1.9	87	1.3	251	16,182	2.8
28-02-13	13:00–15:10	3.6	74	3.2	274	13,429	1.3
05-03-13	09:50–12:10	7.7	48	1.6	142	12,617	2.6
06-03-13	10:10–12:20	11.0	48	1.5	113	11,136	2.7
15-03-13	11:10–13:20	−1.9	59	1.9	246	12,679	1.6
07-06-13	10:00–12:20	19.6	68	2.3	64	10,061	1.3
11-06-13	13:20–15:40	19.3	55	2.8	313	9896	1.5
17-06-13	11:30–13:40	20.7	55	4.0	122	13,080	1.0
19-06-13	09:00–11:10	30.0	59	4.0	165	7755	2.2
02-07-13	14:00–16:20	20.7	53	3.0	274	4789	1.1
09-07-13	13:30–15:40	25.7	43	2.4	318	16,465	1.6
17-07-13	10:00–12:20	23.4	54	2.1	349	10,562	1.7

The variation of BC during the winter and summer period shows a similar pattern in comparison to TNC. BC concentrations were significantly related to busy traffic sites (RO), however, differences were less marked in comparison to TNC. The spatial variation of BC is more distinct in summer (0.5–1.8) than in winter (0.6–1.6). The highest site specific average BC concentrations (Fig. 5) were 4.1 μg m^{-3} at a kerbside spot (cf. Fig. 1, site 16) and the lowest were 1.4 μg m^{-3} at an urban park site (cf. Fig. 1, site 30). Pirjola et al. (2012) reported similar values of BC for near traffic sites in their study in Helsinki.

The LDSA and the mean size of the particle size distribution were measured online with the DiSCmini device (cf. Section 2.2). Both metrics show a pattern that indicates that busy traffic sites are influenced by high particle concentrations with small diameter whereas sites with limited or very low traffic intensities are characterised by larger particles and lower LDSA. For RO the lowermost mean sizes were <40 nm and highest LDSA were ~55 μm^2 cm^{-3} (PA: mean size 65 nm and 25 μm^2 cm^{-3}, Figs. 5 and 6). The body of scientific literature on urban measurements of particle surface area concentrations is limited. At a busy freeway in Lisbon, Portugal average weekday LDSA values of up to 89 μm^2 cm^{-3} were measured that decreased to about 35 μm^2 cm^{-3} during weekends (Albuquerque et al., 2012). At a Los Angeles freeway mean LDSA was reported to reach 153 μm^2 cm^{-3} on average while an urban background site (close to downtown Los Angeles) was characterised by

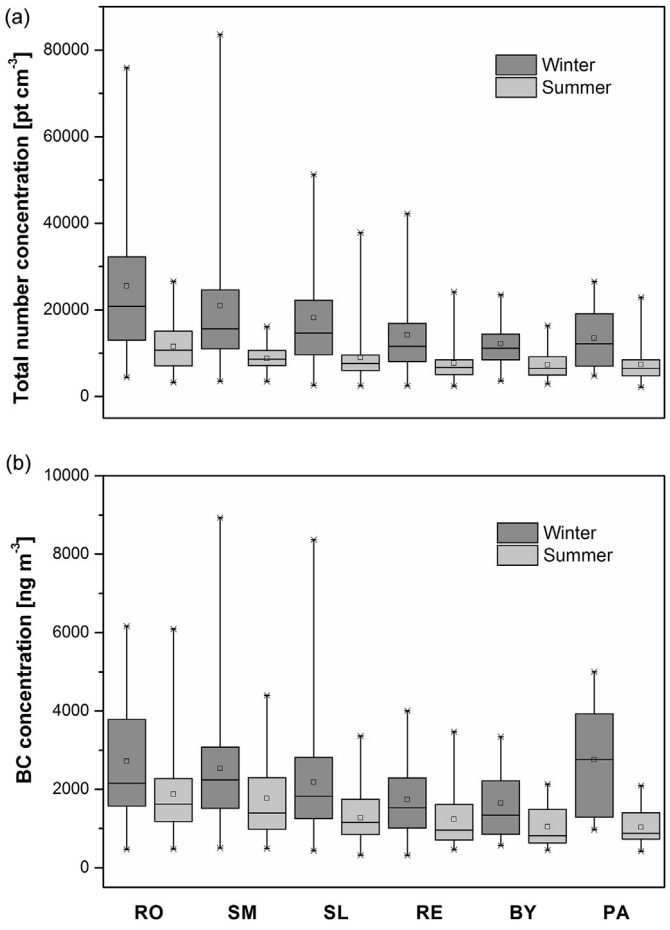

Fig. 4. Variation of averaged TNC (a) and BC (b) concentrations for the different ME during the winter and summer campaign.

average concentrations of about 53 $\mu m^2\ cm^{-3}$ (Ntziachristos et al., 2007). Although not directly comparable the present results at RO are in a similar order of magnitude, even though the traffic intensity (not explicitly documented in the respective studies) is believed to be significantly higher at the freeway sites in the available studies.

