Traffic influenced nitrogen dioxide, ultrafine particle and black carbon concentrations at a busy urban street in Duesseldorf, Germany

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Abstract: In this study the road traffic related air pollution within a busy urban street canyon in the City of Duesseldorf, Germany was investigated. For the investigations the measurement truck of the laboratory for environmental measurement techniques of the HSD was equipped with different measurement devices for gaseous and aerosol species as well as meteorological sensors. The measurement truck was placed next to a stationary air quality measurement container of the Environmental State Agency of North-Rhine-Westphalia (LANUV). The measurement site is located at the Cornelius street, a direct north south connecting street through the city of Duesseldorf, which is highly frequented by road traffic with an daily amount of about 45,000 vehicles. The street is located within the low emission zone (LEZ). Additionally the pass through of vehicles with a higher weight than 7.5 tons is prohibited.

Within this study over a period of several weeks the particle mass concentrations PM_{10}, $PM_{2.5}$ and $PM_{1,}$ and the particle number size distribution (PNSD) were measured over a broad range from 5 nm to 32 µm with a high temporal resolution. Additionally, an aethalometer was used to determine the proportion of black carbon in the particle ensembles. Gaseous pollutants like nitrogen dioxide, ozone and sulphur dioxide were measured as well. Nitrogen dioxide is a typical species originating from combustion processes. In this study the data from NO_2 strongly correlate with ultrafine particle and black carbon concentrations and for this reason all three species are clearly influenced by road traffic. However, this effect is not so pronounced for the particle mass concentrations, which are additionally influenced by the varying local background concentrations.

During the measurements, the averaged concentration for PM_{10} was on a moderate level at 20.6 µg/m³ ($PM_{2.5}$ 13.4 µg/m³, PM_1 10.7 µg/m³). The averaged NO_2 concentration was 54.2 µg/m³ during the measurement period, which is above the yearly limit value of 40 µg/m³. However, the hourly limit value of 200 µg/m³ was not exceeded during the measurement period of this study. The averaged particle number concentration ($PNC_{5\text{-}250\ nm}$) and black carbon concentration were on a low level compared with values from other studies at roadsides. The $PNC_{5\text{-}250\ nm}$ was at 6,400 #/cm³ and for black carbon at 1.8 µg/m³. The lower values for PNC and BC are possibly explained as a result due to the measures from the clean air plan. The measurements of this study took place in summertime. The air pollution concentrations might be higher during colder times of the year due to domestic fuel burning. After all it can be stated, that the additional monitoring of ultrafine particles and black carbon is relevant and important to characterize the composition of the air quality at roadsides and offer a better understanding for the temporal resolution of air pollutants in cities. Moreover, estimations on the risk of air pollution for the human health can more precisely be specified.

Key-Words: urban air pollution, road traffic emissions, ultrafine particles, nitrogen dioxide, black carbon

1 Introduction

Particulate matter and nitrogen oxides are the most problematic species of air pollutants and are still one of the major problems in urban environments and cities. Air pollutants have adverse effects on the human health and can cause respiratory and cardiovascular diseases [1]. The WHO estimates that about 3 million deaths are attributed to the consequence of air pollution worldwide [2].

One of the key sources of air pollutants, especially in cities, comes from the road traffic. Beside nitrogen oxides and particulate matter, road traffic emits a broad range of pollutants like ultrafine particles, black carbon, unburned hydrocarbons, polycyclic aromatic hydrocarbons and volatile organic compounds.

Even though the development of more efficient, environmentally friendly motor engines with reduced emission is still ongoing, there is an opposing trend, which increases the issue, that the limit values of air pollutants given by the EU legislation, ratified into the German legislation by the BImSchV [3] often cannot be met. On the one hand due to the increasing size and weight of the vehicles nowadays more powerful engines have to be installed. On the other hand, the amount of vehicles on the road is still increasing over the past years and there is no indication for a reversal of this trend. When the limit values cannot be met, cities and communities have to develop measures to decrease the air pollution. In terms of clean air plans (§ 47, BImSchG), different approaches to minimize the pollution levels are elaborated. One of these measures are the low emission zones (LEZ). In the LEZ the entrance of vehicles with high emissions, which are mostly diesel-engined or older vehicles, are forbidden. A well investigated long term study by Löschau et al., describes the positive effects of a LEZ in the city of Leipzig [4]. They reported a reduction of 62 % of ultrafine particles ($PNC_{30\text{-}200nm}$), as well as 48 % for black carbon (BC) related to road traffic after five years since the introduction of the LEZ, whereas the particle mass concentration PM_{10} was reduced less than 5 %. In contrast to that the nitrogen oxides levels remain on a high level. Nevertheless it can be stated, that the reduction of PNC and BC emissions have a more significant benefit to human health, as the reduction of the particle mass concentration let expect [5].

