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Variability of particle number concentration and particle size dynamics in an urban street canyon under different meteorological conditions

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HIGHLIGHTS

- ► Effects of turbulent exchange and different street canyon flow situations on aerosol concentrations were studied.
- ► Significant elevation of concentrations < 70 nm within the street canyon throughout daytime hours
- ► Effects of meteorology and traffic on aerosol were parameterised using multiple linear regression.
- ► In most situations, the model performs with a relative uncertainty of < 25%.

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ABSTRACT

During a six-month study period, aerosol number size distributions, mean meteorological conditions and turbulent exchange were measured within an urban street canyon in Essen, Germany. The findings were compared to simultaneous measurements conducted at suburban sites within the study area. The effects of turbulent exchange and different canyon flow situations on aerosol number concentration variability within the street canyon were studied.

In comparison to a suburban background site, the busy urban street canyon aerosol number concentration was significantly elevated in the size range below 70 nm throughout the daytime hours. During the morning rush hour, total number concentrations were a factor of 2.2 higher. On average, the total number concentration at the street canyon site roughly doubled the suburban background concentrations (by a factor of 1.9). The intensity of turbulent mixing within the street canyon was sensitive to the prevailing flow regime. The highest turbulent mixing during cross-canyon flow from directions downwind of the measurement spot was accompanied by the lowest number concentration of all flow regimes observed within the canyon. This behaviour was consistent for the different aerosol size classes considered in this study.

The effects of meteorology and traffic intensity on total aerosol number concentrations were parameterised using a multiple linear regression analysis and indicated that turbulent mixing within the canyon, traffic intensity and NO_x concentrations were the most significant parameters. The model is characterised by an average relative uncertainty of 29%. During situations with a total number concentration > 7500 cm^{-3}, a relative uncertainty of the modelled data of ±25% emerges but displays a larger deviation for low particle concentrations.

1. Introduction

The significance of fine and ultrafine aerosol concentrations in the context of impacts on human health has been well established in numerous studies during recent years (Brunekreef and Holgate, 2002; Lipfert and Wyzga, 2008; Russell and Brunekreef, 2009; Kumar et al., 2010; Knibbs et al., 2011). There is evidence that submicron particles, especially ultrafine particles < 100 nm in diameter (UFP), have a stronger association with health effects than larger particles (e.g., $PM_{2.5}$ and PM_{10}), which are currently regulated by legal limit values (Wichmann and Peters, 2000; Oberdörster and Utell, 2002). Urban areas are of major interest in this context due to the significant emission of submicron and ultrafine particles from stationary and mobile sources. Important stationary sources of ultrafine particles include commercial and industrial combustion processes and district heating during the winter (e.g., Wegner et al., 2012). The emission of aerosol from public and private traffic is the most significant source of mobile particle sources in urban areas. The predominant fraction of traffic aerosol is typically emitted in the ultrafine

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region of the aerosol size spectrum, e.g., up to 90% of the total number concentration (TNC), as documented in a review by Morawska et al. (2008).

Hence, the spatio-temporal distribution of aerosol number concentrations within urban areas is highly variable. This is predominantly a result of the different locations, source types and rates of emission of particles and the complex three-dimensional structure and flow regime within cities (Weber and Weber, 2008; Kuttler, 2012).

Urban and suburban aerosol number size distributions can generally be described by two or three log-normal modes, in the nucleation (<30 nm), Aitken (30–100 nm) and accumulation (>100 nm) size range being indicative of specific emission sources and transformation processes in the atmosphere (e.g., Hussein et al., 2004; Seinfeld and Pandis, 2006; Wegner et al., 2012). While aerosol number concentrations in the accumulation size range are somewhat comparable between urban sites, the heterogeneity in particle size distribution and composition of smaller particles increases the more the measurement site is prone to emission from vehicular traffic (van Dingenen et al., 2004; Krudysz et al., 2009; Moore et al., 2009; Putaud et al., 2010; Wang et al., 2012). In urban street canyons, the significant emission of ultrafine particles and gaseous precursors from traffic coincides with a limited dilution by increased aerodynamic roughness, making them the 'hot-spots' of urban particulate pollution. This phenomenon applies to both particle number and mass concentrations (e.g., Weber et al., 2006a; Morawska et al., 2008; Weber, 2009; Kumar et al., 2011).

Urban scale variation in aerosol number concentrations has recently been intensively studied; however, less is known about the relationships between meteorological and boundary layer processes in urban areas and the resulting particle number concentrations at different sites. In particular, the influence of turbulent flow on particle number concentrations and particle size dynamics (e.g., in urban street canyons) is not properly understood. A number of research studies on the general characteristics of pollutant dispersion in urban street canyons have been performed in recent years (e.g., Vardoulakis et al., 2003). However, field studies on the relationship of local meteorology and micrometeorological effects on number concentrations, i.e., ultrafine particles, are currently limited. In addition to the processes of new particle formation (e.g., from the condensation of hot tailpipe emissions during the winter months, e.g., Olivares et al., 2007) and the physical/chemical transformation of aerosols (e.g., coagulation, condensation) on the resultant airborne concentrations measured at some receptor site within a canyon, the effect of micrometeorological processes, i.e., dilution, deposition and microscale flow patterns, is most prominent. The influence of wind direction on the evolving patterns of microcirculation within canyons (e.g., vortex circulation) has been studied by Weber et al. (2006a), Nikolova et al. (2011) and Mishra et al. (2012). These authors have all cited the significant influence of wind direction and mean wind speed on near surface concentrations. The effect of turbulent flow (mechanical and thermal turbulence) on street canyon aerosol number concentration was investigated to understand its diluting capabilities and dispersion characteristics, i.e., the ventilation capability within street canyons with fresher air from aloft (Longley et al., 2004; Salmond et al., 2010). Tay et al. (2010) investigated the influence of different flow conditions on street canyon ultrafine aerosol fluxes, pointing to a stronger relationship between horizontal wind speed and turbulence intensity rather than between aerosol and sensible heat fluxes. On the contrary, Longley et al. (2004) reported a positive relationship between accumulation mode particles ($0.1<D_p<3$ μm) and sensible heat flux.

The relationships between different meteorological and/or anthropogenic quantities (e.g., traffic fleet and traffic volume) have been successfully used in the past to parameterise urban aerosol mass and number concentrations with different statistical approaches, e.g., multiple linear regression and general additive modelling (Aldrin and Haff, 2005; Voigtländer et al., 2006; Mølgaard et al., 2012). Traffic intensity and wind speed appear to be the variables that most significantly correlate with total number concentrations in the size range $3<D_p<800$ nm (e.g., Voigtländer et al., 2006). Mølgaard et al. (2012) were able to forecast UFP concentrations for the upcoming day with a coefficient of determination of $r^2=0.67$. Generally, statistical model approaches are straightforward applications to parameterise the aerosol number concentration using operationally measured parameters such as meteorological variables or gaseous pollutants. These are important tools for the evaluation of exposure towards urban aerosol.

The aim of the study VAPARTICO (Variability of particle concentrations and particle size dynamics in an urban street canyon under different ambient meteorology) was to study the effects of different local meteorological processes (mean meteorology and turbulence) on the resulting aerosol number concentrations and size dynamics within a busy urban street canyon and at a suburban background site. The focus was on studying the effects of turbulent flow, e.g., momentum and sensible heat fluxes, and different flow situations, e.g., along and the cross canyon flow in the canyon, on aerosol concentrations.

2. Materials and methods

2.1. Study area and instrumentation

The VAPARTICO measurements were performed during a six-month study period from 15 April 2008 to 15 October 2008 at four sites in the city of Essen, Germany (Fig. 1). The four measurement sites were comprised of:

- a busy street canyon in the northern area of Essen (CAN)
- a rooftop site adjacent to the street canyon (ROF)
- a suburban background site at the western border of Essen (SUR) and
- a regional airport site at the Essen–Mülheim airport, situated in southwestern Essen (SOD).

The sites CAN and ROF were fully equipped with instruments to measure aerosol concentrations and meteorological quantities during the full study period (cf. Table 1 for a full list of instrumentation). An intensive observation period (IOP) was held from 16 June to 02 September 2008, during which the sites SUR and SOD were in operation so that all four sites were being measured simultaneously. However, the second scanning mobility particle sizer (SMPS) at SUR did operate until 29 September 2008. Therefore, the aerosol size distributions at CAN and SUR are available for a period of 3.5 months (16 June to 29 September 2008). Below are the details for the study sites and instrumentation.

2.1.1. CAN — street canyon

The street canyon at CAN is orientated SE–NW (135°–310°). Mean building height is H = 17 m while the canyon is W = 21.6 m wide, resulting in a height to width ratio (H/W) = 0.79 (Fig. 2). The residential houses are predominantly comprised of four floors. The street canyon "Gladbecker Straße" (federal road, B224) is characterised by four traffic lanes, two directed towards the centre of Essen while the other two lead towards the northern parts of the Ruhr-Area.