The spatial distribution of BC and TNC within the outdoor microenvironments is characterised by higher concentrations in the traffic exposed microenvironments RO, SM and SL with an average of TNC of about 2.5×10^4 pt cm^{-3} (winter) at RO whereas RE, BY and PA are more distant from strong traffic influence and thus show lower concentrations, e.g. 1.3×10^4 pt cm^{-3} (winter) in PA (Fig. 4). Generally TNC follows the rule that concentrations are higher the more the ME is prone to traffic. This is valid also in summer except that the variation between the ME is considerably smaller, pointing to the fact that atmospheric conditions play an important role. Turbulent mixing and dilution in winter is limited due to a higher frequency of stable atmospheric stratification and decreased solar radiation (e.g. Virtanen et al., 2006; Weber, 2009).

The results of BC generally follow the same pattern. However, the urban park PA is characterised by high winter BC concentrations although this ME is characterised by low TNC concentrations and very limited traffic intensity. One reason for this is believed to be coupled to a bus station situated at a distance of 30–50 m to the sampling point within the park. It seems that emission plumes which persisted over a couple of minutes (after the bus has left the station) affected the concentrations at this sampling site. Similar observations of ultrafine particle peak concentrations in the vicinity of a bus stop were reported from mobile measurements by Weichenthal et al. (2008).

A correlation analysis was performed in order to study whether the TNC and BC data were coupled, i.e. showing a similar spatio-temporal variation. Reche et al. (2011) pointed to the combination of both metrics as a valuable tool to study traffic aerosol. In the present study the correlation between TNC and BC was calculated for each ME and plotted onto the measurement grid (Fig. 7). The

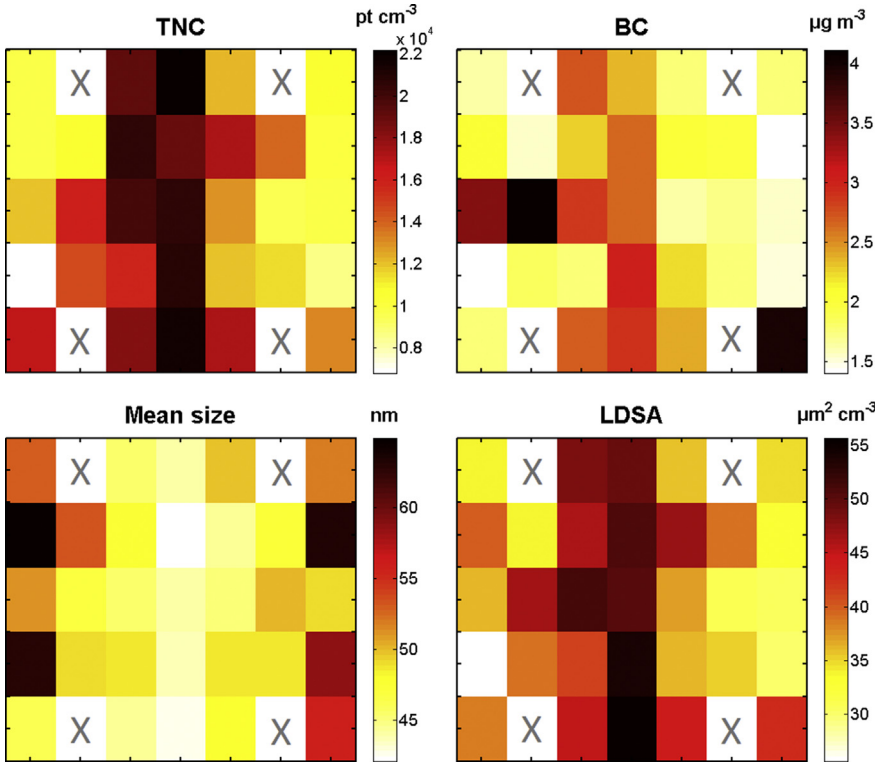

Fig. 5. Spatial distribution of TNC, BC, Size and LDSA for each site during the winter campaign (sectors which are marked with 'x' were not considered).