Several studies have been performed to investigate the temporal and spatial distribution of ultrafine particles especially in cities [6–10]. In this study we measured several air pollutants by a measurement truck at a highly frequented urban road in the city of Duesseldorf over a period of six weeks during the summertime. Duesseldorf is the capital of the state North Rhine-Westphalia (NRW) and has about 600,000 residents. The Cornelius street is a direct north-south connecting street and runs through the middle of the city. The street is located in the LEZ from Duesseldorf, which was introduced in 2009 as a measure from the clean air plan. Since then, only low emission vehicles are allowed to enter the LEZ. As another measure to improve the situation of air pollution, the street is closed for vehicles with a weight over 7.5 tons, except for delivery- and local traffic and for busses and trams.

Today the whole traffic amount is up to 45,000 vehicles per day. In a previous traffic count from the years 2005 till 2011 the daily traffic was 46,200 vehicles per day [11], which is up to now, about 1,200 less vehicles. Most of these vehicles are vehicles with high emissions as well as trucks. This can be seen as a benefit from the clean air plan.

The Cornelius street has a pronounced street canyon structure, characterized by buildings without gaps on both sides of the street, roof levels of about 17 m above ground and a street width of about 30 m. This street canyon structure influences the dilution and the spatial distribution of the air pollutants and was investigated by Weber et al., [12].

At the Cornelius street, the Environmental State Agency NRW (LANUV) operates an official air quality measurement station. The LANUV measures automatically the particle mass concentration PM_{10} and $PM_{2.5}$, NO and NO_2. Other compounds like benzene, heavy metals and polycyclic aromatic hydrocarbons are measured as necessary. In Fig. 1 the averaged diurnal concentrations of nitrogen oxide are shown. The data are gathered in the time between April to September 2016 and were provided by the LANUV. On weekdays the two peaks from the rush hour in the morning and evening, with slow

moving and stop and go traffic clearly can be seen. On the weekends, this effect cannot be observed.

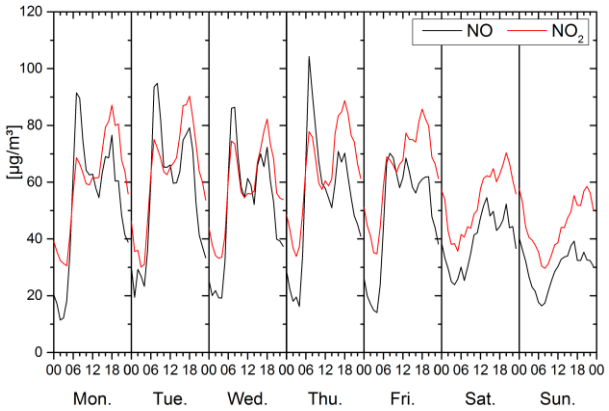

Fig. 1: Diurnal concentration of NO and NO_2 at the air quality station in the Cornelius street (Data: LANUV)

2 Methods

In the time from 22.05.2017 – 03.07.2017 we placed the measurement truck next to the air quality monitoring station from the LANUV. In addition to the air pollutants compounds measured by LANUV, we equipped the measurement truck with several measurement devices. We measured the aerosol mass concentration PM_{10}, $PM_{2.5}$ and PM_1, as well as the particle number size distribution over a broad range from 5 nm up to 32 µm. In addition, an aethalometer was used to determine the proportion of soot in the particle ensembles. Furthermore, we measured gaseous pollutants like nitrogen dioxide, ozone, sulphur dioxide and meteorological parameters like the local wind, temperature, humidity. A more detailed information about the measurement setup is given in the next subchapters.