A measurement container (1.8 m length × 1.0 m width × 2.25 m height) and a 10 m triangular lattice tower were installed at the northeastern kerbside at a distance of approximately 3 m off the northern house wall (cf. Weber et al., 2006a). The container housed a SMPS Mod. TSI 3936 which consisted of a TSI 3080 Electrostatic Classifier with differential mobility analyser (DMA) and a water-based condensational particle counter (CPC) TSI 3785. Ambient aerosol was sampled at a height of 3.1 m agl through a 1.6 m stainless steel tube with an inner diameter of 4 mm. With an aerosol flow rate of 0.3 L min^{-1}, the SMPS performed aerosol size distribution scans in the electrical mobility diameter range $20<D_p<740$ nm

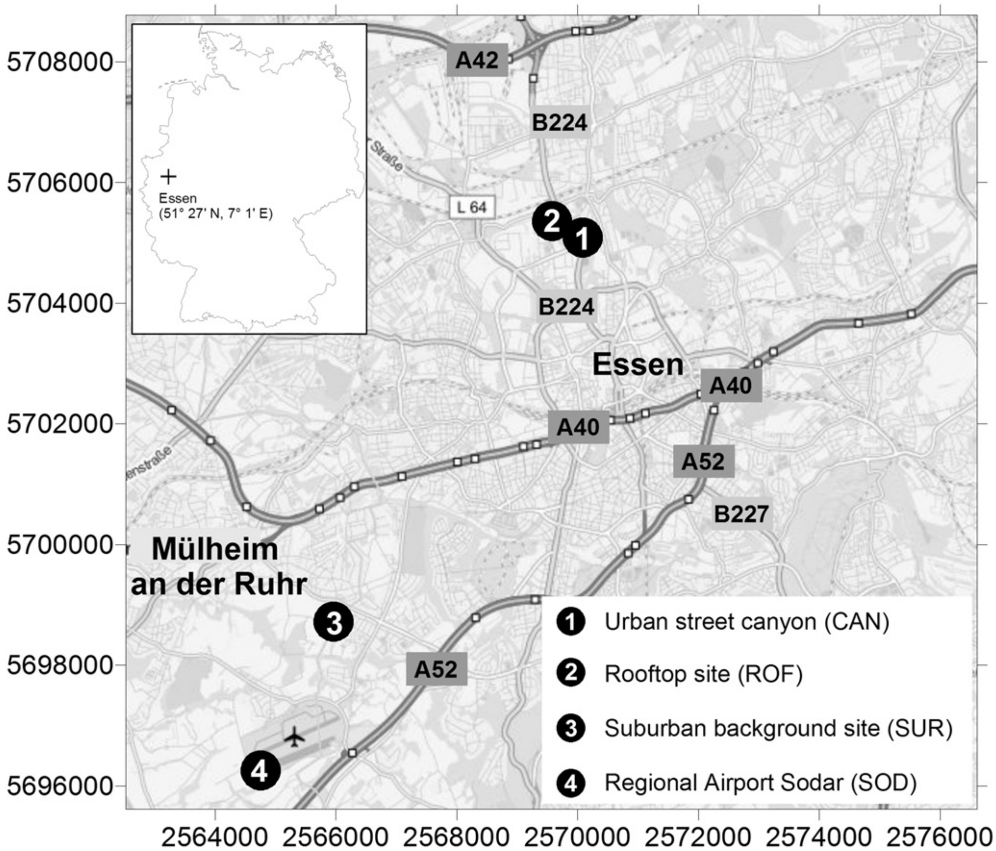


Fig 1. Map of the study area indicating the four urban/suburban measurement sites. The labels refer to significant road transport infrastructure (federal motorways A40, A42 and A52, federal road B224). The map inset marks the location of Essen, Germany.
Map source: www.openstreetmap.org, modified.

every 5 min. Aerosol size distributions in the range $0.3 < D_p < 10$ µm were estimated at a height of 3.4 m agl using an optical particle counter (OPC, Grimm, Mod. 1.107).

The 10 m tower was equipped with three levels of sonic anemometers (USA-1, Metek, Germany) at non-dimensional heights of $z/H =$ 0.22, 0.37, 0.59 (normalised by the average building height H) to measure the horizontal and vertical wind vectors u, v, w and the acoustic temperature T_s at a sampling frequency of 10 Hz. Air temperature and relative humidity (Pt100 and hair hygrometer, Th. Friedrich, Germany) were measured at 2.60 m agl. The full radiation balance was measured using an albedometer (CM7B, Kipp + Zonen) and a pyradiometer (Ph. Schenk, Austria) at 3 m agl.

At CAN, traffic intensity was monitored by the North-Rhine Westphalia State Agency for Nature, Environment and Consumer

Table 1
Overview of instruments that were used during VAPARTICO to measure the near surface aerosol dynamics and meteorological conditions at the four measurement sites.

Site	Quantity	Label	Model	Measurement height (agl)	Sampling interval	Study period
CAN						
	Aerosol size distribution ($0.02 < D_p < 0.74$ µm)	TNC, SD	SMPS TSI 3936	3.80 m	5 min	E
	Particle number and volume distribution ($0.3 < D_p < 10$ µm)	TOV	OPC Grimm 1.107	3.40 m	1 min	E
	Turbulence	u, v, w, T_s	Sonic Metek USA-1	3.75 m	10 Hz	E
		u, v, w, T_s	Sonic Metek USA-1	6.50 m	10 Hz	E
		u, v, w, T_s	Sonic Metek USA-1	10.00 m	10 Hz	E
	Air temperature	T_a	Th. Friedrichs Pt100	2.60 m	10 s	E
	Relative humidity	rH	Th. Friedrichs, hygrometer	2.60 m	10 s	E
	Short- and long-wave radiation	K↓, K↑, L↓, L↑	Ph. Schenk Net radiometer, Kipp + Zonen CM7B albedometer	3.00 m	10 s	E
	Nitrogen oxide	NO_x	LANUV NRW	3.00 m	30 min	IOP
ROF						
	Particle number and volume distribution ($0.3 < D_p < 10$ µm)	TOV	OPC Grimm 1.107	27.50 m	1 min	E
SUR						
	Aerosol size distribution ($0.02 < D_p < 0.74$ µm)	TNC, SD	SMPS TSI 3936	2.30 m	5 min	IOP
	Particle number and volume distribution ($0.3 < D_p < 10$ µm)	TOV	OPC Grimm 1.107	2.30 m	5 min	IOP
	Turbulence	u, v, w, Ts	Sonic Metek USA-1	4.50 m	10 Hz	IOP
SOD						
	Boundary layer measurements	u, v, w	SODAR Scintec MFAS	0.50 m	30 min	IOP

TNC = total number concentration ($20 < D_p < 745$ nm); SD = size distribution; TOV = total volume concentration; u, v, w = horizontal and vertical wind vectors; T_s = sonic temperature; T_a = air temperature; rH = relative humidity; K↓, K↑, L↓, L↑ = upward and downward shortwave and longwave solar radiation.
Study period: E = entire period, IOP = intensive observation period.

Fig. 2. The measurement sites street canyon (CAN) and rooftop (ROF) in Essen, Germany. Left picture: view to the NNW, the container housing the measurement equipment was installed at the northeastern kerbside. Right picture: downward view from the measurement site ROF to the SE into the street canyon.
Photos: Kordowski, Weber.

Protection (LANUV NRW) starting on 22 May 2008. By using conduction loops, 30 min of traffic data, sorted into different vehicle categories, were monitored. From this data set, three different traffic categories were classified for further data analysis: PCAR (passenger cars), HDV (heavy duty vehicles, lorries, articulated lorries and buses) and VEH (containing all vehicle categories). The classification into three categories was performed to study the influence of the traffic fleet mixture on aerosol concentrations in more detail. Passenger cars (PCAR) are the predominant fraction of the traffic fleet, either gasoline or diesel vehicles, while a heavy-duty vehicle (HDV) class was established because predominantly diesel vehicles are believed to fall into this subcategory.

2.1.2. ROF — rooftop site

At a distance of 300 m to the NE of CAN, particle size distributions were quantified using an optical particle counter on a rooftop at a height of 27.5 m agl. The OPC operated at a sampling frequency of 6 s. One min averages were aggregated and stored to data storage cards. These measurements were performed to derive a near canyon background concentration that is not directly influenced by traffic. The residential building itself has a height of 25 m and is one of the largest buildings in this area (cf. Weber and Kordowski, 2010). The mean building height in the surrounding of the ROF was calculated from a digital elevation model with $H = 17$ m agl.

2.1.3. SUR — suburban background site

At the western border of the city of Essen, meteorological and aerosol measurements were performed at the suburban station Harscheidweg (Weber and Kordowski, 2010). This station is situated at a height of 135 m above sea level and is predominantly surrounded by land used for agriculture. To the SW and S, the terrain slopes to approximately 100 m asl. At a distance of approximately 150 m to the NE of the site, a residential area with two storey houses is located. The inner urban area of the city of Essen is situated at a distance of approximately 6 km to the NE of the site. To the north of SUR, a two lane road crossing the area from NW to ESE with an average daily traffic intensity of approximately 21,000 veh d^{-1} is situated.

Both an SMPS and OPC were installed during the IOP from 16 June to 29 September 2008. The TSI 3936 SMPS consisted of an Electrostatic Classifier 3080 and a butanol-based CPC 3010. Aerosol was sampled at a height of 2.3 m agl through a 1.5 m sampling line with an inner diameter of 4 mm. Flow rates, scan times and diameter size ranges are according to the SMPS settings at CAN. A Grimm 1.107 OPC was installed to sample particles at a height of 2.3 m agl. Turbulence was measured using a sonic anemometer (USA-1, Metek, Germany) at a height of 4.5 m agl.