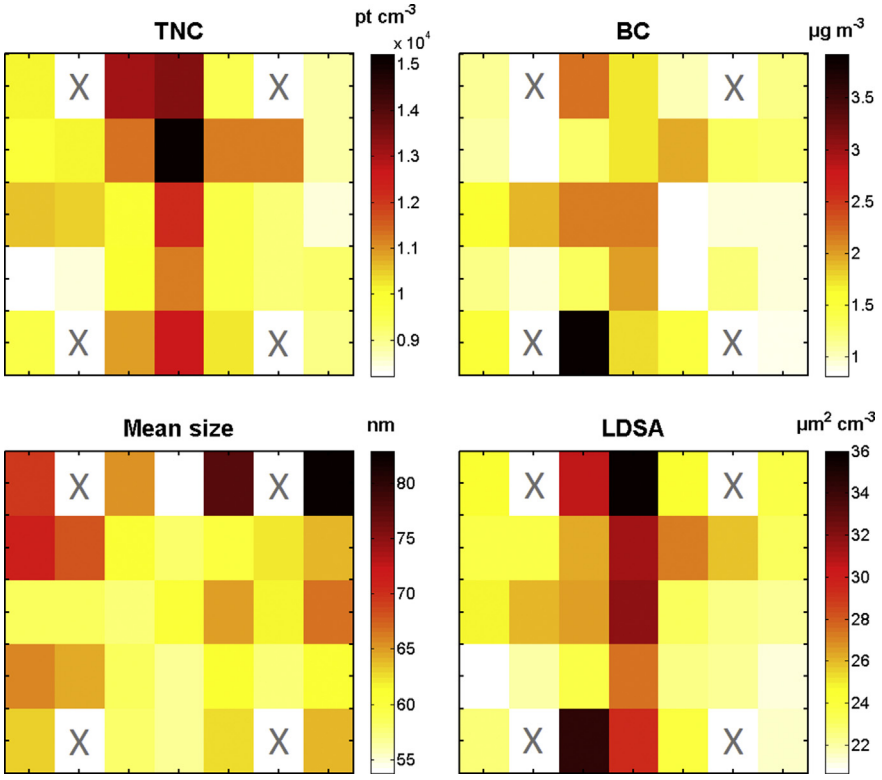

Fig. 6. Spatial distribution of TNC, BC, Size and LDSA for each site during the summer campaign (sectors which are marked with 'x' were not considered).

picture is spatially much clearer for summer where the main road sticks out with correlation coefficients of up to 0.9 ($0.35 < R < 0.9$) while correlation is lower at the other sites ($0.2 < R < 0.4$). In winter the coefficients are higher on average, however, the spatial distribution is more homogeneous, i.e. the highest correlation coefficients still turn up at the main road as is the case during summer, however, the correlation-gradient between main road and surrounding areas is smaller.

In summary, the variation of TNC and BC during winter and summer is clearly related to traffic intensity and meteorological conditions. We believe that in winter, due to the influence of additional emission sources (domestic heating, residential wood burning for heating purposes) and the higher frequency of stable atmospheric stratification, the spatial picture is less confined to the traffic influence as is the case for summer. The effect of wood burning on air pollution especially in residential areas has also been reported by others (Tissari et al., 2008; Bari et al., 2011). This is in contrast to the summer season where traffic emissions are the dominant source of particulate emissions. It results in the marked spatial distribution of the TNC–BC correlation which is especially evident at the main road (Fig. 7). The lower variation of pollutants during summer is coupled to more instable atmospheric conditions.

3.2. Spatio-temporal variability of particle number size distributions

After the spatio-temporal behaviour of particle and black carbon concentrations was studied a deeper look into the spatio-temporal behaviour of the NSD is conducted by analysing the average NSD for each of the six ME for the winter and summer period (Fig. 8).

The ME size distributions were subjected to log-normal mode fitting in order to study the modal characteristics of the number spectra (Fig. 8). The DistFit software was used to fit log-normal modes to the aerosol data (Chimera Technologies, 2009). In urban areas three log normal modes generally offer a good description of

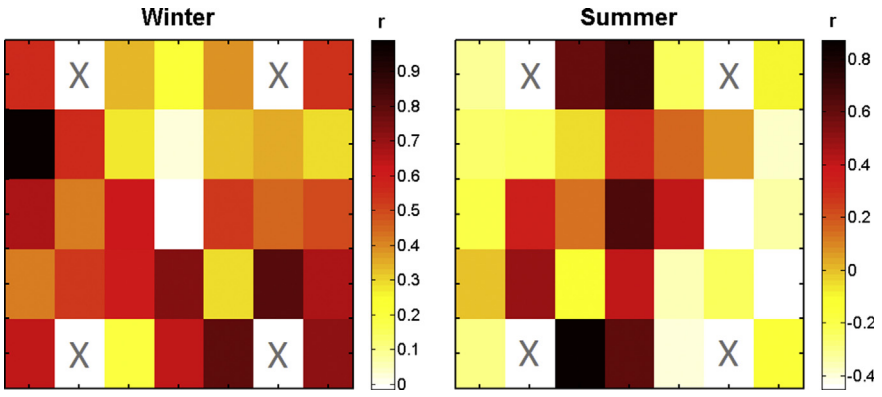

Fig. 7. Spatial correlation between TNC and BC concentrations for each site during the winter and summer campaign (sectors which are marked with 'x' were not considered).

