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Article

Direct measurement of CO_2 and particle emissions from an urban area

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Abstract

From July 9th through September 24th, 2009, turbulent particle number fluxes and CO_2 fluxes were measured above the city area of Münster, north-west Germany. The goal was to characterize the respective vertical fluxes in the urban boundary area. The measurements were conducted at a height of 65 m a.g.l. on a military radio tower at 10 Hz temporal resolution. Fluxes were calculated applying the eddy covariance method. To determine the impact of traffic emissions on particle number fluxes and CO_2 fluxes, hourly traffic activities for 45° sectors, related to the tower, were calculated. Averaged diurnal and sectoral fluxes are consistently directed upward, implying that the urban area of Münster acts continuously as particle (number) and CO_2 source.

Traffic activities vary in the course of the day and within the 45° sectors. The latter is attributable to differences in land use between the sectors. In the course of the day, two peaks are discernible, during the morning and the evening rush hours, respectively. Averaged diurnal particle (number) fluxes are correlated significantly to traffic activity. Accordingly, traffic related emissions are the main sources for urban particle (number) fluxes. Averaged sectoral CO_2 fluxes also correlate fairly well with sectoral traffic activities. In addition, daytime photosynthesis is a controlling variable for the CO_2 flux, leading to lower upward fluxes in daytime. The contribution of the photosynthetic activity of the vegetation in the urban area to the CO_2 flux is quantified. Further, the contribution of traffic related emissions to the CO_2 flux is computed by applying emission factors for carbon dioxide to the traffic activity. They contribute in daytime about 40 to 50 % to the CO_2 flux, whereby, nightly contributions are minimal.

Zusammenfassung

Vom 9. Juli bis zum 24. September 2009 wurden in Münster, Nordwest-Deutschland, turbulente vertikale Partikel-Flüsse und CO_2-Flüsse gemessen. Die Messung wurde in einer Höhe von 65 m über Grund auf einem Funkturm der Bundeswehr mit zeitlicher Auflösung von 10 Hz durchgeführt. Die Berechnung der Flüsse erfolgte unter Anwendung der Eddy-Kovarianz-Methode. Um den Einfluss verkehrsbedingter Emissionen auf die Partikel-Anzahl-Flüsse und die CO_2-Flüsse zu bestimmen, wurden stündliche Verkehrsbelastungen für 45° Sektoren, vom Messturm ausgehend, berechnet. Die gemittelten Flüsse sind im Tagesverlauf und innerhalb der Sektoren durchgängig aufwärts gerichtet. Das bedeutet, dass das Stadtgebiet Münsters durchgehend als Partikel-Anzahl- und CO_2-Quelle fungiert.

Die Verkehrsbelastungen variieren im Tagesverlauf und innerhalb der 45° Sektoren. Zweiteres ist Unterschieden in der Landnutzung zwischen den Sektoren zuzuordnen. Im Tagesverlauf sind zwei Maxima, während der morgendlichen und der abendlichen Hauptverkehrszeit, erkennbar. Die für den Tag gemittelten Partikel-Flüsse korrelieren signifikant mit der Verkehrsbelastung. Dementsprechend sind verkehrsbedingte Emissionen die Hauptquelle für den städtischen Partikel-Anzahl-Fluss. Die sektorspezifisch gemittelten CO_2-Flüsse korrelieren ebenfalls sehr gut mit der jeweiligen Verkehrsbelastung. Neben dem Einfluss der Emissionen verschiedener urbaner Quellen ist auch die photosynthetische Aktivität eine entscheidende Einflussvariable für die Tagesverläufe des CO_2-Flusses. Der Anteil der photosynthetisch aktiven Vegetation im Stadtgebiet auf den CO_2-Fluss wurde quantifiziert. Um den Anteil der Emissionen des Straßenverkehrs auf den CO_2-Fluss zu berechnen, wurden Emissionsfaktoren für CO_2 auf die Verkehrsbelastung angewendet. Der Anteil des Straßenverkehrs auf den CO_2-Fluss liegt tagsüber bei 40 bis 50 %, wohingegen der Anteil nachts minimal ist.

1 Introduction

Carbon dioxide and aerosol particles have an important impact on climate change. The high contribution of carbon dioxide to climate forcing is well-known and is associated with only small uncertainties (IPCC, 2007). In contrast to that, aerosol particles have, due to their direct and indirect effects, a negative influence on climate forcing. There are much larger uncertainties in these processes. Therefore the quantification of surface-fluxes of carbon dioxide and, in particular, aerosol particles are of great interest for global warming estimates and research (IPCC, 2007). Furthermore, there is an adverse health effect of fine aerosol particles, especially in urban areas (IBALD-MULLI et al., 2004). There are strong indications that urban areas are often both a sink for particulate matter, dominated by larger particles, and a source for particle numbers, dominated by fine and ultra-fine particles (NEMITZ et al., 2000; DORSEY et al., 2002; SCHMIDT and KLEMM, 2008). SEINFELD and PANDIS (2006) subdivide urban particle sources in four parts: Two parts are combustion of fossil fuel and industrial processes. As the third part, there are non-industrial sources such as construction sites, wind erosion and re-suspension. Finally, there are emissions from traffic, which are not associated with the combustion process itself but rather with, for example, the abrasion of brakes and tires. The high loads of particles in the urban air and their negative health effects lead to the directives 1999/30/EC and 2008/50/EC of the European Union. These directives include regulations for limits and exceedance of air pollutants. Consequently, knowledge about urban particle dynamics and properties are of great interest for various actors.