2.1.4. SOD — SODAR regional airport

At the regional airport "Essen/Mülheim" in the SW of the city of Essen, boundary layer measurements were performed by SODAR. The airport is situated on the city border of Essen and Mülheim, comprises an area of approximately 141 ha and is situated at 124 m asl. The densely built up areas of Essen are situated at a distance of 5.5 km to the NE of the site.

The SODAR was installed at a height of 0.5 m agl during the IOP. The monostatic SODAR (Scintec AG, Germany, Mod. MFAS) measures horizontal and vertical wind vectors and turbulence characteristics using sound pulses that are emitted by the instrument and backscattered by atmospheric temperature in homogeneities (e.g., Emeis, 2010). The backscattered signal is received and analysed by the instrument's software. Ten metre layer averages were derived and stored at a time resolution of 30 min. Because of the noise exposure to residents, the SODAR had to be operated with limited sound power. Therefore, data availability at measurement heights $z > 250$ m agl was constricted. In this study, only data from height levels $30 < z < 200$ m agl will be presented.

For subsequent data analysis, all gathered data at the different sites were aggregated to 30 min averages.

2.2. Data handling and quality control of the data

2.2.1. Classification of street canyon wind directions

To analyse the different effects of microscale circulation regimes on the aerosol concentrations within the street canyon (e.g., vortex circulations), the data were categorised into different flow regimes, owing to the direction of the reference wind. Reference wind speed and direction were defined by the measurements taken at 10 m agl at SUR.

Owing to the orientation of the street canyon axis, flow directions were classified into situations of along canyon (ALC) or cross canyon flow (CRC, cf. Fig. 3). Flow along the canyon axis (ALC) was defined for a reference wind direction at an angle of 20° around the street canyon axis, i.e., $125° < \phi_{ref} < 145°$ and $305° < \phi_{ref} < 325°$. Cross canyon flow is defined for flow angles perpendicular to the street canyon axis, i.e., CRC_{SW} $145° < \phi_{ref} < 305°$ for flow from the SW and CRC_{NE} $325° < \phi_{ref} < 125°$ for flow from the NE. Therefore, the CRC flow classification does not only account for wind directions strictly perpendicular to the canyon axis but also to those being directed at some angle to the canyon axis. Owing to this classification scheme, 5% of the data can be characterised as ALC flow while 38% are cross canyon flow from the NE (CRC_{NE}), and the majority of situations are CRC_{SW} with a frequency of 57%.

2.2.2. Comparison of SMPS and OPC

Prior to the campaign, the different particle measurement systems were operated simultaneously in the laboratory and the field. The SMPS systems demonstrated a good correlation of total number concentration measurements; however, the butanol-based CPC (SUR) overestimated the CAN total number measurements by approximately 30% ($r^2 = 0.98$). This magnitude of deviation is in agreement with other comparison studies (e.g., Birmili et al., 2007). However, the number concentration data in the size channels of both SMPS was corrected to the CAN SMPS. The OPC displayed an average deviation of approximately 15% in the number concentration measurements between the devices. The different systems were corrected to each other. However, whenever we refer to particle number concentrations throughout the paper, these were measured using the SMPS systems at CAN and SUR in the size range $20 < D_p < 745$ nm. The OPC data in the size range $0.3 < D_p < 10$ μm were only used to calculate total particle volume concentrations (TOV) for the different sites.

During the course of the measurement campaign, some hitherto unknown failures occurred with the SMPS system operated at CAN. The SMPS was actually operated with a lower cut-off at $D_p =$ 14.1 nm. However, the small size channels of the SMPS in the range from $14.1 < D_p < 24.1$ nm were characterised by frequent "zero concentration counts", which started approximately 9 weeks after the first measurements. Fig. 4 indicates that the smallest size channel observed approximately 50% zero counts during the study period. The first eight size channels were characterised by a high percentage of zero counts, which decreased to approximately 2% for the size range $18.8 < D_p < 24.1$ nm and did not occur for larger particle sizes. The reason for the zero counts in the small size channels is still unsolved and was also unknown to the manufacturer of the device. However, a similar failure did occur with other SMPS systems produced by this manufacturer (Kaminski, pers. communication). We found that when changing to a 10 min size distribution resolution with a 5 min "off-time" between scans (from approximately 30 s in the 5 min resolution mode), the problem did not occur. Currently this problem has still not been satisfactorily resolved. Birmili et al. (2007) reported a significant underestimation of particle counts in the smallest size channels, which was caused by a poorly grounded DMA. This has its strongest effects on measurements in the smallest particle sizes because the lowest voltages are required there. Poor grounding of the DMA is also believed to be responsible for the effects detected during the measurements in VAPARTICO; however, no clear failure was detected during maintenance by the manufacturer. A

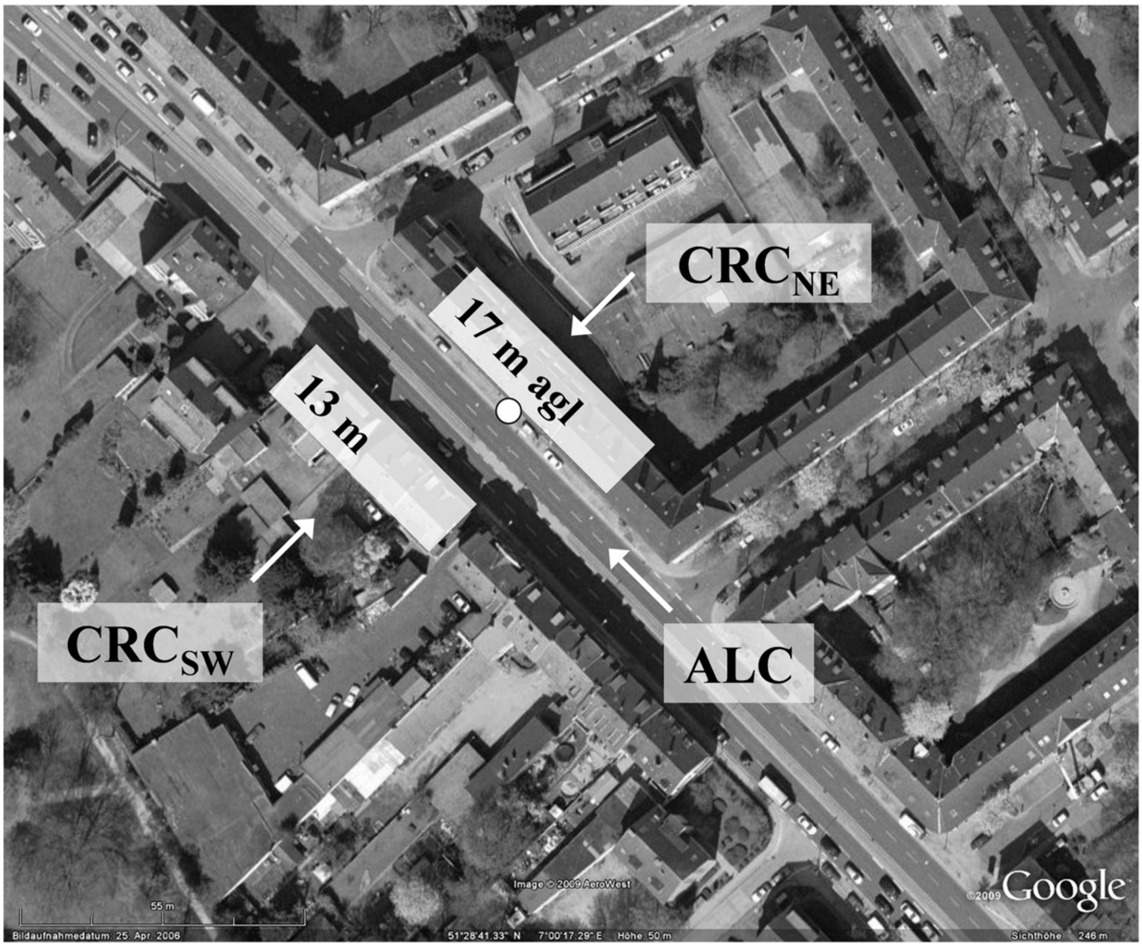


Fig. 3. Site details and canyon geometry. The insets indicate the mean building heights at both sides of the canyon and the classification of the flow regimes (see text). Map source: Google Earth.