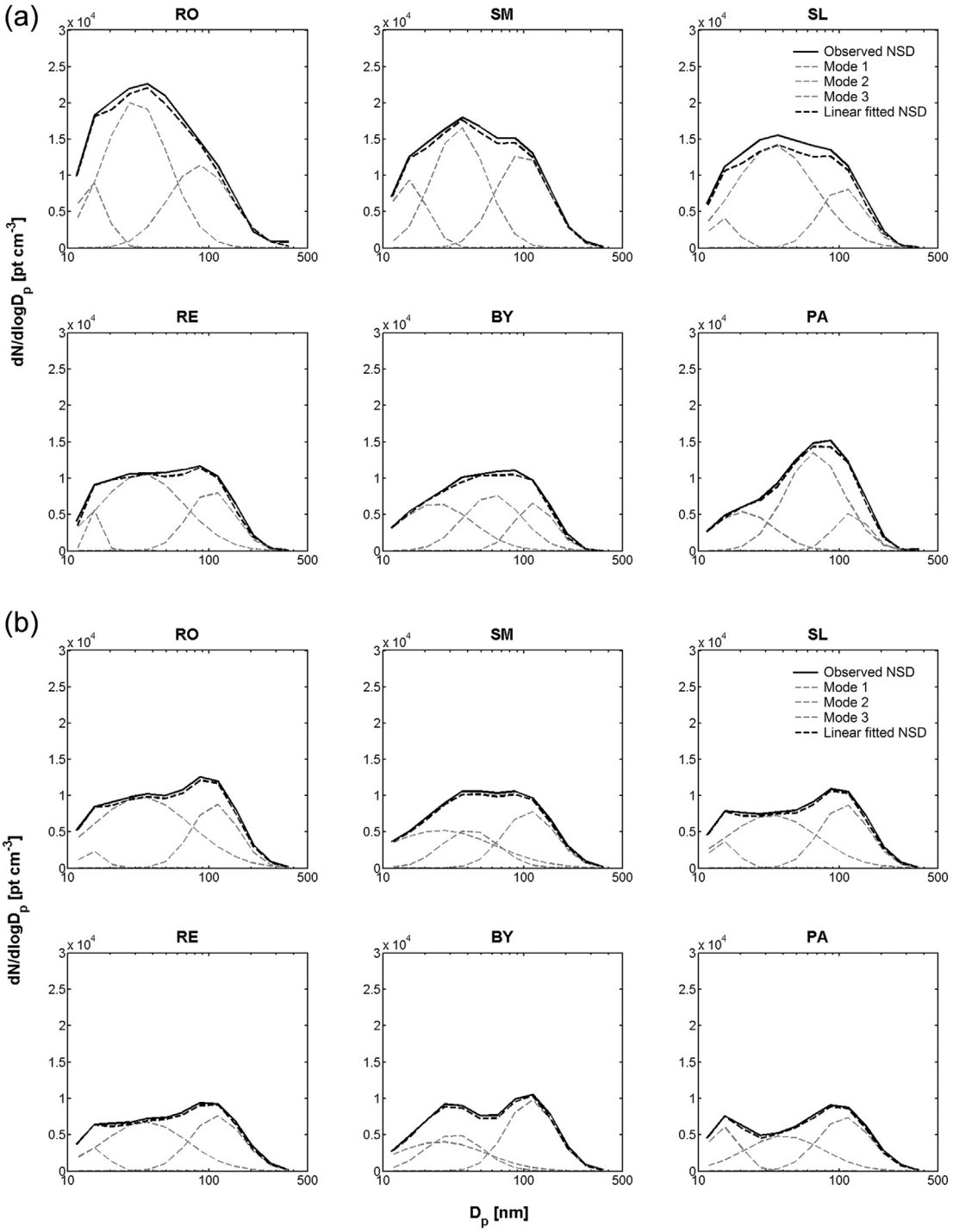

Fig. 8. Averaged size distributions and log normal modes for each microenvironment during winter (a) and summer (b) period. The measured NSD is shown in solid black lines, log normal mode 1–3 in dashed grey lines and linear fitted curve is shown in dashed black lines.

the measured number size distributions (Wegner et al., 2012; Hussein et al., 2014). Traffic sites are characterised by high total number concentrations up to $>5.0 \times 10^4$ pt cm^{-3} and $>7.5 \times 10^4$ pt cm^{-3} at RO, SM and SL whereas RE, BY and PA sites are characterised by lower number concentrations of around 30–50%. The ME that are prone to traffic (RO, SL, SM) are characterised by nucleation mode peaks at about 17–18 nm and Atiken mode peaks at 40 nm indicating the presence of fresh aerosol from traffic emission. During winter the condensation of hot tailpipe emissions in cold ambient air will be one of the main reasons for significant concentrations of nucleation particles <30 nm (cf. Olivares et al., 2007). According to observation by others emission from gasoline vehicles are located in the size range between 15 and 40 nm while diesel emission are found at around 15 nm with a second mode at about 60–80 nm (Harris and Maricq, 2001; Wehner et al., 2002; Kittelson et al., 2004; Morawska et al., 2008). In those ME which are not directly influenced by road traffic (BY, PA) the modal peak diameters shift to larger sizes in all modes indicating the dominance of aged aerosol. However, the picture in summer is less clear. The total number concentrations in all ME are considerably smaller