2.1 Measurement truck

The truck we used for the investigation is a Mercedes Benz transporter which is air-conditioned and has several installation facilities for the measurement devices. It has two sampling inlets which can be extended about 80 cm above the top of the roof. One sampling inlet leads into the housing of the truck where the sampling air is distributed with a manifold to the single devices. The other sampling inlet is dedicated to the aerosol measurements and therefore separated from other sampling lines. Additional it is equipped with a nafion-dryer to reduce humidity in the sampling air. Moreover, the truck is equipped with a pylon for the meteorological sensors which can be extended to the height of 7 m over ground. The truck has installed large capacity batteries for independent operation of the measurement instruments for several hours. On Fig. 2 one can see the truck during the campaign. The measurement techniques are explained in the next subchapter.

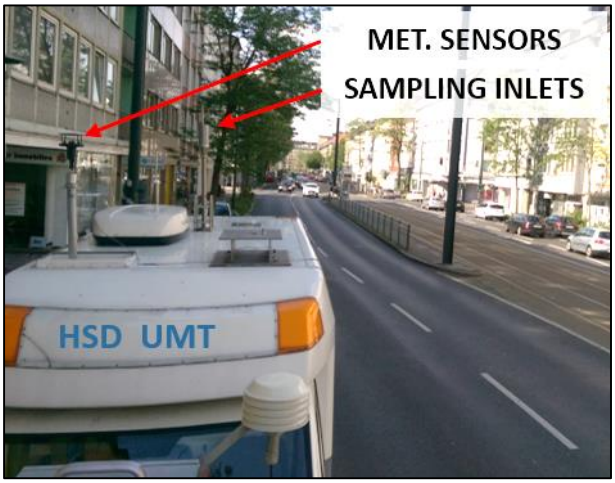

Fig. 2: View from the LANUV-measurement station along the Cornelius street. In front is the HSD measurement truck with the meteorological sensors and the sampling inlets.

2.2 Measurement techniques

2.2.1 Aerosol measurement techniques

For the detection of aerosol, we use different techniques to determine aerosols over a broad range from a few nanometers up to several ten micrometers. A scanning mobility particle sizer (SMPS) and an optical particle counter (OPC EDM180) are the main instruments for the size resolved aerosol measurement. Additionally, we equipped the truck with a Grimm HLX device which is a combination of an optical particle counter (OPC1.109) and a faraday-cup aerosol electrometer (FCAE). Both systems have a short and straight radial sampling inlet, so that the sampled aerosols in the coarse mode are not disturbed in the size distribution due to sampling losses in long sampling line. The sampling line from the main system has an extra nafion-dryer to reduce humidity in the sampling air. For the determination of soot and unburned hydrocarbons we used a seven-wavelength aethaolometer.

The SMPS (Grimm Aerosol GmbH, type 5420) is a combination of a differential mobility analyzer (DMA, type Vienna) with a condensation particle counter (CPC). The DMA uses high voltage to separate ultrafine particles with a specific diameter, which are counted afterwards with the CPC. With a known transfer function between voltage and particle mobility the device is able to classify particles from 5 nm up to 350 nm in 41 channels. The CPC uses a heated saturator with an oversaturated butanol atmosphere and a cooled condensation chamber. First the particles pass the saturator, where they adsorb some butanol. After that they pass the condensation chamber where the butanol condensates and because of this, the particles grow up by size and can be detected. Depending on the range and number of channels the time resolution is up to 4 min for a single scan over the whole range with 41 channels. One can reduce the range and number of channels to get a higher time resolution. After some premeasurements, here we used a configuration to classify ultrafine particles in a range from 5 nm to 250 nm in 22 channels with a time resolution of 120 sec. for each scan.

The FCAE (Grimm Aerosol GmbH, type NC1.320) measures the ultrafine particle number concentration as well as the size integrated particle diameter within a range from 25 nm up to 300 nm by a unipolar diffusion charger with an electric potential and Faraday detector. The time resolution is 10 seconds.

The OPCs (Grimm Aerosol GmbH, type EDM180 and 1.109) classifies and counts aerosols by measuring the scattered light from single particles and has a time resolution of 6 sec. The particles are classified by its size in 31 channels from 250 nm up to 32 µm. From the particle size number distribution obtained, the mass concentrations can be calculated. Therefore, a special calibration function is used, which is empirical evaluated for the measurement of environmental and urban aerosols.