Because of its impact on climate change, carbon dioxide is one of the most challenging gases for science (e.g. PAWLAK et al., 2009). Especially measurements of CO_2 concentrations and fluxes in urban areas, which are main sources for anthropogenically emitted carbon dioxide, contribute to knowledge about the dynamics of this greenhouse gas (VOGT et al., 2006). Emissions of carbon dioxide directly affect the local CO_2 concentrations, and, due to the long atmospheric lifetime of CO_2, it indirectly affects the global atmospheric conditions (e.g. IPCC, 2007). By interaction between the anthropogenic sources and the natural sink, including the photosynthetic activity of the vegetation, a complex system of exchange between surface and atmosphere in urban areas evolves (VOGT et al., 2006). In the past, CO_2 emissions were mainly quantified based on estimates of the consumption of fossil fuel (GRIMMOND et al., 2002). More recent approaches predominantly are based on direct measurements in or above urban areas (e.g. GRIMMOND et al., 2002; NEMITZ et al., 2002; VOGT et al., 2006; SCHMIDT et al., 2008). But measurements of carbon dioxide in or above cities are underrepresented compared to measurements in natural environments. Due to the importance of cities as main anthropogenic sources of CO_2, recently more and more studies focus on urban areas (BURRI et al., 2009). Because of climatic and structural variability of cities, any generalization of results from urban areas is limited (GRIMMOND et al., 2004; VOGT et al., 2006). Results of studies that treat urban CO_2 emissions, are available for, e.g., Chicago (GRIMMOND et al., 2002), Edinburgh (NEMITZ et al., 2002), Marseilles (GRIMMOND et al., 2004), Tokyo (MORIWAKI and KANDA, 2004; MORIWAKI et al., 2006), Basel (VOGT et al., 2006), Sapporo (MIYAOKA et al., 2007), London (RIGBY et al., 2008), Helsinki (VESALA et al., 2008), Münster (SCHMIDT et al., 2008), Cairo (BURRI et al., 2009), and Lodz (PAWLAK et al., 2009).

For both aerosol particles and carbon dioxide, road traffic is an important source in urban areas. For CO_2 in Münster, the calculated contribution of traffic to the annually averaged emission is about 27 % (AMT FÜR GRÜNFLÄCHEN UND UMWELTSCHUTZ MÜNSTER, 2005). According to BEZIRKSREGIERUNG MÜNSTER (2009), 82 % of urban emissions of particulate matter smaller than 10 μm (PM10) originate from traffic, whereas only 13 % and 5 % arise from industry and smaller firing systems, respectively. If the regional background and long-range transport is included, only 10 to 25 % of PM10 immissions originate from traffic, and about 75 to 90 % are attributable to regional background. That agrees very well with results of GIETL et al. (2008), where about 23 % of the particle mass concentration at a roadside was related to traffic emissions and 77 % reflected the regional background concentration. Similar rates are published by MOLNAR et al. (2002) and KETZEL et al. (2004) with at least 50 % of the particle mass concentration in urban air typically originating from regional or long range transport. For particle number concentrations, much less information is available. Sub-micrometer aerosol particles are released due to combustion of fossil energy and due to abrasion of brakes, tires, and road surface. GARG et al. (2000) calculate an average amount of 35 % for abrasion of brakes of vehicles for emitted particulate matter to the atmosphere. Another source for particle emissions is re-suspension of deposited particles of natural or anthropogenic origin (ROGGE et al., 1993). Because of differences in composition and amount of emissions, vehicles are divided in vehicle classes such as light duty vehicles (LDV), heavy duty vehicles (HDV), busses, and motorcycles. In addition to this, vehicles can be classified by size, age, and technology of the engine. In KIRCHSTETTER et al. (1999), the emitted particle number per unit mass of fuel burned for diesel-powered HDV is described as 15 to 20 times higher than for LDV. This is similar to results of WANG et al. (2010) from a study from Copenhagen, where HDV emit 20 times more particles than LDV, and IMHOF (2005), where computed emission factors for HDV are 10 to 30 times higher than for LDV.

The aim of this study is to examine the characteristics of particle and carbon dioxide emissions for the urban area of Münster. The results should give insights in their diurnal behaviours. Especially the impact of traffic as a source of particle number and carbon dioxide and the differences in land use within the urban area of Münster on emissions are of major interest. The existing knowledge about urban particle and CO_2 emissions should be further developed, and a comparison of our results to other studies from other cities should reveal differences and similarities in the respective behaviour.

2 Methods and material

2.1 Study and site descriptions

From July 9th through September 24th 2009, the turbulent fluxes of carbon dioxide and aerosol particle number were measured in Münster. The city of Münster is located in the Northwest of Germany. With about 274 000 inhabitants, Münster is a regional center surrounded by smaller cities and villages alternating with agricultural areas. Despite of the rural appearance, Münster acts as a major carbon dioxide and particle number source, comparable to other cities (SCHMIDT and KLEMM, 2008).

The measuring system was mounted at the top of a military radio tower at 65 m a.g.l. in the urban area near the city center (Fig. 1). The location is about 40 m above the rooftops of the surrounding buildings, so that direct influences by nearby particle or carbon dioxide sources are excluded. Furthermore, because of the measuring height, the data are representative for large footprint source areas (up to 1 km^2) within the urban area (SCHMIDT and KLEMM, 2008). The sensors were mounted 2 m above the upmost level of the tower to minimize any interference by the structure of the tower. In the surrounding of the tower, the sector 1 and 2 (0°(N) to 90°(E)) from the measurement location is dominated by residential areas with suburban structure, frequently combined with gardens. Adjacent to that, there are agricultural areas and small forested patches. In the sector 3 and 4 (90° to 180°), the potential source area is rather heterogeneous. Parts contain residential areas like in the NE sector, parts contain allotment garden grounds, agricultural areas, and forests. The little inland harbour and a few medium sized industrial areas are located towards South within the sectors 4 and 5. In the sectors 5 and 6 (180° to 270°), there are also the train station, the cargo train station, densely built-up areas, and less green areas. The recreational area "Aasee", which covers 90 ha, starts in 2 km distance from the tower at about 260°. The sector 7 and 8 (270° to 360°) contain the main part of the city center, which is a densely build-up residential and central business area. Beneath the city center, dense residential areas, university buildings, and small green areas are dominating.