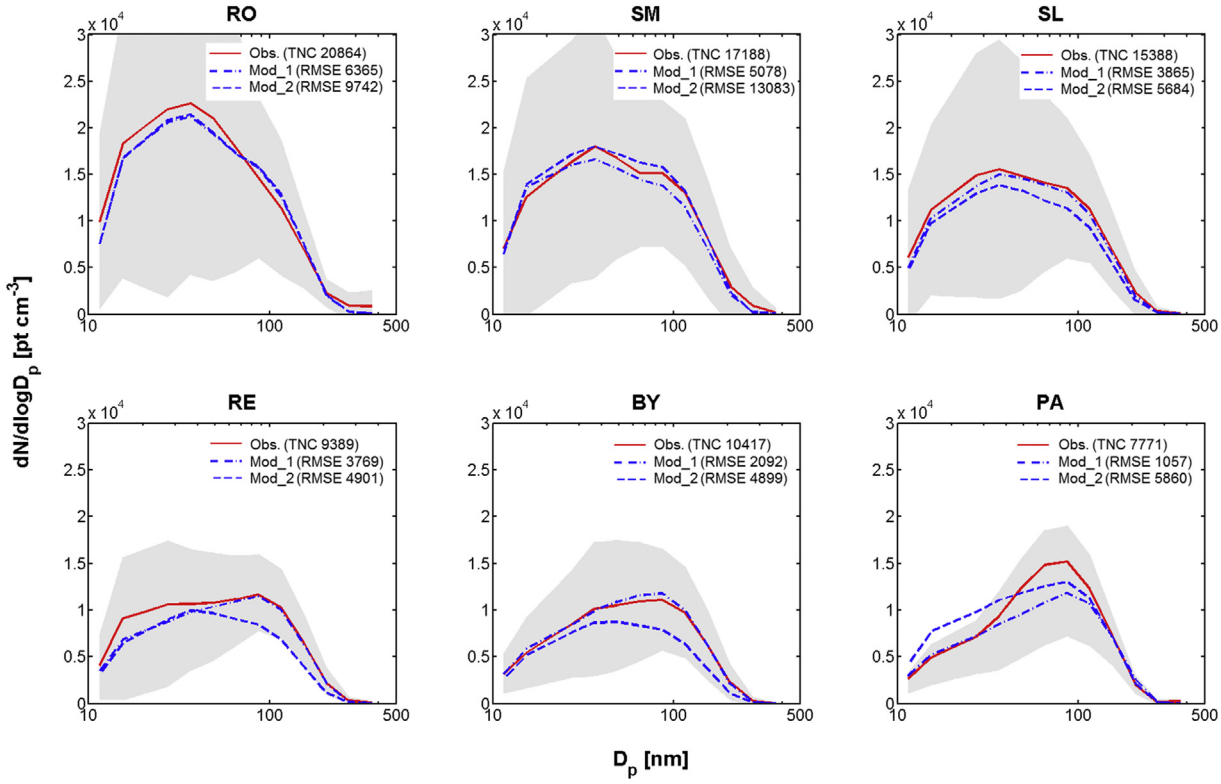

Fig. 9. Averaged particle number size distributions for each microenvironment during the winter period. The measured NSD is shown in solid red lines (Obs.), the modelled NSD is shown in dash-dot blue lines (Model 1) and dashed blue lines (Model 2). Grey area marks the interquartile range (25–75 percentile). Values in brackets indicate TNC [pt cm^{-3}] for a full NSD and the respective RMSE for Model 1 and Model 2. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

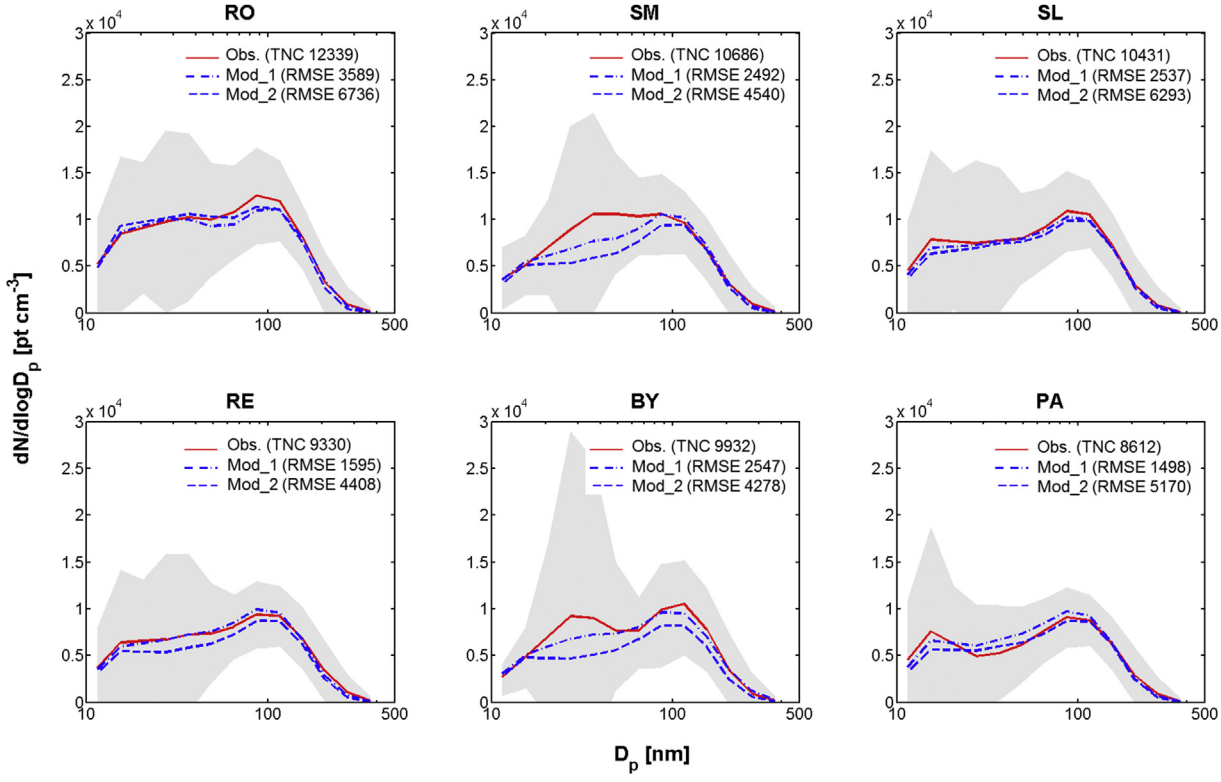

Fig. 10. Same as Fig. 9 but for summer period.

than during winter and, again, follow the rule of a decrease with increasing distance to traffic. However, a clear spatial relation of the modal peak diameter with distance to traffic is not observed in the log-normal fits.