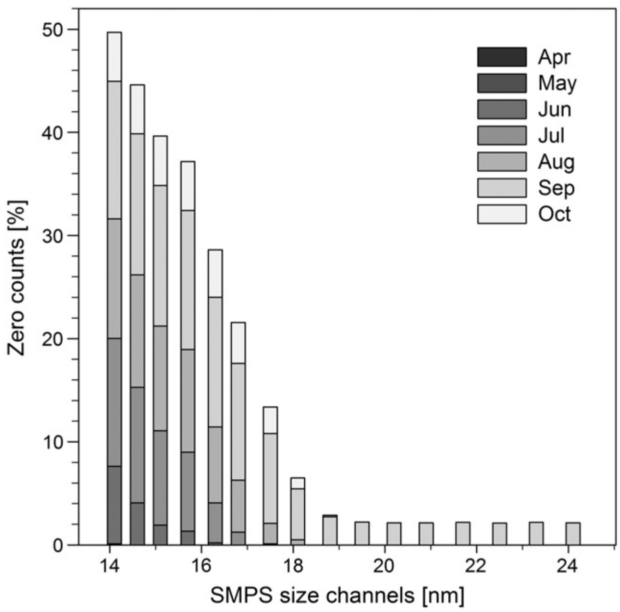


Fig. 4. Monthly frequency of 'zero concentration counts' in the lower size channels of the SMPS at CAN during the study period.

comparison of the SMPS to a fast electrical mobility sizer (FMPS, TSI Mod. 3091) demonstrated a good correlation and agreement of particle number concentrations and size distributions (Kaminski et al., 2010). A 17% overestimation of the FMPS in the ultrafine particle size range $14<D_p<100$ nm was found during comparison measurements at CAN.

A high frequency of zero counts significantly influences the (average) aerosol number size distribution (cf. Fig. 5 left). With an increasing number of zero counts in the lower size channels, the average size distributions significantly decreases at diameters below 30 nm. To not lose the information in these size channels, a correction scheme based on three steps was applied:

- First, all size channels below 20 nm had to be deleted and were not used in subsequent analyses because the frequency of zero counts was too large to apply plausible correction schemes.
- Second, monthly normalised average size distributions were calculated (Fig. 5 left). The size distributions were normalised by the monthly average total number concentration. It is evident that in June the size distributions start to significantly drop for aerosol <30 nm in comparison to April and May. Therefore, a monthly correction factor was calculated to correct for the size distributions from June to October based on the undisturbed monthly size distribution from April and May ('base case'). The correction factor (Corr.Factor, Fig. 5, right) was calculated according to:

$$\text{Corr.Factor} = 1 + \left(SD_n / \langle SD_{\text{April,May}} \rangle\right) \quad (1)$$

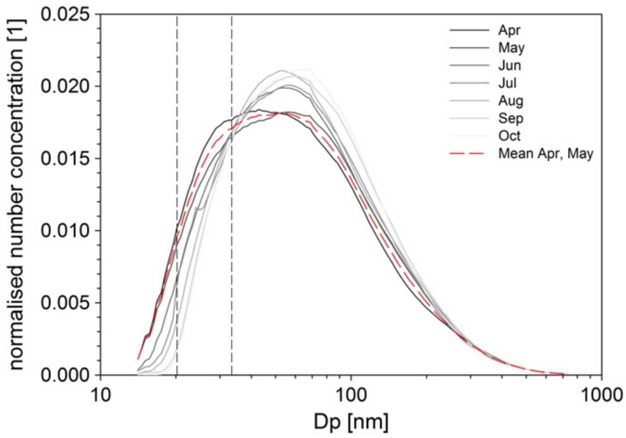
with SD_n being the normalised size distribution of the respective month and $\langle SD_{\text{April,May}} \rangle$ the mean normalised size distribution of the 'base case' from April and May.
- Third, the Corr.Factor was applied to the SMPS size channels $20.2<D_p<33.4$ nm to calculate the corrected size distributions for the months June to October. The correction was not applied to size channels >33.4 nm because 'zero counts' were not detected for these particle sizes. However, the number concentrations of particle sizes below 30 nm might still be underestimated and have to be treated with some caution.

3. Results and discussion

3.1. Characteristics of traffic fleet and traffic intensity within the street canyon

The traffic intensity, defined as the number of vehicles per unit time, at CAN is characterised by an average daily traffic intensity of 41,000 veh d^{-1} (±8500 veh d^{-1}). The traffic volume is relatively constant during the week, with approximately 45,000 veh d^{-1} (weekdays from Monday to Friday), but drops to lower values of 38,000 veh d^{-1} and 23,000 veh d^{-1} on Saturdays and Sundays, respectively (Fig. 6). On average, 88% of the traffic fleet is comprised of PCAR while 6% are HDV (the remaining 6% are either unclassified or motor bikes).

The average diurnal course of traffic intensity for all vehicles in CAN is characterised by a bimodal shape (Fig. 7), which shows a first sharp peak during the morning rush hour between 06:30 and 09:00 CET. Approximately 20% of the daily traffic volume is passing the measuring station at CAN during this time period. The morning rush hour peak is followed by a decline of traffic intensity and a subsequent build up with a broad 'plateau-like' afternoon rush hour peak between 14:30 and 17:30 CET. During this time period, approximately 25% of the daily traffic volume is counted. While approximately 45% of the daily traffic volume occurs during rush hour,



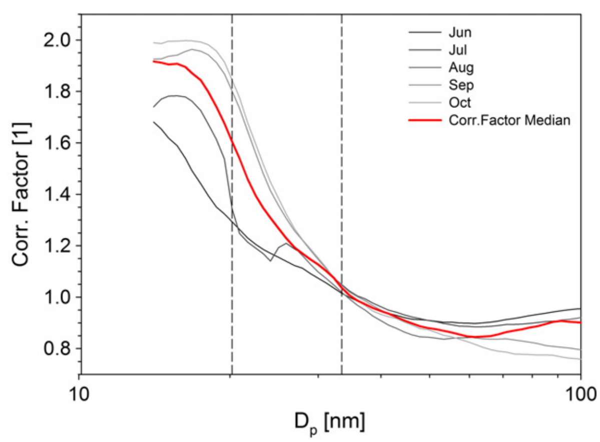


Fig. 5. Comparison measurements of (left) the uncorrected aerosol number size distributions of both instruments in the size range $14<D_p<736$ nm, based on 5 min averages (n = 553), and (right) monthly correction factors in the size range $14<D_p<100$ nm. The vertical dashed lines indicate the size range $20<D_p<34$ nm, in which the correction scheme was applied.

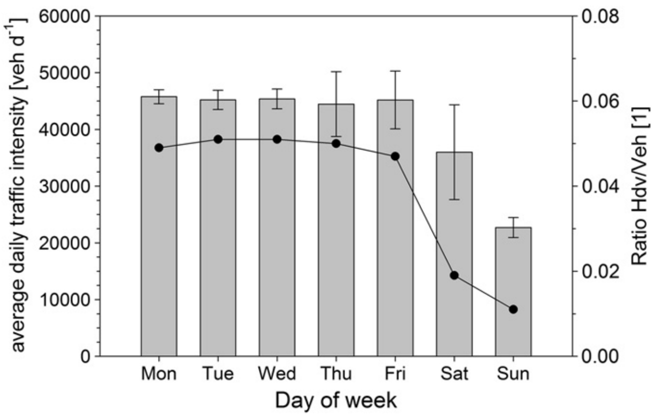


Fig. 6. Average daily traffic intensity at CAN, measured during the time period from 22 May to 11 October 2009 (left). Standard deviations are indicated by error bars. The ratio of HDV/VEH is marked by the black line (data basis: 30 min sums).

approximately 85% of the daytime traffic volume is registered during 06:30 and 20:00 CET. The shape of the diurnal course of HDV is different from VEH. No clear maximum (e.g., during the morning or afternoon rush hour) exists, but a broad plateau during office hours occurs from about 07:00 to 16:00 CET, with approximately 68% of daily HDV passing CAN during this time period.

The average diurnal courses for different days of the week are characterised by very similar shapes for the weekdays, Monday to Friday (Fig. 7). The temporal evolution and the absolute number of vehicles is about approximately the same on weekdays. The diurnal courses of VEH on Saturdays and Sundays have a unimodal shape, with a maximum at approximately 14:00 CET. Although the absolute number of vehicles is lower on Saturdays in comparison to the weekdays (see above), the noon maximum on Saturdays is higher than the corresponding estimates during weekdays (1530 veh 30 min^{-1} and 1390 veh 30 min^{-1}, respectively).

3.2. Characterisation of canyon meteorology and turbulent exchange inside the canyon

The undisturbed frequency distribution of wind direction during the measurement period at SUR is characterised by a maximum from SW and SSW (15%), with the second maximum from northeastern directions (12%, Fig. 8). However, street canyons are characterised by significant modifications of the undisturbed wind and flow conditions above the canyon (e.g., Eliasson et al., 2006; Weber and Weber, 2008; Salizzoni et al., 2009). To assess the effects of microscale flow modifications on the dispersion of aerosols within the street canyon, the mean flow and turbulence regime was analysed. A prominent feature of street canyon flow is the channelling of wind along the street canyon axis (e.g., Christen, 2005). At CAN, wind is more or less channelled along the street canyon axis for the reference wind directions 45°<ϕ_{ref}<200° and 240°<ϕ_{ref}<340° (Fig. 9). For northern reference wind directions, the channelling of flow is not strictly along the canyon axis for the bottom and middle sonic levels. This phenomenon is believed to be due to the lower building heights on the southern side of the street, thus reducing the intensity of the vortex circulation (cf. Fig. 3). At the reversal point from northerly to southerly wind directions, i.e., approximately 200°<ϕ_{ref}<240°, channelling along the canyon axis is also less pronounced. However, for all situations of cross canyon flow, a deflection of wind from the opposite house walls, indicating a vortex circulation within the canyon, can be observed. The vortex circulation at this site is described in more detail in Weber et al. (2006a,b).