2.2 Measurement

The measuring system consists of three instruments: The 81000V Ultrasonic Anemometer (R. M. Young Company, Traverse City, Michigan 49686, USA) measures the speed of the three wind components u, v, w, and the sonic temperature. The Li-7500 Open Path CO_2/H_2O Infrared Gas Analyzer (LI-COR, Inc., Lincoln, Nebraska 68504, USA) measures the densities of CO_2 and H_2O by absorption of infrared light. The aerosol particle number concentration ($D_p > 11$ nm) was measured with the Condensation Particle Counter Model 3760A (TSI Incorporated, St. Paul, Minnesota 55126, USA). To transport the aerosol through the 3.4 m silicon sampling tube (diameter: 0.5 cm), a flow of 1.5 lpm was produced by an external pump and confined by a critical orifice. For optical detection n-butane is condensed onto the aerosol particles, which leads to an enlargement of the particles. The instruments were operated with a temporal resolution of 10 Hz. According to the predominant south-westerly wind direction, the sensors and sensor inlets were located detached but close to each other, so that errors caused by the setup are kept at a minimum.

The distances from the sonic anemometer to the particle inlet and the CO_2/H_2O infrared gas analyzer were 20 cm and 30 cm, respectively.

2.3 Traffic data

A total of about 159 000 motor vehicles are licensed in Münster, whereof 87 000 vehicles are petrol-driven and 70 000 are diesel-powered. The remaining 2 000 vehicles are gas-powered (BEZIRKSREGIERUNG MÜNSTER, 2009). Data to the traffic situation, which is used in this study, is based on different data sources provided by the department of urban planning in Münster (Stadtplanungsamt Münster), and a traffic census by GIETL and KLEMM (2009). The available data sets had been partly gathered manually, and partly gathered automatically through hourly traffic counting over several years (2001 to 2008) in the urban area of Münster. Also a database for daily averaged traffic load on main roads was utilized. Traffic data of several years is used with the assumption that the variation of traffic from year to year can be neglected. From these data the average traffic activity in units of travelled vehicle-kilometers per ground area and per time unit ($\mathrm{veh\,km\,m^{-2}\,s^{-1}}$) for 45° sectors from the tower are computed. Averaged diurnal variations are calculated. In addition, variations of traffic activity between weekdays and weekends are acknowledged. While this data should reflect the traffic activity appropriately for long-term observations, differences between the actual traffic situation and the calculation are conceivable. Potential causes are variations due to changing weather conditions, seasonal variations, holiday season, or major construction sites. A circular area with 3 km radius centered around the tower was used to calculate the traffic activity from available traffic data. Data within the circle was considered, whereas data from outside the circle was excluded. This area reflects the dominating footprint areas for fluxes as measured at the tower based on results of SCHMIDT and KLEMM (2008), who calculated source areas applying the methods of SCHMID (1994). An overview to the calculation of footprints is given in SCHMID (2002). The highest traffic intensities can be expected in the sectors 5 to 8 (180° to 360°) because of industrial areas and a dense building structure, which is related to heavy duty vehicles and an intensive passenger car flow, respectively.

2.4 Eddy covariance

Flux measurements by use of the eddy covariance technique are implemented by highly resolved direct measurements, which are calculated into fluxes by algorithms based on several assumptions and simplifications (FOKEN, 2008). It is important to perform the measurements within the surface layer or in a way that no internal boundary layer could exhibit any impact on the measurements, respectively. Due to the measuring height of 65 m a.g.l., the impact of surrounding structures is excluded to the greatest possible extent. With regard to the requirements for eddy covariance measurements, the commonly used formula for turbulent fluxes calculated by the eddy covariance is:

$$F = \overline{w'x'} \tag{1}$$

where *F* is the flux over 30 minutes, *w* is the vertical wind speed, and *x* is the variable (scalar) for which the flux is calculated. Primes indicate the difference of individual 10 Hz measurements from the 30-min average of the scalar, whereas the overbar indicates the 30-min average. Consequently, the right-hand side of equation 1 is the covariance between *w* and *x*.

Turbulent eddy fluxes indicate the feature of the surface to act as a source or sink for the scalar *x* under investigation. To include temporally and spatially high resolved turbulent elements, data are recorded with 10 Hz. In return, fluxes are calculated over 30-minute intervals to ensure that also large turbulence elements with low frequencies are incorporated. Wind measurements in complex surroundings are affected by flow distortions due to buildings, setup construction, or other sensors (DYER, 1981, 1982; GRIESSBAUM and SCHMIDT, 2009; MENNEN et al., 1996; WIERINGA, 1980). In this study, flow distortion only arises from the structure of the tower and nearby sensors. The correction of the flow distortion is performed by using the method of GRIESSBAUM and SCHMIDT (2009). The mean corrections from flow distortion range from 4 % up to 7 %, the median range from 6 % up to 10 %. The small CO_2 sensor head is not considered in the simulation, but the data from 322.5° through 37.5° from the anemometer are excluded from data analysis due to potential flow distortion as caused by the northern location of CO_2 sensor head. According to SCHMIDT and KLEMM (2008), data quality for the flux computation is tested and assured by performing time lag correction, based on shifting the data with the maximum value of an autocorrelation analysis, coordinate rotation for the streamline fit (PAW U et al., 2000; WILCZAK et al., 2001), linear detrending, WPL correction to correct fluctuations of density induced by turbulent exchange of latent and sensible heat (WEBB et al., 1980), stationarity testing (FOKEN and WICHURA, 1996), and the calculation of the integral turbulence characteristics (ITC). The transport of the aerosol through the sampling tube leads to an attenuation of the fluctuation, which produces an underestimation of the fluxes. This effect is corrected following MASSMAN (1991), LENSCHOW and RAUPACH (1991) and HELD and KLEMM (2006). The mean correction for the attenuation of the fluctuation is 11.6 % and the median correction is 7.3 % with 3.4 % as 5 % percentile and 23 % as 95 % percentile. To ensure a good data quality, further quality tests are implemented (FOKEN, 2008). After exclusion of incorrect data out of over 3 000 30-minute intervals, approximately 900 intervals for the particle flux and about 1 100 intervals for the CO_2 flux are available for further analysis.