3.3. Modelling number size distributions

Multiple linear regression analysis was used to model the particle NSD in the different ME (cf. Section 2.4).

Generally, both models are capable of calculating the particle number size distribution with good agreement to the observed NSD (Figs. 9 and 10). The average size integrated (TNC) deviation to observed data in all ME during winter is 7% for Model 1 and 12% for Model 2. Better agreement between model and observations is given for the near-traffic microenvironments RO, SM and SL, for which deviations are <6% for both models (except a 14% deviation for SL in Model 2). The model results for RE, BY and PA show somewhat limited agreement with the observed values, average deviations range between 5 and 23%.

Generally Model 1 shows better results than Model 2, since it incorporates a larger number of input data and offers some information of the size of the particles, i.e. mean size of the distribution as derived by the DiSCmini device (cf. Section 2.2). However, even without any information on the particle size included, Model 2 with only three input parameters works reasonably well, given the fact, that it is characterised by an average deviation from measured data in the ME of 12% in winter and 14% in summer, respectively. Generally, both models loose some detail in the size range >100 nm, whereas the ultrafine size range is satisfyingly covered in both approaches (Figs. 11 and 12). This is of relevance since the dominant fraction of the urban aerosol lies within the ultrafine size range and thus is important in exposure assessment. However, in the ME which are characterised by limited influence from traffic (RE, BY, PA) the model shows larger deviation in the accumulation mode >100 nm. Here, the pollutant information TNC and BC as well as the wind speed alone is not sufficient to explain variation in the particle size. Accumulation mode particle at BY and PA are underestimated by up to 70%. Model 1 works much better in the accumulation mode, however, the size parameter explains a significant part of the variance in the larger size channels, but is not available for Model 2 (Figs. 11 and 12).

The performance of the models during the summer season is similar to winter, with the exception that the deviations between both model approaches are smaller. Even in the more traffic remote ME the agreement between both models is comparable, i.e. the size information in Model 1 does not produce significantly better results. The relative influence of the size information in Model 1 during summer is strongly limited in the multiple regression approach (Fig. 13). On average the size integrated deviation of both models to observed NSD during summer is 6% (Model 1) and 14% (Model 2). The model approach presented in this study is not directly comparable to others reported in literature since most focus on longer time-series or were used to forecast number concentrations based on meteorological and traffic data, e.g. for the upcoming day (e.g. Clifford et al., 2011; Mølgaard et al., 2012). However, the present model should generally be applicable to model time series of NSD at traffic sites based on TNC, BC and wind speed data.