Another important feature of flow and dispersion in street canyons is the significant reduction of undisturbed wind conditions (reference wind conditions). At all measurement heights within the canyon, the wind speed is significantly reduced to between 30% and 40% of the reference value (Fig. 10, left). At the canyon bottom, the wind speed is almost linearly reduced to 31% of the reference value (Fig. 10, right).

The turbulent exchange within a street canyon is crucial to determining the dispersion of pollutants. Turbulence parameters show a distinct increase in turbulence at the upper part of the canyon due to the enhanced exchange and transport of momentum from the shear layer forming across the canyon top. The shapes of the vertical profiles of the turbulent exchange at CAN (data not shown here) are in agreement with earlier measurements performed in the vicinity of this site (Weber et al., 2006a). At the reference site SUR, turbulence parameters are generally lower in comparison to the in-canyon values and are comparable to neutral surface layer scaling (e.g., Kaimal and Finnigan, 1994; Eliasson et al., 2006; Weber and Kordowski, 2010).

When turbulent exchange is classified according to the different flow regimes, interesting effects can be observed. The mean wind is largest during ALC but is smaller during CRC_{NE} and CRC_{SW}. The level of turbulent mixing as indicated by σ_w and u* increases and shows a maximum during CRC_{SW}. This phenomenon especially applies to the upper measurement level at z/H = 0.59 (Fig. 11), which has implications for the dispersion of particles, as will be described in a later section of this paper.

3.3. Aerosol size distributions and number concentrations

The average aerosol statistics at the urban street canyon and the background site indicate pronounced differences between the sites.

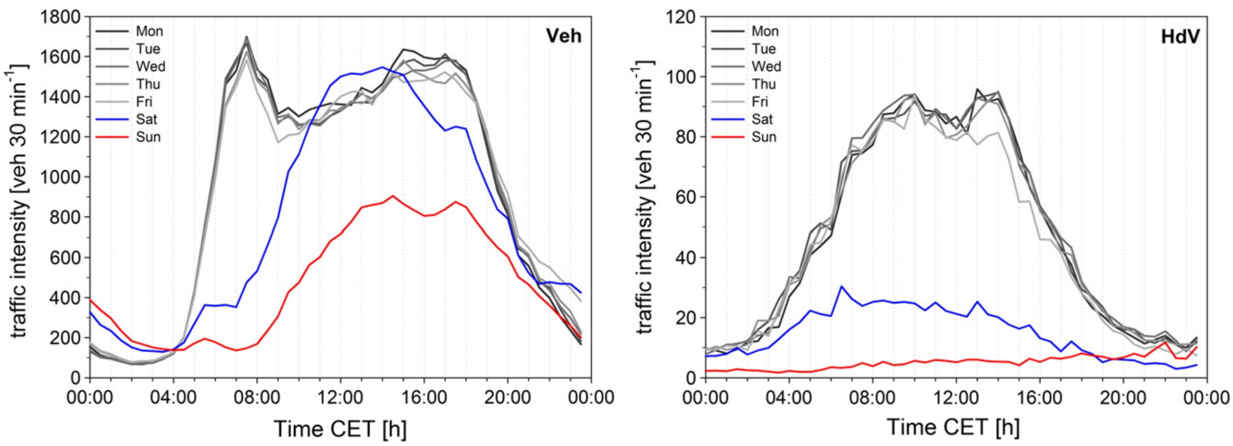


Fig. 7. Average diurnal courses of all weekdays, classified according to the traffic categories VEH and HDV (data basis: 30 min sums).

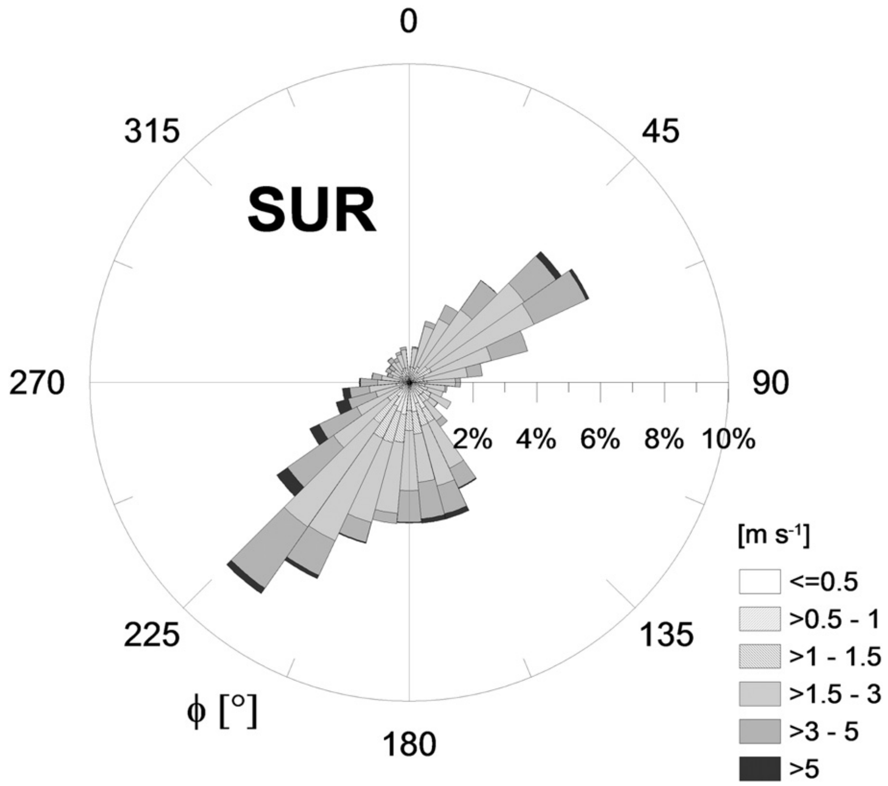


Fig. 8. Wind direction speed and frequency distribution at SUR, measured during the VAPARTICO study period.

The absolute average number concentration at CAN is 13,160 cm^{-3}, with the maximum 30 min number concentration reaching 107,760 cm^{-3} in comparison to 31,738 cm^{-3} at SUR. The total average number concentration in the size range $20<D_p<745$ nm at CAN roughly doubles the average concentration at SUR. The average number concentrations observed at CAN are somewhat lower in comparison to other European street canyon sites, e.g., approximately 17×10^3 cm^{-3} were measured on average during a winter period in Leipzig, Germany (Voigtländer et al., 2006), 40×10^3 cm^{-3} during fall conditions in Lahti, Finland (Vakeva et al., 1999) and 60–70×10^3 cm^{-3} in Stockholm, Sweden (Gidhagen et al., 2005). In a review summarising measurements at 18 roadside and 7 street canyon sites, Morawska et al. (2008) reported mean concentrations of 48×10^3 cm^{-3} and 42×10^3 cm^{-3}, respectively. However, especially within street canyons, the total particle number concentration is sensitive to different lower cut offs of the mobility spectrometers, the actual distance of the aerosol inlet to the road, the traffic intensity and ambient meteorological conditions.

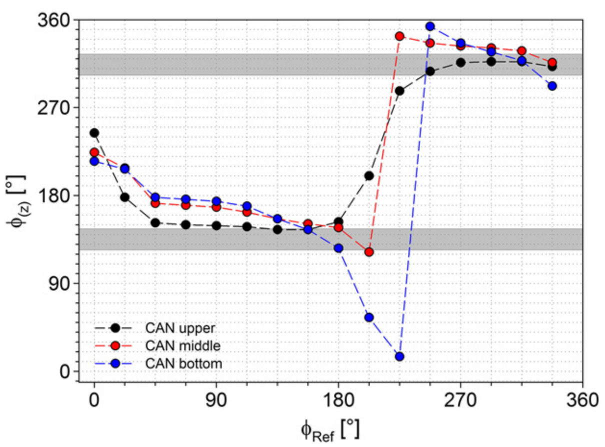


Fig. 9. Channelling of the in-canyon wind directions at the different measurement heights, dependent on the reference wind direction at SUR.

The median aerosol number size distributions at CAN are characterised by a tri-modal size distribution, with a nucleation mode peak diameter of 25 nm and a mode number concentration of 1315 cm^{-3}. The Aitken mode peaks at 45 nm with 10,070 cm^{-3} (Fig. 12, left). Both modes are comparable to other traffic sites at which aerosol concentration peaks in the nucleation mode, e.g., $15<D_p<30$ nm and typical emissions by gasoline vehicles in the size range 15–40 nm are observed (Harris and Maricq, 2001; Wehner et al., 2002; Morawska et al., 2008). Two Aitken modes at SUR, peaking at 31 and 66 nm, point to the influence of aged (traffic) aerosol from the two lane road to the N of the site (cf. Section 2).

During the day, elevated morning rush hour concentrations at CAN between 06 and 09 CET are visible in the size range from 20 to 70 nm (Fig. 12, right). The maximum number concentration with approximately 25,000 cm^{-3} is found at around 25 to 35 nm. Afterwards, concentrations gradually decrease with a rising mixing layer height; however, aerosol concentrations remain elevated in the size range 20 to 70 nm until the early evening. The afternoon rush hour, which is characterised by a higher number of passing vehicles in comparison to the morning rush hour (cf. Fig. 7), is clearly not visible in the average size distribution, which is due to a stronger instability and dispersion in the daytime boundary layer in comparison to the more stable conditions in the morning boundary layer. A ratio calculated between the sites indicates that ultrafine number concentrations at CAN are a factor 2.2 higher in comparison to SUR during the morning rush hour and remain significantly elevated throughout most of the daytime.