3 Results and discussion

3.1 Traffic activity

The calculated average traffic activity for weekdays and weekends is presented Fig. 2. The Saturdays are analyzed together with the Sundays as weekends because the differences between Saturdays resemble each other very much, whereas the other days of the week show a typical and different weekday pattern (see also WANG et al. (2010)). In the following weekend describe Saturdays and Sundays and weekday describe Mondays to Fridays. For weekdays, two peaks can be identified, representing the morning and afternoon rush hours, respectively. In the morning, the traffic activity increases rapidly from 05 hrs to 08 hrs local time. After that, the averaged traffic activity remains more or less stable until the afternoon rush hour starts at 14 hrs. On workdays, the maximum traffic activity is reached at 17 hrs, when the commuter traffic and the residual, not commuter-related traffic, which is far greater during afternoons than during mornings, arise simultaneously. On weekends, the traffic activity differs. Because of strongly reduced commuter traffic, the morning and afternoon rush hours are not discernible. The only maximum is reached at 15 hrs. With the exception of higher nightly traffic activity on weekends from 00 hrs to 05 hrs, which is related to nightlife activities, weekend traffic is persistently lower than that on weekdays. Similar patterns to the variation of traffic from Copenhagen are shown in WANG et al. (2010).

The averaged traffic activities as differentiated in 45° sectors with reference to the measuring site, are shown in Fig. 3. For the eastern sectors, the calculated traffic activities are considerably smaller than those of the western sectors. This characteristic is attributable to the land use of the sectors (Fig. 1), with dense urban areas in the western sectors and heterogeneous aspects in the eastern sectors, as outlined in section 2.1.

The amount of HDV, including buses, is computed to be between 5 and 13 % and between 4 and 8 % of the hourly averaged values for weekdays and weekends, respectively. Overall, HDV contributes 7 %. The overall amount of vehicle kilometers in Münster is 5.4×10^6 veh km d^{-1} (BEZIRKSREGIERUNG MÜNSTER, 2009) and about 2×10^6 veh km d^{-1} for the considered circular area around the experimental tower, with 3 km radius.

3.2 Measured particle number flux and urban sources

To describe the relation of the results of particle flux measurement to the calculated traffic activities, the sectoral traffic activities are compared in Fig. 3 to aerosol particle fluxes for the corresponding wind directions. The correlation is fairly good. With the exception of two southern sectors, the variations in traffic activity with respect to the wind direction are reflected in the corresponding averaged sector fluxes. In the eastern sectors, the values for averaged sectoral particle number fluxes, in correspondence to the minor traffic activities, are comparatively low, whereas the particle number fluxes in the western sectors are considerably higher. These similarities indicate a strong impact of traffic activity on measured particle emission fluxes. In the sector 4 (135° to 180°), the road network is less dense. The influence of traffic on particle emission is less, but other particle sources are more pronounced. The variability of fluxes within this sector is large. Source of this large variability is not clear, but it is likely to be related to a large variability of non-traffic emission sources. In the sector 5 (180° to 225°), the ratio of main roads to smaller roads is higher than anywhere else. This probably leads to an overestimation of traffic intensity in this sector.

Averaged particle number fluxes for weekdays and weekends are shown in Fig. 4. For workdays the hourly averaged flux ranges between $11 \times 10^6\ \mathrm{m^{-2}\ s^{-1}}$ and $151 \times 10^6\ \mathrm{m^{-2}\ s^{-1}}$. The minimum value occurs at 04 hrs and the maximum at 12 hrs, respectively. At 05 hrs, the measured particle fluxes increase sharply, which is related to the morning rush hour with intense traffic activity. According to the values of the traffic activity, the diurnal cycle of the particle flux shows similar behaviour. However, a rather striking difference consists in the absence of an afternoon peak for the particle flux. During daytime, there is a well developed turbulence regime (AGARWAL et al., 1995) and a deeper boundary layer, which results in efficient dilution of particles. Thus, the maximum of the flux occurs at noon rather than during afternoon rush hour. Similar results with a maximum at noon in particle fluxes are described e.g. for Edinburgh (DORSEY et al., 2002), Stockholm (MÅRTENSSON et al., 2006), and Helsinki (JÄRVI et al., 2009). The correlation between particle flux and traffic activity ($\mathrm{R}^2 = 0.60$ for weekdays, $\mathrm{R}^2 = 0.57$ for weekends) is significant and confirms the influence of traffic to the particle flux. The offsets ($\mathrm{F}_0 = 16 \times 10^6\ \mathrm{m^{-2}\ s^{-1}}$ for weekdays, $\mathrm{F}_0 = 27 \times 10^6\ \mathrm{m^{-2}\ s^{-1}}$ for weekends) represent other urban particle sources, such as industry or smaller firing systems. Compared to traffic related emissions, other sources contribute only little to particle emissions. Although some values for 30-minute fluxes are negative and reflect downward directed fluxes, all averaged values are positive. So, the particle number fluxes are in average consistently directed upward. That implies that the urban area acts as a continuous particle number source. For weekends, the hourly averaged flux ranges between $14 \times 10^6\ \mathrm{m^{-2}\ s^{-1}}$ and $134 \times 10^6\ \mathrm{m^{-2}\ s^{-1}}$. The minimum occurs again at 04 hrs, and the maximum at 14 hrs, respectively. In comparison to fluxes on workdays, the morning increase is retarded and less pronounced. The maximum is almost as high as on weekdays. Overnight particle fluxes on weekends are slightly higher than on workdays. That is attributable to enhanced overnight traffic activity on weekends. Moreover, even on weekends, the urban area of Münster acts as particle source and there are no downward directed averaged fluxes. Individual 30-minute downward fluxes are infrequently occurring exceptions.