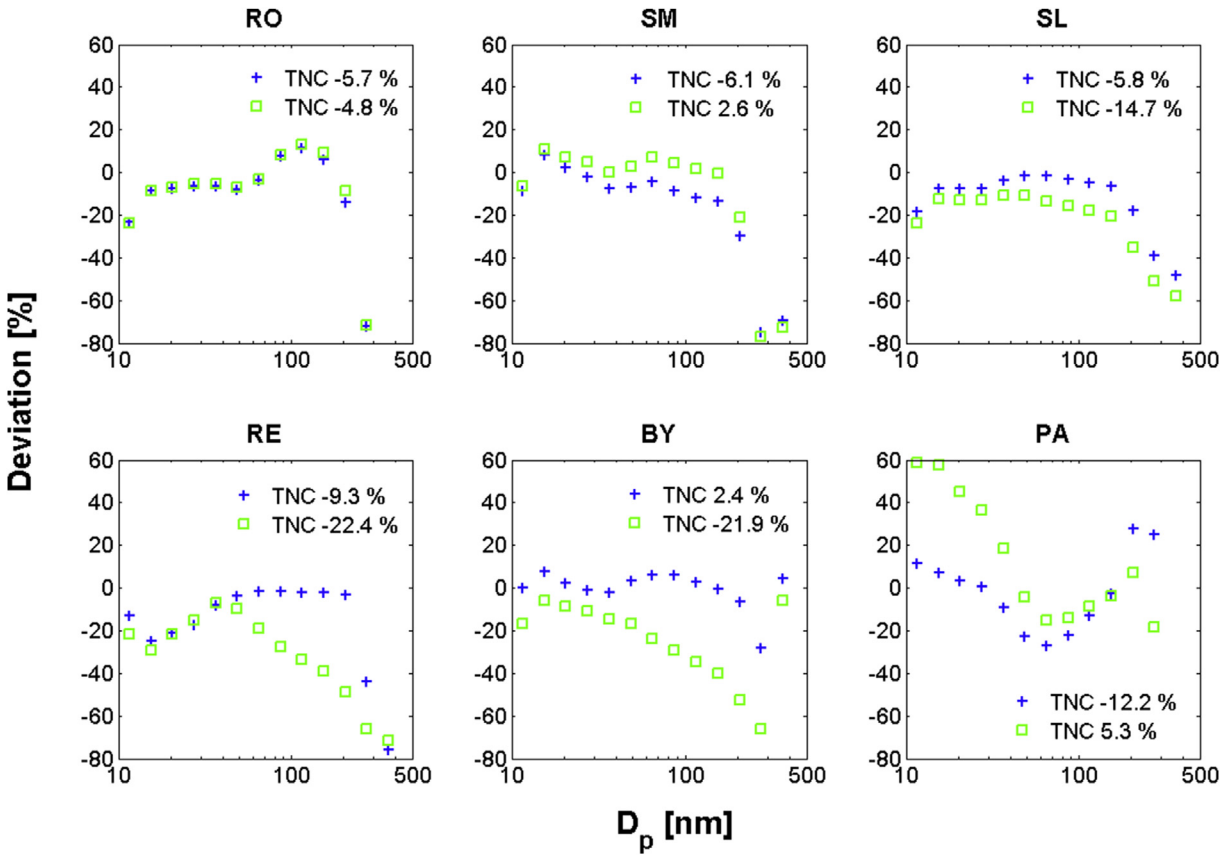

Fig. 11. Size dependent relative deviation between observed and modelled number size distributions for the winter period (Model 1: blue markers, Model 2: green squares). Bold numbers indicate average relative deviation between observed and modelled TNC. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

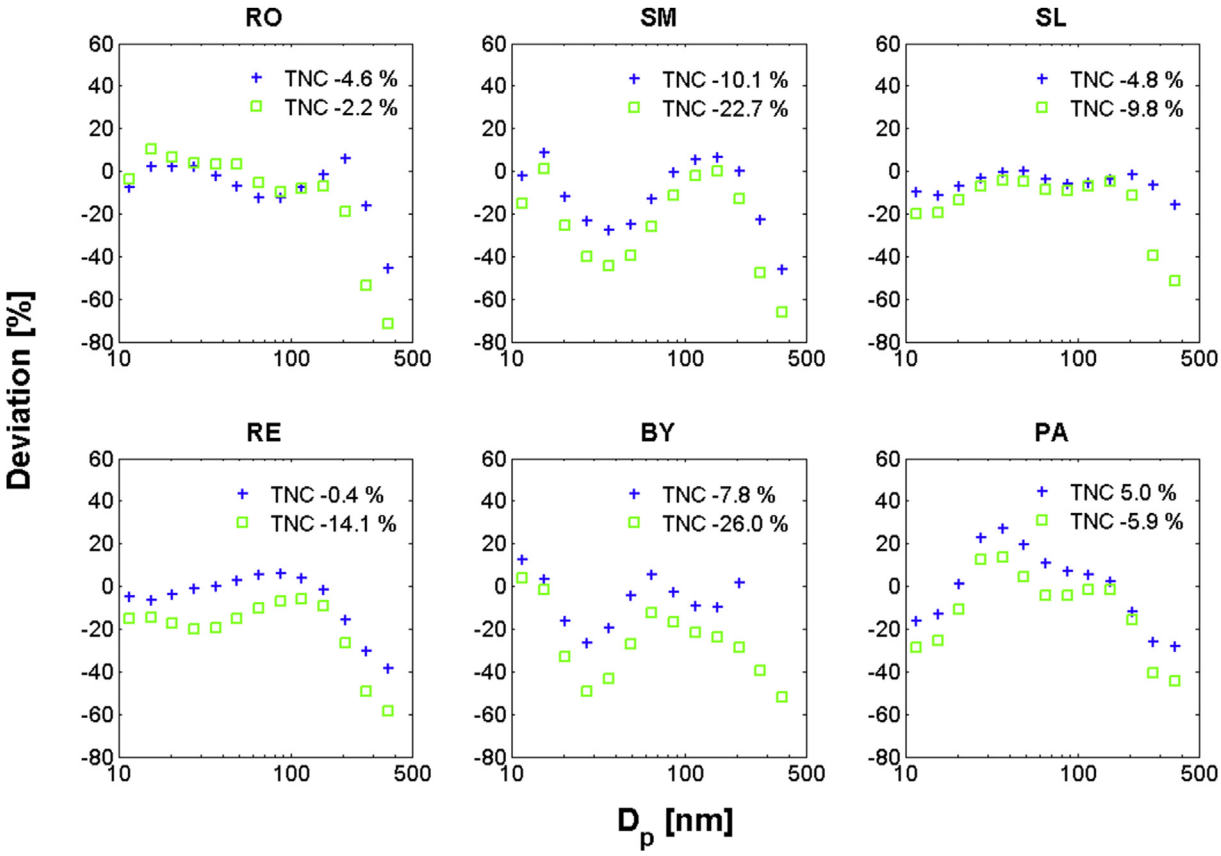

Fig. 12. Same as Fig. 11 but for summer period.