The aethalometer (MAGEE Scientific/Aerosol d.o.o, type AE 33) is used for the determination of unburnt hydrocarbons as well as organic carbon and black carbon. A well-defined volume flow is led through a filter tape, where the particles are deposited. Thus the light transmittance of the filter material is decreasing depending on the increasing load of particles. As a light source, laser diodes at 7 different wavelengths from 370 nm up to 950 nm are used. Due to wavelength depending light absorption ability of different soot types, one can get additional information about which kind of soot is present. The temporal resolution can be adjusted variably to a minimum of one second.

2.2.2 Gaseous measurement techniques

For the measurement of nitrogen dioxide, we used a cavity attenuated phase shift spectrometer (CAPS, Environnement s.a, type AS32M). The air is lead into an optical cavity cell where an LED produces a square wave modulated light at 425 nm. At presence from NO_2 in the cell, the leak of light from the cavity will be modulated, which is a quantity of the concentration. The time resolution is about one second, but the response time (t90) is about 16 sec. For the ozone measurement we used a miniaturized UV absorption spectrometer with a light source of 254 nm (personal ozone monitor POM, 2B Technologies) The system has a special shaped measuring cell and thus, even though it is small and light weighted, has a low detection limit of 1.5 ppb_v, a resolution of 0.1 ppb_v and a time resolution from 2 sec. and a response time (t90) of 4 sec. For the measurement of sulphur dioxide we used a standard UV fluorescence analyzer (Horiba, type APSA-370) The SO_2 molecules are excited by UV radiation and emit light at a particular wavelength when returning to their energetic ground level. The amount of this emitted light is proportional to the concentration. The device has a time resolution of 5 sec. However, the response time (t90) is limited constructively at 15 sec.

3 Results

3.1 Data Acquisition

Due to the different time resolutions of each instrument, we averaged the data to 10-minutes values. Other data processing was not done.

3.2 Results

Nitrogen dioxide is a typical species from combustion processes. Therefore, we performed a