To validate our results, fluxes are calculated by applying emission factors on the traffic activity of Münster. The used emission factors for a mixed vehicle fleet, with

$1.4 \times 10^{14}\ \mathrm{veh}^{-1}\ \mathrm{km}^{-1}$, arise from a study of MÅRTENSSON et al. (2006) to similar flux measurements in Stockholm. Due to similar mean contributions of HDV in Münster (7 %) and Stockholm (6 %) (MÅRTENSSON et al., 2006), respectively, the errors resulting from the use of emission factors for a mixed vehicle fleet, are neglectable. The results of the calculation of diurnal fluxes for the considered circular area resemble the measured particle fluxes, especially for weekends, and show the complimentary nature of our measurement and the emission factors for a mixed fleet in Stockholm. The calculated flux is based on traffic activity and the emission factors. The calculated weekday flux shows an afternoon peak. This is the main difference between our measurement and the calculated flux. Considering the similar magnitude of the measured and the calculated fluxes, we hypothesize that the employed emission factors from Stockholm are overestimations for the traffic, typical for the urban area of Münster. If the data as presented in Fig. 4 is expected to be valid, there would be no more room for any particle emissions from other sources. This contradicts the results from diurnal emission analysis as shown above. Possible reasons for this contradiction are differences in amounts of the various emission sources between urban areas of different cities. Possibly the amount of traffic as particle source is larger in Stockholm as compared to Münster. As another potential reason, there are possibly lowered emissions of vehicles, due to improved technology of the engines between 2002 and 2009.

3.3 Measured CO_2 flux and urban sources

A comparison between carbon dioxide fluxes and traffic activity for 45° sectors is shown in Fig. 5. Throughout, in the sectors 1 to 4 (0° to 180°), representing heterogeneous land use source areas, the fluxes are smaller than those from the sectors 5 to 8 (180° to 360°), which represent the city center and densely built-up areas. Although traffic is only one of several urban sources, a clear dependence is discernible. It is conceivable, that traffic as a carbon dioxide source plays an important and controlling role for the spatial variation. The largest variations of carbon dioxide fluxes appear in the range from 90° to 225° (sectors 3 to 5), while the fluxes in the other sectors occur more consistent.

Averaged diurnal carbon dioxide fluxes for weekdays and weekends, respectively, are shown in Fig. 6. For workdays the hourly averaged fluxes range between $3.8\ \mu\mathrm{mol\ m}^{-2}\ \mathrm{s}^{-1}$ at 10 hrs and $11.2\ \mu\mathrm{mol\ m}^{-2}\ \mathrm{s}^{-1}$ at 08 hrs. The diurnal course features a maximum in the morning, which is a result of increased emissions from traffic and coincides with the morning rush hour. The flux sharply decreases to a minimum at 10 hrs and stays rather constant, exhibiting a slight increase, though. This low level is related to photosynthesis of the vegetation, which picks up CO_2 during daytime. It reduces the ground level carbon dioxide concentration and, thus, the net emission flux. In the evening, when photosynthesis is completed and carbon dioxide concentrations rise again due to plant respiration and anthropogenic sources, an increasing CO_2 emission flux evolves. The decrease of the flux in the early morning hours can be explained by extreme low urban emissions. Despite the photosynthetically induced daily depression of the flux, no net downward fluxes occur in our averaged data sets. So, on weekdays, the urban area acts as a net source of carbon dioxide.

On weekends, the diurnal appearance of carbon dioxide fluxes, with an hourly averaged flux ranging between $1.8\,\mu\mathrm{mol\,m^{-2}\,s^{-1}}$ at 15 hrs and $7.5\,\mu\mathrm{mol\,m^{-2}\,s^{-1}}$ at 23 hrs, is different from workday fluxes. Due to the absence of a morning rush hour, no clear peak appears in the diurnal cycle. Likewise on weekdays, the flux decreases during daytime because of the photosynthetic activity of the urban vegetation. Towards evening, when uptake of CO_2 by the vegetation ceases, the carbon dioxide flux increases and levels out at about $6\,\mu\mathrm{mol\,m^{-2}\,s^{-1}}$. The measured fluxes are comparable to results to urban CO_2 fluxes from Melbourne (COUTTS et al., 2007), Helsinki (JÄRVI, 2009), and Essen (KORDOWSKI and KUTTLER, 2010). Especially the results from Essen (KORDOWSKI and KUTTLER, 2010), which is located in north-west Germany and is subjected to similar environmental conditions, validate our measurements during summer due to the measurement period covering one year. Despite the lower fluxes by day on weekends as compared to weekdays, again the averaged carbon dioxide flux direction is consistently upward. The vegetation in Münster is apparently incapable of balancing the ground level CO_2 emissions.

In the following, we quantify the contribution of the photosynthetic activity of the vegetation to the CO_2 flux. A regression between CO_2 flux and particle number flux during night time (22 - 05 hrs) is determined (Fig. 7). This correlation is interpretable under the assumption that the particle number emission flux and the CO_2 emission flux behave similarly during the nights. Under these conditions, for both fluxes the main controlling variables are the urban emissions. Traffic is the major source for both fluxes.

The linear regression (coefficient of determination $(R^2) = 0.18$; correlation coefficient $(R) = 0.43$) is applied on the particle fluxes of the entire day to derive the CO_2 fluxes that would reflect the urban emission, without the mitigating effect of the vegetation. The result (included in Fig. 6) is an averaged CO_2 flux without a daytime depression. The computed flux "without" vegetation impact increases until 11 hrs on weekdays and 14 hrs on weekends. Towards evening, the fluxes decrease until the minimum is reached at 04 hrs for weekdays and weekends. These hypothetical fluxes range from $4.7\ \mu\mathrm{mol\ m^{-2}\ s^{-1}}$ at 4 hrs to $11.3\ \mu\mathrm{mol\ m^{-2}\ s^{-1}}$ at 14 hrs and $5.2\ \mu\mathrm{mol\ m^{-2}\ s^{-1}}$ at 04 hrs and $10.7\ \mu\mathrm{mol\ m^{-2}\ s^{-1}}$ at 14 hrs for weekdays and weekends, respectively. Certainly, these values are projected and hence reflect a possible behaviour with potential discrepancy from real conditions.