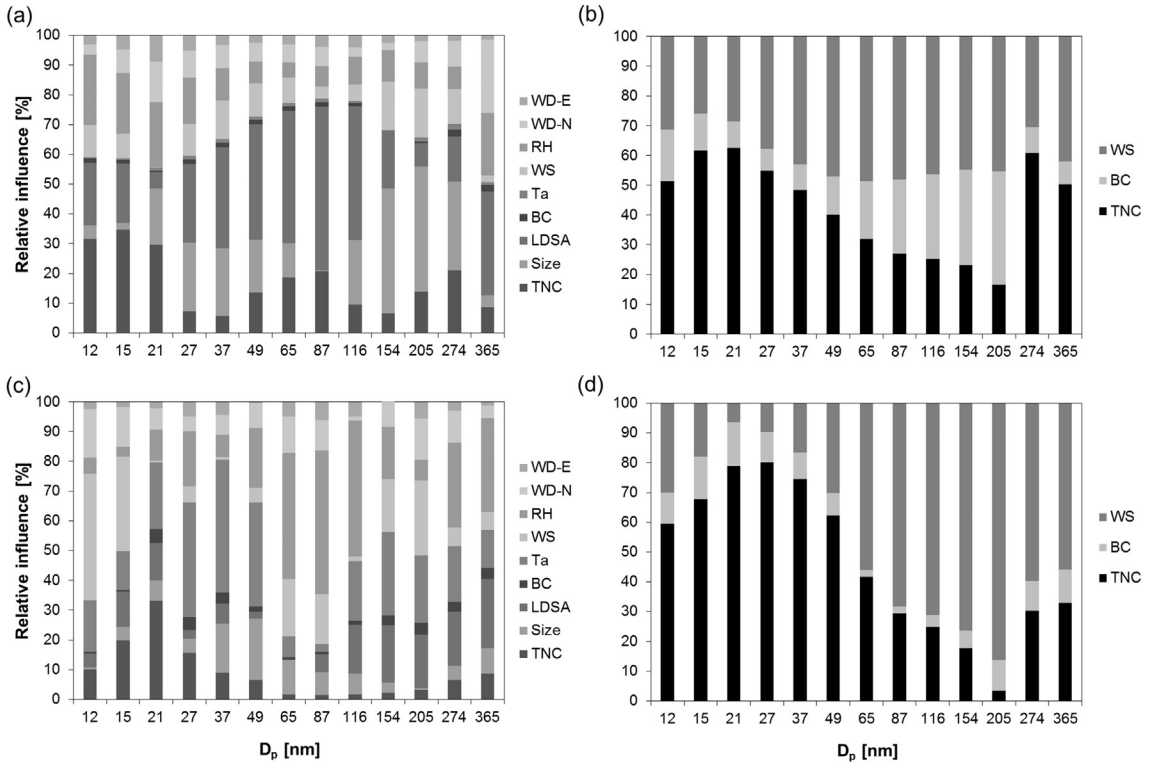

Fig. 13. Relative influence of parameters on the number size distribution during the winter period for Model 1 (a) and Model 2 (b) and during the summer campaign for Model 1 (c) and Model 2 (d). The different acronyms are described in the text, WD-E and WD-N refer to the East and North vector of the wind, respectively.

4. Conclusion

A mobile measurement campaign in an urban study area was conducted during the winter period 2012/13 and summer 2013 to analyse the microscale variation of particle and black carbon concentrations in six different outdoor microenvironments. The characteristics of the spatio-temporal variation were studied during comparable clear and calm weather situations.

The results indicate that higher concentrations of TNC and BC are obviously related to the distance of the measurement spot to traffic (on the busy main road) whereas sites which are more distant to traffic were characterised by lower concentrations. The difference is largest during winter. The higher correlation and spatially more homogeneous distribution of particle number and BC concentrations during winter is an effect of the combined influence of multiple sources (traffic, district heating, wood burning) and specific atmospheric conditions with more frequent situations of temperature inversions and stable atmospheric stratification in comparison to summer where traffic emissions are the dominating source. Therefore the measurement spots close to the main road are characterised by a higher spatial correlation between TNC and BC in summer.

The analysis of average concentrations in the six outdoor ME clearly reflects the results of the spatio-temporal distribution of air pollution concentrations at single measurement points. The microenvironments RO, SM and SL were characterised by higher number concentrations and modal peaks that point to fresh traffic emission. The sites RE, BY and PA which are more remote to traffic were characterised by lower concentrations and larger modal peak diameters.

Multiple regression analysis was used to investigate the relationship of the measured parameters on the NSD. The analysis indicates that only three parameters, which can be easily measured, can describe the NSD in each ME sufficiently. The model performance is limited for sites with low traffic activity, but works sufficient for high and medium traffic sites which is due to the close relationship of TNC and BC concerning traffic emissions. We believe that further implementation of urban structure/morphology parameters (e.g. building density and volume, roughness parameters) might significantly enhance model performance. This will be subject to future research.

The findings imply that a mobile approach is suitable to analyse the spatio-temporal variation of BC and TNC and to derive characteristic spatial patterns of both pollutants. The study of exposure towards particles using ME should take temporal variation throughout the year, as introduced by different atmospheric conditions on the seasonal cycle (e.g. accumulation vs. dilution), into account. The urban fabric (e.g. distance to roads and traffic, road type, building and street geometry etc.) clearly affects the concentration of particles and BC in urban microenvironments introducing considerable spatial variation that has to be considered in order to derive realistic patterns of individual exposure. It will be subject to future research to use the present model to estimate time series of NSD at traffic sites based on TNC, BC and wind speed data.

Acknowledgements

Hagen Mittendorf (Climatology and Environmental Meteorology, Technische Universität Braunschweig) is acknowledged for his technical assistance in manufacturing and testing the 'AerosolBox'. We additionally acknowledge Kay Weinhold (TROPOS) for his advice and assistance during the comparison measurements. MR acknowledges partly funding of his research in the framework of the 'Zukunftsfonds'-initiative of the Technische Universität Braunschweig.

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