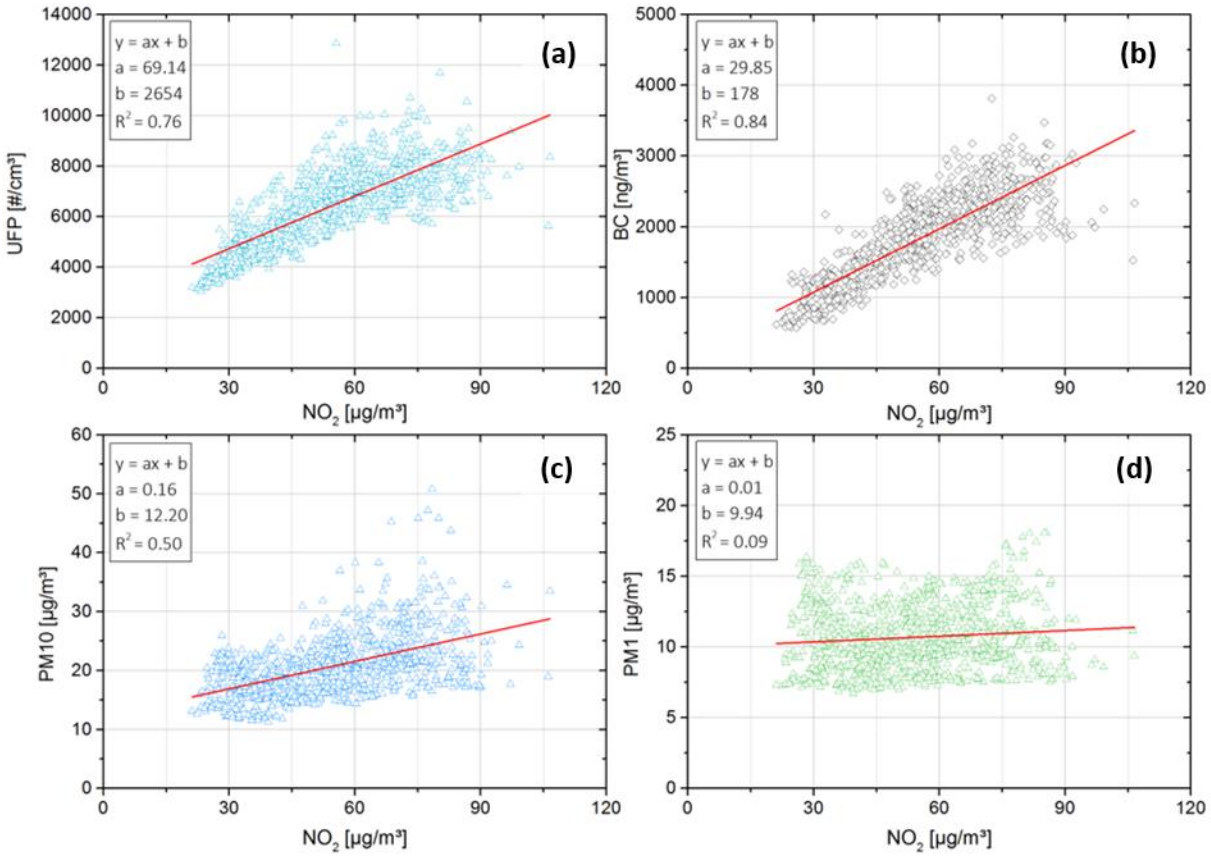

Fig. 3: Regression analysis from NO_2 with the UFP, BC, PM_{10} & PM_1

regression analysis of the data which is shown in Fig. 3. It can be seen, that there is a strong correlation between NO_2 and ultrafine particles as well as NO_2 and BC. However, the correlation between the particle mass concentration PM_{10} and NO_2 is not that good and between the PM_1 and NO_2 there is almost no positive correlation. It can be stated, that NO_2, UFP and BC are clearly influenced by road traffic. The particulate matter does not show this trend strictly within this study.

For a better characterization of the temporal distribution from the air pollutants at the measuring site, we averaged the ten-minute values of each day to generate the diurnal trend over the whole week. In Fig. 4 the results are shown. In the upper graph (a) the particle mass concentration for PM_{10} and PM_1 in µg/m³ are shown. In (b) is the particle number concentration $PNC_{5\text{-}250\ nm}$ in #/cm³, in (c) there is the black carbon concentration in ng/m³ and in the last graph (d) is the concentration of NO_2.

It can be seen, that the diurnal trends for nitrogen dioxide, $PNC_{5\text{-}250\ nm}$ and black carbon corresponding strongly to each other and show the same characteristic peaks as seen in the data from LANUV in Fig. 1. However, this effect is not so pronounced for the particle mass concentrations, which are additionally influenced by varying local background concentrations.

A basic trend can be observed for the three traffic influenced species, following the amount of road traffic. During the weekdays at around five a.m. the concentrations for the traffic influenced species begin to strongly increase up to a first peak between seven a.m. and ten a.m., which is followed by a short decreasing. At twelve a.m. the values increase again and have their second maximum peak between five and six p.m. In some cases, after the rush hour in the morning the concentrations stay on a high level during the whole day, without the short sink of the values at noon. Between seven p.m. and eight p.m. the concentrations begin to sink continuously and reach their daily minimum between three and four a.m. in the night. During the weekend, the values show a deviating trend. On Saturdays the daily