To further validate this calculation and to directly quantify the contribution of traffic emissions to the CO_2 flux, this traffic related CO_2 flux is computed by applying emission factors for carbon dioxide to the traffic activity. The emission factors, which contain a differentiation between HDV and LDV, originate from a study from the project report by BOULTER et al. (2009). These emission factors were compared and updated with a differentiation between emission factors for petrol-driven and diesel-powered vehicles from other studies (e.g. BARTELT, 2005) to ensure a good quality of emission factors for traffic related carbon dioxide emissions. The fluxes range from $0.3\ \mu\mathrm{mol\ m^{-2}\ s^{-1}}$ at 04 hrs to $6.3\ \mu\mathrm{mol\ m^{-2}\ s^{-1}}$ at 17 hrs and $0.4\ \mu\mathrm{mol\ m^{-2}\ s^{-1}}$ at 05 hrs and $4.3\ \mu\mathrm{mol\ m^{-2}\ s^{-1}}$ at 15 hrs for weekdays and weekends, respectively. In addition to the annually averaged distribution of sources as mentioned above (AMT FÜR GRÜNFLÄCHEN UND UMWELTSCHUTZ MÜNSTER, 2005), this study offers more detailed information about diurnal variations of sources and their impact on carbon dioxide fluxes. The computational strategies to compute the CO_2 flux from traffic emissions is similar for the two methods. On the one hand, a CO_2 emission factor is directly applied to the traffic pattern. On the other hand, an experimentally determined CO_2 / particle number emission ratio is applied to the particle number flux. The latter leads to the emission estimate "without vegetation". The results of the two methods are very similar to each other. Deviations occur on weekday afternoons, when the maximum for the averaged CO_2 fluxes without daytime depression is at 11 hrs compared to 17 hrs for traffic emission related CO_2 fluxes. The main difference between the two results consists in an offset between the two fluxes. This offset represents the flux attributable to other, non-traffic CO_2 emitters, with a more or less stable baseline level between 2 $\mu\mathrm{mol\,m^{-2}\,s^{-1}}$ and 6 $\mu\mathrm{mol\,m^{-2}\,s^{-1}}$ for both, weekdays and weekends. Considering the diurnal courses of the computed fluxes, it is obvious that the diurnal variation of the measured carbon dioxide flux is mainly caused by traffic emissions. The nightly fluxes on weekdays, related to traffic emission, are small and contribute only marginally to the CO_2 fluxes. During daytime, the contribution of traffic-related fluxes to the total CO_2 flux is approximately 50 %. On weekends, similar diurnal amounts are achieved with slightly smaller contributions in daytime (about 40 %), though. For weekdays the diurnal averaged amount of traffic emissions to the CO_2 flux is 40 % and 28 % for weekends. This agrees fairly well with results of AMT FÜR GRÜNFLÄCHEN UND UMWELTSCHUTZ MÜNSTER (2005), where the average annual amount of traffic emissions to the CO_2 flux is 27 %. Due to lower emission from smaller firing systems during summer, when this measurement took place, the amount of traffic emission is overestimated in comparison to all year averages. The main remaining amount is attributable to energy related emissions. During 2005, a total of 556000 t originated from traffic related emission, and 1207000 t from energy related emission (AMT FÜR GRÜNFLÄCHEN UND UMWELTSCHUTZ MÜNSTER, 2005). The impact of the remaining emitters has a more constant distribution in the course of the day. Assuming an absence of traffic emissions contributing to the fluxes, the urban area of Münster probably would act as carbon dioxide sink in daytime. Likewise, a reduction of emissions of the other sources could affect a daytime downward flux and reduce the total urban carbon dioxide emission.

3.4 Error discussion

The error discussion is included to be able to assume the quality of the data presented in this paper. Errors can be made at the data acquisition and the data processing. The first is related to the inaccuracy of the used instruments and the execution of the measurements. Since measurement errors are excluded in the following data processing, errors of the acquisition of data are limited to instrument inaccuracy. Manufacturers state the coincidence of the Condensation Particle Counter as about 10 % at 10000 particles per cm^3 (TSI Incorporated, St. Paul, Minnesota 55126, USA), the inaccuracy of the Ultrasonic Anemometer as < 3 % for the wind speed and $< 5°$ for the wind direction (R. M. Young Company, Traverse City, Michigan 49686, USA), and similar values for the inaccuracies of the CO_2/H_2O Infrared Gas Analyzer (LI-COR, Inc., Lincoln, Nebraska 68504, USA). FOKEN (2008) describes the accuracy of the applied eddy covariance technique to be 5 to 10 % with regard to the described requirements. Since traffic data are provided, the statements to the inaccuracy have to be limited to the data processing. The error, which is made by using data from several years, is neglected due to only marginal changes of the traffic conditions in that period. Another smaller error, which is made in the processing of the traffic data, is the selection of data from the circle with 3 km radius for the calculation of the traffic activity. Due to the dominating footprint areas for fluxes measured at the tower this area is chosen to be the basis for traffic activity calculation. Summarized the inaccuracy of traffic activity data can be assumed as < 20 %. The traffic related fluxes, calculated by application of emission factors to the traffic activity, adopt the errors made by calculation of the traffic activity. On the assumption that the vehicle fleet of Münster is comparable to the vehicle fleet on which the calculation of the emission factors is based, further errors are neglectable. Errors made by calculating the CO_2 flux without impact of the vegetation is charged with larger uncertainties. Due to the statistically insignificant correlation coefficient (Fig. 7) the values for this calculated flux only can be considered to be an estimation. With respect to this insignificancy and the associated uncertainties the calculation is presented in this study due to the discernible plausibility of this estimation (Fig. 6). Accordingly the results to the CO_2 flux without impact of the vegetation have to be treated carefully.