To evaluate the concentration surplus of particles at CAN, the parameter UFP* (i.e., the difference between the ultrafine particle concentration at CAN and SUR) was calculated (Fig. 13). The morning rush hour is characterised by a concentration surplus of approximately 8000 particles cm^{-3} at the street canyon while, during daylight hours, the concentration difference does not decrease to values

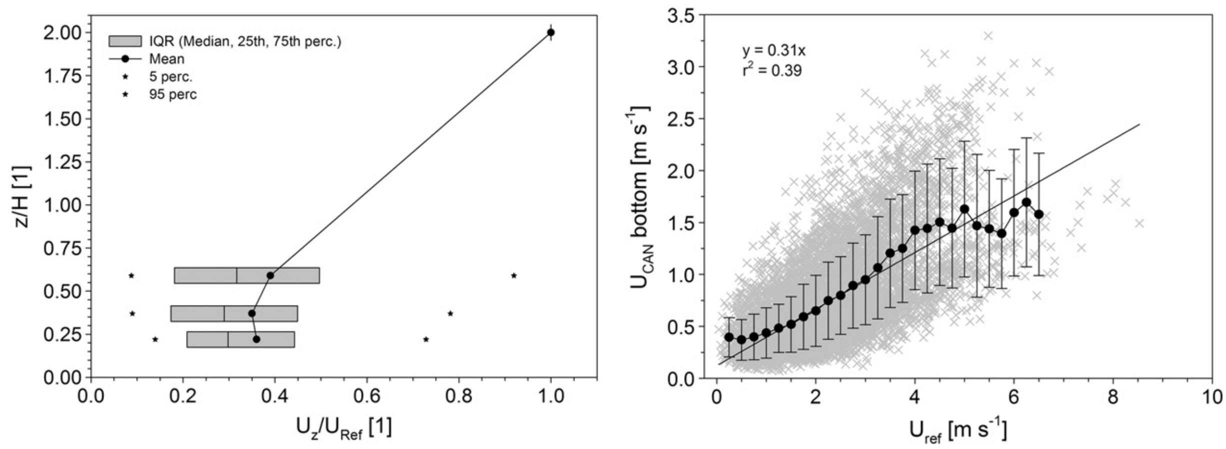


Fig. 10. Reduction of the in-canyon wind speeds at the different measurement heights, dependent on the reference wind direction at SUR (left) and the linear relationship between the bottom wind speed at z/H = 0.22 and the wind speed at SUR (right).

below 5000 to 6000 cm^{-3}. NO_x concentrations were used as a traffic indicator in other street canyon studies due to a close (linear) relationship between NO_x and total particle number concentration (Ketzel et al., 2003; Olivares et al., 2007). The normalised concentration value UFP/NO_x is somewhat constant at approximately 160 to 200×10^6 particles μg^{-1} throughout the day, indicating the close temporal relationship due to traffic as a common emission source of ultrafine particles and NO_x. This ratio is in agreement with observations from other street canyon studies (e.g., Olivares et al., 2007). This close relationship will be useful for the parameterisation of particle number concentration in Section 3.3.

The flow regimes introduced in the previous section were analysed for differences in aerosol number concentrations in different size bins (Fig. 14). Both size distributions and number concentrations at CAN are significantly different during the flow regimes, with the highest number concentration during CRC_{NE}. The shapes of the number size distributions do not change on average during a vortex-like circulation in the street canyon, but concentration differences of about $CRC_{NE}/CRC_{SW} = 1.9$ emerge. This behaviour is similar when observing the ratios in different number concentration size bins (Fig. 14, right). CRC_{SW} is characterised by the lowest concentrations because the vortex is able to transport pollutants away from the container kerbside (cf. Weber et al., 2006a), and the most intense turbulent mixing was observed for CRC_{SW} (cf. Fig. 11).

3.4. Quantification of the effects of traffic and meteorological parameters on aerosol concentrations

To study the effect of traffic and meteorological parameters on aerosol concentrations, a Spearman rank correlation analysis, including most of the measured parameters, was applied. The rank correlation analysis was performed in a first step to identify the main parameters that have a significant influence on aerosol concentrations (Table 2).

Wind speed and turbulence parameters are significantly negatively correlated to aerosol concentrations while traffic intensity shows a positive correlation (cf. Voigtländer et al., 2006). The correlation of mean wind speed is significantly lower than the standard deviation of vertical wind speed, which is a good characterisation of the intensity of turbulent mixing within the canyon. The SODAR signal in the lower level (50–99 m agl), characterising the undisturbed boundary layer wind regime, provides a similar signal in comparison with the mean wind speed at 10 m agl (SUR). However, σ_w displays the strongest correlation of the meteorological parameters, especially

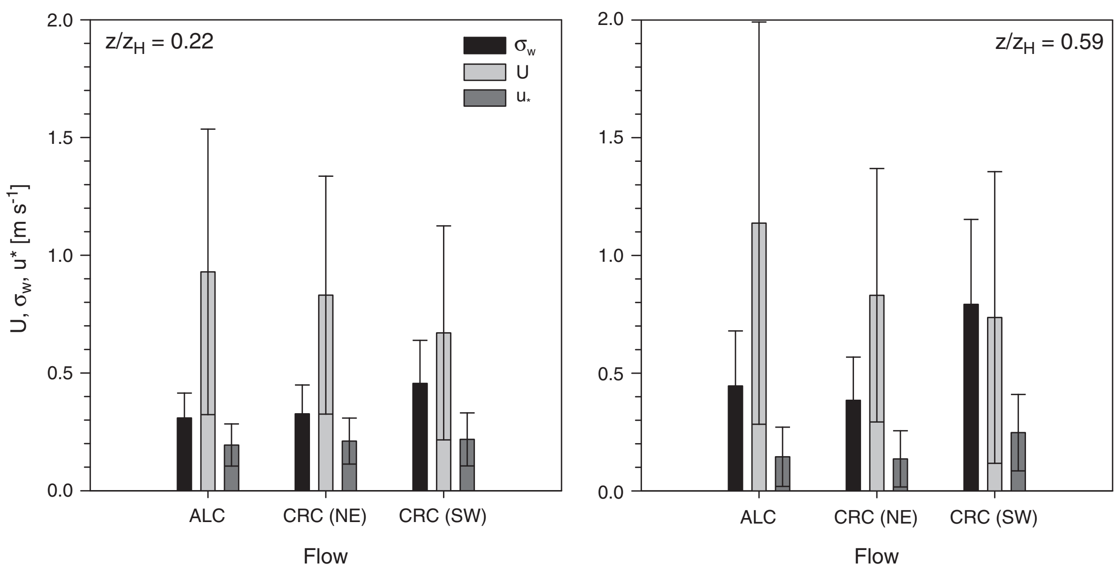


Fig. 11. Turbulence parameters classified into different flow regimes for the upper sonic at z/H = 0.59 and the bottom sonic at z/H = 0.22.