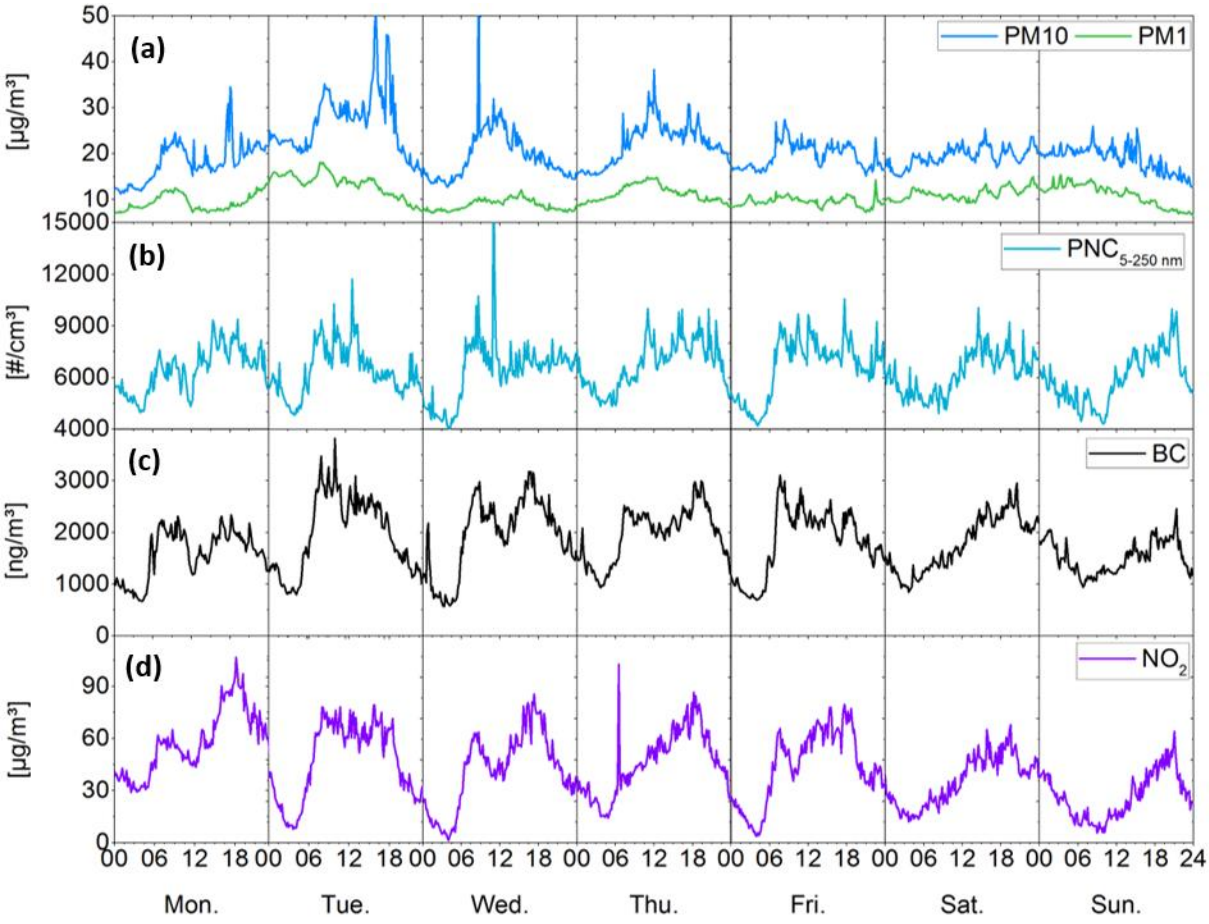

Fig. 4: Diurnal trends for particle mass concentration PM_{10}, PM_1, particle number concentration ($PNC_{5\text{-}250\ nm}$), black carbon and nitrogen dioxide.

minimum is reached earlier in the night at around two a.m. After six a.m. the values start to increase slowly to have their maximum at seven p.m. This can be traced back to visitors which do shopping or cultural activities in the city. At Sundays the daily minimum can be observed only at six a.m. A possible reason might be, during Saturday night, there is private traffic and traffic by taxies for people with Saturday night activities. Moreover, the minimum is exceeded until ten a.m. Then the values increase slowly again during the day and have their maximum at ten p.m. The time-shifted maximum might be explained to commuters which arrive or leave the city in the early evening. After ten p.m. the values drop significantly. Finally, it has to be mentioned, that the diurnal trends are idealized due to averaging and there might be deviations on individual days.

The concentrations for PM_{10} and PM_1 are on a moderate level. The averaged concentration for PM_{10} was at 20.6 µg/m³ ($PM_{2.5}$ (not shown in the graph) 13.4 µg/m³) and for PM_1 10.7 µg/m³. The values from NO_2 were on a much higher level and range from 20 µg/m³ at nighttime up to 105 µg/m³ at daytime. The averaged NO_2 concentration was 54.2 µg/m³, which would be above the yearly limit value from 40 µg/m³, if the concentration would stay at this level. Due to the fact that the measurements took place in summertime, the values might increase by domestic fuel burning during the colder times of the year. Also the reduction of NO_2 by O_3 will be less in the winter because of the attenuated solar radiation and the minor sunshine duration.