4 Summary and conclusions

The measurement of the aerosol particle number flux and the CO_2 flux, applying the eddy covariance method, above the urban area of Münster is focused on the impact of urban emissions on the fluxes and the relative contributions of different sources. The measurement height of 65 m a.g.l. gives access to large flux source (footprint) areas, reflecting the net emission from the urban area in a better way than ground based measurements. The general interest lies in the flux regime for particle number and carbon dioxide above the urban canopy. Special interest lies on traffic as a main driver for both particle number and CO_2 emissions. We analyzed its influence as a function of wind direction and diurnal variation.

Therefore, traffic data and measured data are divided spatially into 45° sectors from the tower, and temporally into hourly periods. Furthermore, a distinction of the data between weekdays and weekends is made. Particle number fluxes vary between sectors, reflecting the magnitude of the traffic activity in the associated sector fairly well. Accordingly, diurnal traffic activities (Fig. 2) and diurnal particle number fluxes (Fig. 4) correlate significantly ($R^2 = 0.60$ for weekdays, $R^2 = 0.57$ for weekends). Especially the impact of the morning rush hour on particle number fluxes is clearly identifiable. The ordinate offsets of the regression between traffic activity and measured particle flux ($F_0 = 16 \times 10^6\ m^{-2}\ s^{-1}$ for weekdays, $F_0 = 27 \times 10^6\ m^{-2}\ s^{-1}$ for weekends) represent other urban particle sources. The impact of traffic related emissions as main particle number source is confirmed by these minor offsets for non-traffic related particle sources on the one hand, and by the calculation of traffic related particle number fluxes based on emission factors for traffic from a similar study from Stockholm on the other hand. According to the major amount of traffic emissions to particle fluxes, reductions of particle upward fluxes is most likely achievable by reducing urban traffic activity.

For carbon dioxide fluxes, similar characteristics can be observed. The CO_2 fluxes vary between the sectors, representing heterogeneous land use source areas. Although traffic is only one of several urban sources, a clear dependency is discernible. It is thus conceivable that traffic emissions as CO_2 source have a controlling impact on the spatial variation. Averaged diurnal carbon dioxide fluxes are mainly influenced by the CO_2 emissions and daytime photosynthesis (Fig. 6), with smaller emissions in daytime and higher ones during the nights. Specifically high upward fluxes were observed on weekdays during the morning rush hour before the photosynthetic activity started. The contribution of the photosynthetic activity of the vegetation in the urban area to the CO_2 flux is quantified by applying a regression between CO_2 flux and particle number flux during night time. The result (Fig. 6) shows clearly increased carbon dioxide fluxes during daytime. This calculation is validated by computing the contribution of traffic emissions to the CO_2 flux, based on applying emission factors for carbon dioxide to the traffic activity. The diurnal courses of these two calculated fluxes are similar, so that the main contribution to the diurnal variation of the flux can be allocated to traffic emissions. The offset between the fluxes represents the flux attributable to the other CO_2 emitters. Assuming a significant reduction of traffic could be achieved, the carbon dioxide emissions from the urban area of Münster probably would become negative in daytime.

This study provides an accurate characterization of the spatial and temporal behaviour of measured particle number and CO_2 fluxes for the urban area of Münster. With respect to their possible errors and uncertainties, further characterizations, based on the calculated fluxes, show interesting details of the spatial and temporal behaviour. To achieve a more exact source allocation, a more detailed and greater database for traffic activity and a database for the other urban CO_2 sources would be useful. An increased duration of the measurement could improve the representativeness of the results. For particle flux characterization, size resolved fluxes in combination with chemical analysis would help to expand the view towards particle mass, rather than just particle number, fluxes.

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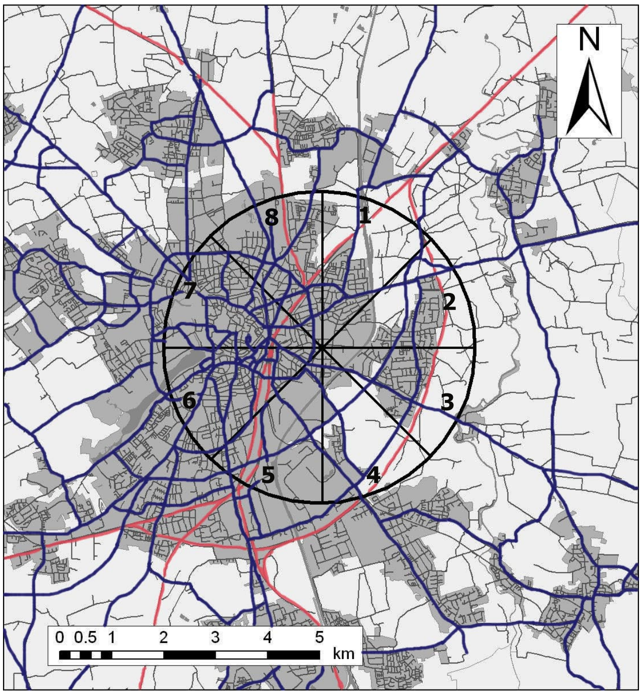

Figure 1: Map of Münster. The measurement tower is located in the center. The pie diagram insert represents the 45° sectors with 3 km radius. The grey shade symbolizes the urban area of Münster, blue and red lines symbolize main roads and rail tracks, respectively.