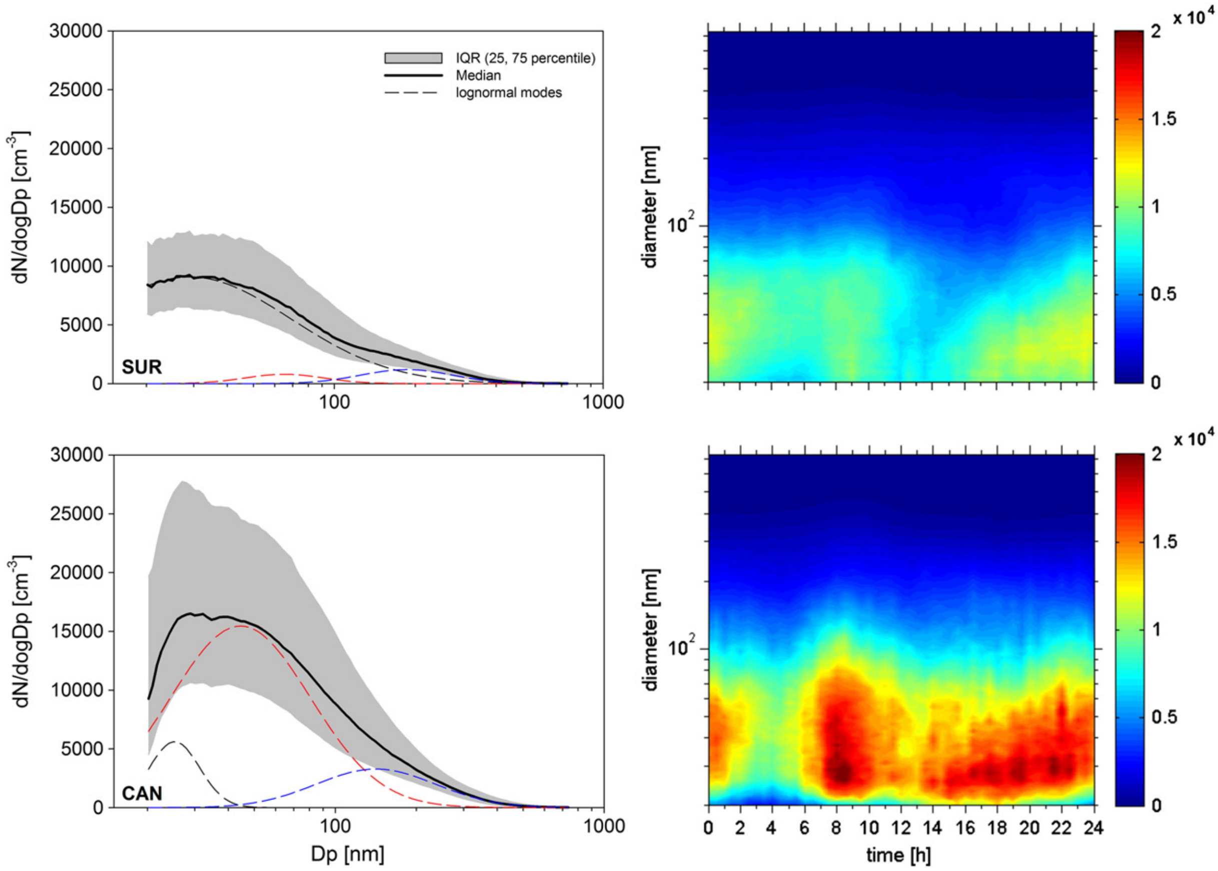


Fig. 12. Average aerosol number size distributions at CAN and SUR for the IOP. Black line indicates the median, and the grey area marks the interquartile range (25 to 75 percentile). The dashed lines indicate lognormal modes fitted to the measured size distributions (left). Concentrations are given in dN/dlogD_p.

for larger particles. The correlation of other meteorological variables (kinematic heat flux and atmospheric stability) was negligible. This finding is in agreement with Voigtländer et al. (2006), who reported wind parameters and the number of cars to be the only quantities with significant influence on measured total aerosol number concentrations ($3<D_p<800$ nm) in a street canyon in Leipzig, Germany.

In our study, traffic data contributes with the strongest signal for the smallest particle size fraction (20–30 nm) and for UFP, which is related to the peak emission of traffic in the size range < 100 nm (e.g., Morawska et al., 2008). Differences in the correlation of the traffic fleet (PCAR and HDV) with aerosol are not clearly observable. The values of the correlation coefficient are similar for both parameters, except in the 20–30 nm size range where PCAR shows a stronger signal. Generally, the magnitude of the correlation coefficient with traffic parameters is lower in comparison to other street canyon studies (e.g., Voigtländer et al., 2006). Any stronger influence of

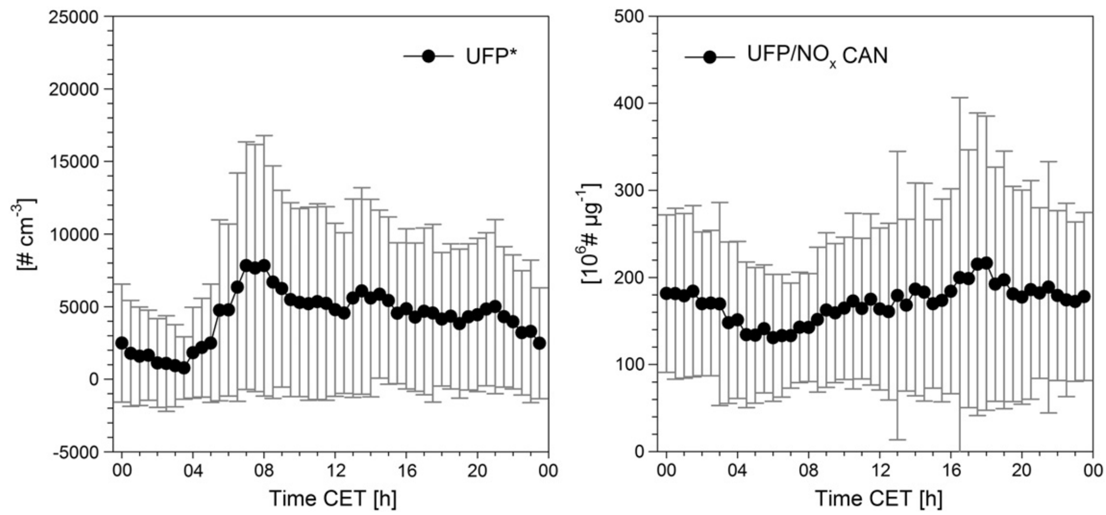


Fig. 13. Average diurnal courses of the concentration difference in ultrafine particles between the canyon and suburban background site ($UFP^* = UFP_{CAN} - UFP_{SUR}$) and the ultrafine particle concentration at CAN, normalised by the NO_x concentration at CAN.

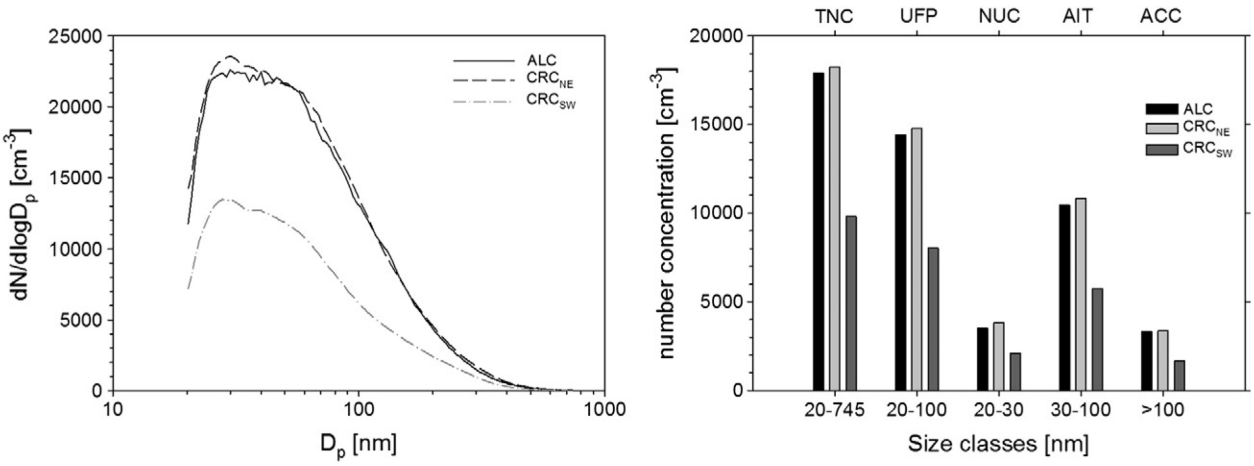


Fig. 14. Mean aerosol number size distributions (left) and number concentrations in different size bins (right) at CAN, classified into different flow regimes for the entire study period. The acronyms for the size classes refer to total number concentration (TNC), ultrafine particles (UFP), nucleation mode (NUC), Aitken mode (AIT) and accumulation mode (ACC). See the text for details on the upper and lower boundaries for the specific size classes.

mean meteorology or boundary layer meteorology, as evaluated using the SODAR or sonic anemometer measurements at SOD and SUR, was not found in the rank correlation analysis.

Based on these findings, a limited number of parameters were selected to parameterise aerosol number concentrations at CAN into different size bins using a multiple linear regression model (Fig. 15). NO_x is an 'indirect' traffic indicator and was shown to be closely linked to the (ultrafine) particle number concentration due to traffic as a common emission source (e.g., Gidhagen et al., 2004; Olivares et al., 2007). Therefore, it is a robust parameter to parameterise number concentrations and was incorporated into the statistical model.

The results of the multiple regression show a weak to moderate correlation of aerosol number concentrations with CAN_σ_w and PCAR while SUR_U and HDV were not significant. However, the strongest correlation in all size classes with $r > 0.5$ was found with NO_x concentrations. The correlation coefficients of σ_w and PCAR are characterised by a distinct variation with particle size. While PCAR shows the strongest correlation with the smaller particle size classes (the emission peak of vehicles), turbulent mixing is strongly linked to larger particles (dispersion and resuspension).

Because the total aerosol number concentration (TNC) integrates all particle size ranges and shows an 'average' correlation with all analysed size classes, it was parameterised based on the findings of the regression analysis (Fig. 16). Overall, the model performs reasonably well, i.e., the mean relative residual between observed TNC_{obs} and parameterised TNC_{par} values is 29% (median 22%) and the root-mean-square error is 4640 cm^{-3}. The explanation of variance ($r^2 = 0.68$) is in agreement with other statistical models (e.g., Mølgaard et al., 2012). The analysis of the residuals of the modelled TNC_{par} indicates an underestimation of the observed data for number concentrations > 15,000 cm^{-3}, but a distinct overestimation of TNC_{obs} at low particle concentrations, < 7500 cm^{-3} (Fig. 16, right). The mean overestimation in this size regime is approximately 50% to 75%. For particle concentrations above 7500 cm^{-3}, the model is characterised by a relative uncertainty of ±25%.

Lower particle concentrations are occurring predominantly during nocturnal and weekend periods, during which traffic flow is reduced compared to daylight weekday hours. The model shows a larger deviation because the relationship between traffic, aerosol and NO_x is less robust during these periods. Nocturnal periods might also be

Table 2
Overview of the Spearman rank correlation coefficients for the aerosol concentrations and other quantities based on 30 min averages during IOP (n = 3856). # indicates that the data are not significant at p = 0.05.