For the ultrafine particles, the $PNC_{5\text{-}250\ nm}$ has a minimum of 4,000 #/cm³ at night and a maximum of 12,000 #/cm³ at day. Due to averaging the data, short time peaks can be much higher. Compared to values from other studies in terms of investigating ultrafine particles at roadsides [4,7] the averaged values within this study are at 6,400 #/cm³, which is at the lower limit.

The concentration of black carbon at nighttime is about 600 ng/m³ and up to 4,000 ng/m³ at daytime. Typical yearly concentrations at roadside measurement station are between 2.5 µg/m³ and 3.5 µg/m³ [13]. The average BC concentration in the present study was lower at 1.8 µg/m³. This may be explained for two reasons. Baumbach et al., [13] describe the percentage distribution of BC sources. They show that 37 % is related to road traffic (51 % incl. other transportation traffic e.g., inland shipping, rail traffic, etc.) and about 40 % related to domestic fuel burning. Therefore, the BC concentration might be higher during the colder times of the year. Another reason is, that there is maybe a possible effect of the LEZ and the additional prohibition on the Cornelius street for vehicles with a weight greater than 7.5 tons. This effect could also be observed at a roadside measurement station in the city of Leipzig described by Löschau et al. [14]. There, the amount of BC was decreased about 48 % since the activation of the LEZ. The yearly averaged BC values are of the same order of magnitude as in our observations. The proportion of soot in the particle mass concentration PM_1 is about 17 % within this study.

4 Conclusion

The data showed, that there are typical air pollution species like NO_2, ultrafine particles and black carbon, which are strongly influenced by the combustion processes from the road traffic. These pollutants agree well together in the regression analysis. However, this effect is not so pronounced for the particle mass concentrations PM_{10} and PM_1, which are additionally influenced by the varying local background concentrations.

The temporal distribution of NO_2, ultrafine particles and black carbon show a typical trend, which can be traced back to the amount of road traffic. The minimum values are observed at night at around four a.m. During the day there are two daily maximums. One is between seven a.m. and ten a.m. and the second between five p.m. and six p.m. Both maxima can be traced back to the increased amount of traffic during the rush hours. At the weekend the temporal behavior is different. The daily minimum is time shifted and can be observed in the early morning hours. After six a.m. at Saturdays and ten a.m. at Sundays the values begin to increase to reach their maximum at seven p.m. at Saturdays and ten p.m. at Sundays. Finally, it has to be mentioned, that the diurnal trends are idealized due to averaging and there might be deviations on individual days.

During the measurement period the averaged particle mass concentrations for PM_{10} was on a moderate level at 20.6 µg/m³ ($PM_{2.5}$ 13.4 µg/m³ and PM_1 10.7 µg/m³) and under the yearly limit value (PM_{10} 40 µg/m³, $PM_{2.5}$ 25 µg/m³). The averaged NO_2 concentration was on a relatively high level at 54.2 µg/m³. Assuming that the NO_2 concentration would stay at this level, the yearly threshold from 40 µg/m³ would be exceeded. However, the hourly limit value from 200 µg/m³ was not exceeded during the measurement period. The averaged particle number concentration ($PNC_{5\text{-}250\ nm}$) and black carbon concentration were on a low level compared with values from other studies at roadsides. The $PNC_{5\text{-}250\ nm}$ was at 6,400 #/cm³ and for black carbon at 1.8 µg/m³. The lower values for PNC and BC are possibly explained as a result due to the measures from the low emission zone and the additional prohibited entrance of vehicles with a weight above 7.5 tons. Similar positive effects from LEZ was reported by Löschau at al., [14]. Five years after the introduction of the LEZ in Leipzig, only a small reduction for the particle mass concentration PM_{10}, but a very clear reduction for the ultrafine particles and black carbon could be observed. Because the measurements took place in summertime, the values for all the measured air pollutants might be higher due to domestic fuel burning during the colder times of the year.

After all it can be stated, that the additional monitoring of ultrafine particles and black carbon as well, are relevant and important parameters to characterize the composition of the air quality next to roads and offer a better understanding for the temporal resolution of air pollutants in cities. Moreover, estimations on the risk of air pollution for the human health can more precisely been specified.

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