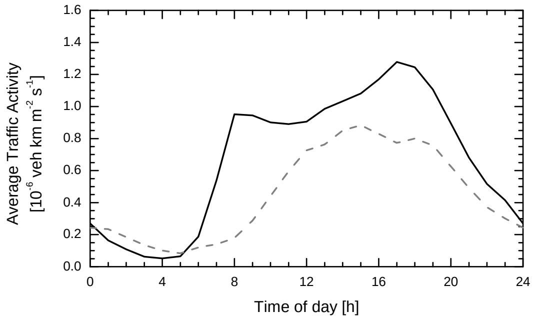

Figure 2: Calculated diurnal average traffic activity for weekdays (Mondays to Fridays), as black line, and weekends (Saturdays and Sundays), as grey broken line.

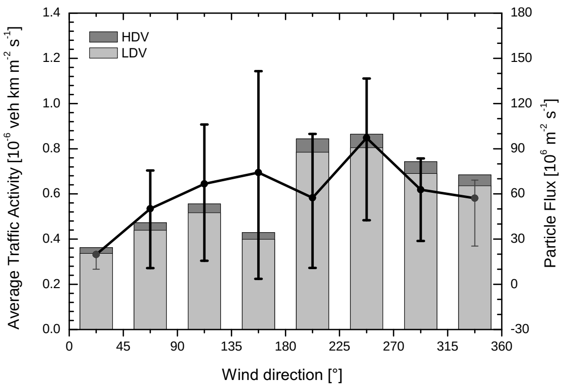

Figure 3: Traffic activity and particle number flux averaged over 45° sectors. Dark grey color represents heavy duty vehicles (HDV) and light grey color represents light duty vehicles (LDV).The particle flux is illustrated as black line with 25 % and 75 % percentile. The flux of the northern sectors is visualized as thin dark grey lines since the data basis is reduced due to the impact of the sensor head.

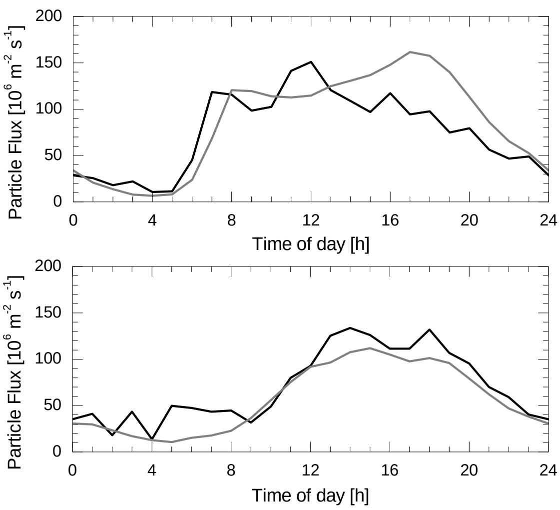

Figure 4: Measured diurnal particle number flux (black line) and calculated diurnal particle number flux (grey line) for weekdays (top panel) and weekends (bottom panel). The calculated diurnal particle number flux is based on traffic activity (section 2.3) and emission factors for traffic.

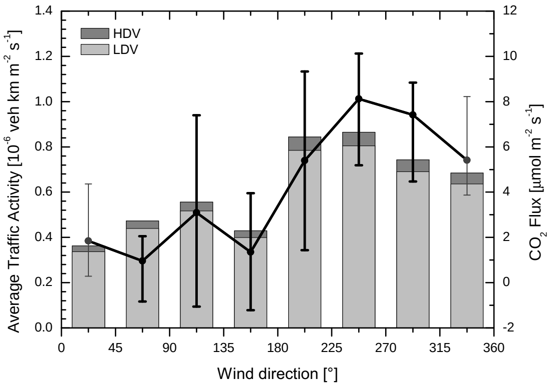

Figure 5: Traffic activity (see also Fig. 3) and carbon dioxide flux averaged over 45° sectors. Dark grey color represents heavy duty vehicles (HDV) and light grey color represents light duty vehicles (LDV). The CO_2 flux is illustrated as black line with 25 % and 75 % percentile. The flux of the northern sectors is visualized as thin dark grey lines since the data basis is reduced due to the impact of the sensor head.

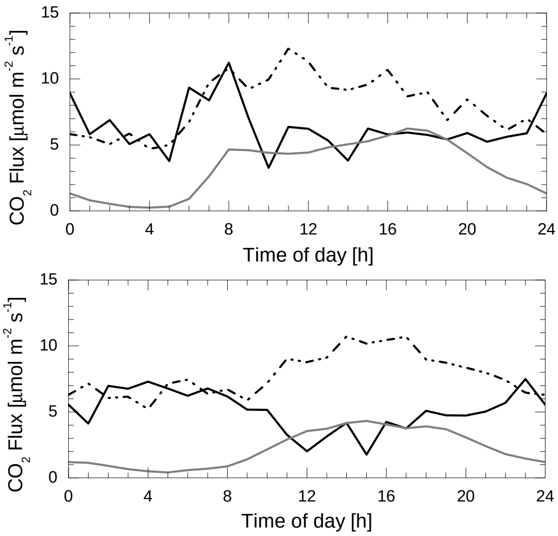

Figure 6: Measured diurnal CO_2 flux (black line), CO_2 flux with excluded impact of vegetation (black broken line) and calculated diurnal CO_2 flux (grey line) for weekdays (top panel) and weekends (bottom panel). The calculated diurnal CO_2 flux is based on computation out of emission factors for traffic and traffic activity.

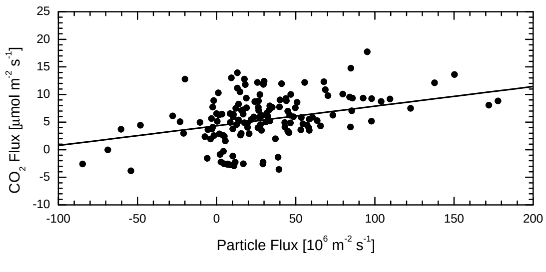

Figure 7: Correlation of particle number flux and CO_2 flux over night to estimate the vegetation impact on CO_2 fluxes. The black line represents the linear regression between particle number flux and CO_2 flux ($R^2 = 0.18$).