Site	Aerosol	Meteorology							Traffic[a]			
		σ_w[a]	u*[a]	w′T′[a]	U SUR[b]	K↓ SUR[b]	SODAR σ_w 50–99[c]	SODAR shear 50–99[d]	VEH	PCAR	HDV	NO_x
CAN	TNC	−0.40	−0.13	−0.17	−0.35	0.12	−0.15	−0.36	0.25	0.25	0.23	0.81
	20–30 nm	−0.12	0.05	0.06	−0.17	0.29	0.04	−0.25	0.45	0.44	0.35	0.61
	UFP	−0.36	−0.11	−0.15	−0.33	0.14	−0.12	−0.34	0.27	0.27	0.24	0.78
	30–100 nm	−0.43	−0.16	−0.22	−0.37	0.07	−0.18	−0.35	0.20	0.19	0.20	0.79
	>100 nm	−0.48	−0.18	−0.24	−0.42	0.06	−0.21	−0.36	0.17	0.16	0.20	0.84
	>1000 nm	0.03	−0.06	0.08	−0.07	#	−0.15	#	0.04	#	0.24	#
	TOV	−0.19	−0.12	−0.06	−0.24	0.06	−0.19	−0.12	0.13	0.11	0.31	0.39
ROF	>1000 nm	#	−0.08	0.12	−0.10	0.05	−0.09	−0.05	0.05	0.04	0.22	0.05
	TOV	−0.32	−0.18	−0.09	−0.31	0.03	−0.17	−0.24	0.09	0.07	0.21	0.50
SUR	TNC	−0.36	−0.31	#	−0.33	−0.11	−0.26	−0.26				0.30
	20–30 nm	−0.04	#	#	−0.08	−0.03	−0.08	#				0.18
	UFP	0.29	−0.26	−0.10	−0.27	−0.12	−0.24	−0.20				0.29
	30–100 nm	−0.38	−0.33	−0.15	−0.34	−0.16	−0.29	−0.26				0.30
	>100 nm	−0.49	−0.46	#	−0.44	#	−0.26	−0.34				0.22
	>1000	−0.07	−0.06	0.31	−0.07	#	−0.04	−0.06				0.31
	TOV	−0.30	−0.27	0.28	−0.22	0.05	−0.11	−0.18				0.23

[a] Measured at CAN, u* = friction velocity in m s^{-1}, w′T′ = kinematic heat flux in K m s^{-1}.
[b] Measured at SUR.
[c] Measured at SOD, σ_w is calculated for 10 m SODAR levels in the height range between 50 and 99 m agl.
[d] Measured at SOD, shear gives the wind shear between the SODAR levels in the height range between 50 and 99 m agl.

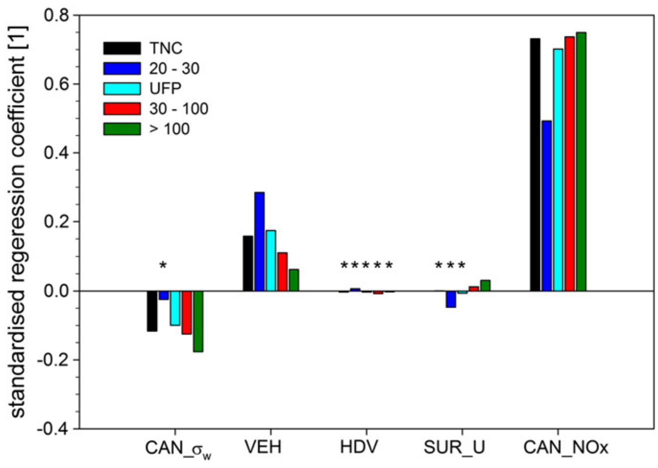


Fig. 15. Results of the multiple linear regression analysis (standardised coefficients) with the parameters σ_w, number of passenger cars at CAN (PCAR), number of heavy duty vehicles (HDV), the mean wind speed at 10 m agl at SUR (SUR_U) and the NO_x concentration at CAN. The star indicates data that are not significant ($\alpha > 0.05$).

characterised by a different NO_2/NO ratio, e.g., due to advection of NO_2 towards the site, which will affect the relationship between NO_x and aerosol. However, because only the NO_x data were available for analysis, this behaviour was not analysed in more detail.

4. Summary and conclusions

To study the effects of meteorological parameters, turbulent mixing and traffic intensity on aerosol number concentrations and size distributions, measurements at different sites were performed. The two main sites were chosen to characterise aerosol found at a suburban background site and a heavily trafficked urban street canyon. The modified flow and turbulence structure at the street canyon site and its influence on the dispersion of aerosol concentration was one of the main research tasks in VAPARTICO. Additional boundary layer measurements were performed at a regional airport to provide insight into the possible effects of "mesoscale" boundary layer processes on near-surface aerosol concentrations.

The busy urban street canyon is characterised by higher total number concentrations, with a factor of 2.2 in comparison with the background site. Ultrafine particles demonstrated a concentration surplus of approximately 8000 particles cm^{-3} during the morning rush hour. The busy street canyon aerosol has its number concentration peak at smaller size ranges of approximately 20–30 nm, which are related to traffic emissions. Aerosol concentrations in the size range below 70 nm are elevated at CAN throughout the daytime hours.

Within the urban street canyon flow regimes are complex. A vortex-like circulation during cross canyon flow (normal to the street canyon axis) is observed. The intensity of turbulent mixing is sensitive to the prevailing flow regime, with the highest turbulent mixing during cross canyon flow from southwestern directions. These situations result in the lowest number concentrations of the flow regimes within the canyon. During CRC_{SW} situations, the vortex is also able to transport pollutants away from the measurement container.

To quantify the effects of meteorology and traffic intensity on aerosol concentrations, rank correlation and multiple regression analysis were applied to the data. The rank correlation identified turbulent mixing, traffic intensity and NO_x concentrations to be best associated to aerosol number concentrations. Turbulent mixing was negatively correlated while the other two parameters exhibited a positive relationship. Based on these results, selected parameters were subjected to a multiple regression analysis to parameterise total number concentrations as integral parameters for aerosols of different size ranges. In addition to the strong relationship between NO_x and aerosol, the traffic intensity of passenger cars turned out to be the second most important variable to explain the variance in TNC. Heavy duty vehicles and mean wind speed were of negligible influence. The parameterisation of TNC with the input data σ_w, passenger cars and NO_x concentrations resulted in an overall uncertainty of 29% of TNC. However, an analysis of the residuals demonstrated two effects for different concentration regimes. While a significant overestimation occurs for low particle concentrations (e.g., $<7500\ cm^{-3}$), a moderate deviation of $\pm 25\%$ was found for particle concentrations above 7500 cm^{-3}.

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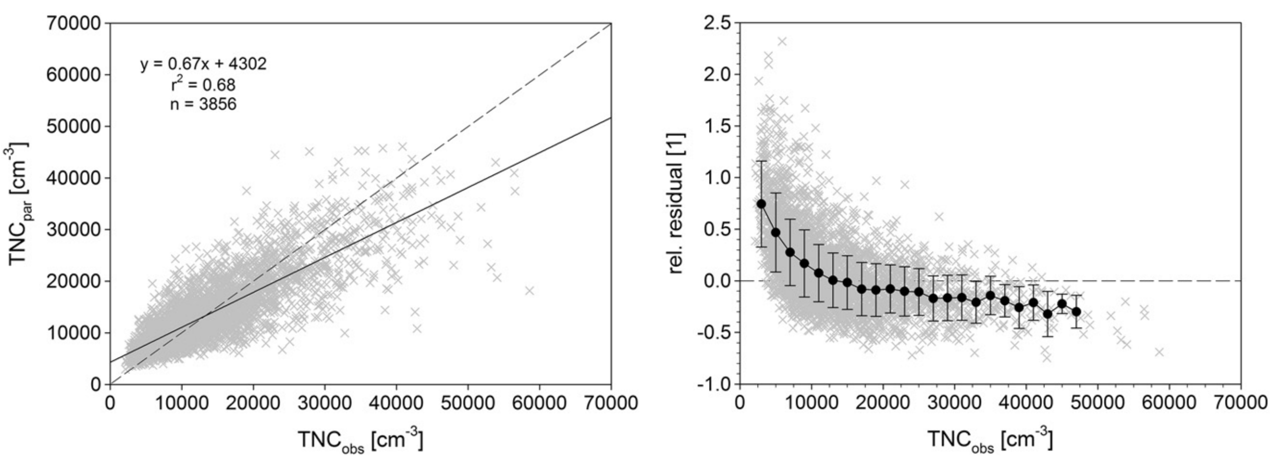

Fig. 16. (Left) Scatterplot of parameterised total number concentration (TNC_{par}) and observed total number concentrations (TNC_{obs}) at CAN, based on the results of the stepwise multiple linear regression analysis (input parameters: σ_w, PCAR, NO_x). The dashed line indicates the 1:1 line. (Right) Scatterplot of TNC_{obs} vs. residuals of the modelled TNC_{mod}. The black line indicates average relative residuals and standard deviations in 2500 cm^{-3} number concentration bins.

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