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The relationship between 0.25–2.5 µm aerosol and CO_2 emissions over a city

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Abstract

Unlike exhaust emissions, non-exhaust traffic emissions are completely unregulated and there are large uncertainties in the non-exhaust emission factors required to estimate the emissions of these aerosols. This study provides the first published results of direct measurements of size resolved emission factors for particles in the size range 0.25–2.5 µm using a new approach deriving aerosol emission factors from the CO_2 emission fluxes. Because the aerosol and CO_2 emissions have a common source and because the CO_2 emission per fuel or traffic amount are much less uncertain than the aerosol emissions, this approach has obvious advantages. Therefore aerosol fluxes were measured during one year using the eddy covariance method at the top of a 118 m high communication tower over Stockholm, Sweden. Maximum CO_2 and particle fluxes coincides with the wind direction with densest traffic within the footprint area. Negative fluxes (uptake of CO_2 and deposition of particles) coincides with an urban forest area. The fluxes of CO_2 were used to obtain emission factors for particles by assuming that the CO_2 fluxes could converted to amounts of fuel burnt. The estimated emission factors for the fleet mix in the measurement area are, in number 1.4×10^{11} [particle veh^{-1} km^{-1}]. Assuming spherical particles of density 1600 kg/m^3 this corresponds to 27.5 mg veh^{-1} km^{-1}. Wind speed influence the emission factor indicating that wind induced turbulence may be important.

1 Introduction

Road traffic is one of the major contributors to air pollution in many urban areas (Ruuskanen et al., 2001; Gidhagen et al., 2005). Airborne particulate matter (PM) may be expressed in terms of number mass, surface area, or volume (Harrisson et al., 2000), but PM_{10} and $PM_{2.5}$ are the usual metrics used in regulations of air pollution within Europe. This despite the fact that information about bulk particle mass concentrations and emissions of PM_{10} or $PM_{2.5}$ is of limited value for assessing climate and health

ACPD
10, 21521–21545, 2010

effects of aerosol pollution. Particle size resolved information on emissions is urgently needed to understand processes controlling emissions and the importance for health and climate. In order to make accurate air quality and traffic measurements, good source apportionment is needed. This in turn means that emission inventories should include relationships between meteorology, traffic intensity, fuel load, etc.

A number of studies have been made and approaches used (see below) to quantify road traffic emissions for different applications.

The

- laboratory dynamometer test, which provides emission factors (EFs) for individual vehicles including gasoline/diesel light duty vehicles and heavy duty EFs (see e.g., Westerholm and Egeback, 1994; Sjögren et al., 1996; Hall et al., 2001; etc.)
- car-chasing experiments and the FEAT-technique provide EFs for individual vehicles in real world driving (Kittelson et al., 2000; Sjödin and Lenner, 1995).
- open-road studies Open-road studies are based on a combination of roadside measurements of air pollutants and models to account for the dispersion of the exhaust gas plume. Information about the evolution of EFs for PM_{10} and PM_1 (Gehrig et al., 2003) as well as for particle number, active particle surface area, and black carbon (BC) (Jamriska and Morawska, 2001) has been derived using this technique.
- road tunnel measurements which provide EFs from the entire fleet during partly real-world conditions (e.g. McLaren et al., 1996; Kristensson et al., 2004; Colberg et al., 2005; Hueglin et al., 2006).
- eddy covariance method which provides also EFs for entire fleet for actual real world conditions (Dorsey et al., 2002; Mårtensson et al., 2006; Martin et al., 2008; Järvi et al., 2009). It requires fast response instrumentation.

In this study we focus on emission measurements using the eddy covariance method. An advantage of this method is that it can provide information about the emission

ACPD
10, 21521–21545, 2010

from a large vehicle fleet during real-world driving and under the influence of different meteorological conditions that might affect the emissions. On the other hand the obtained emissions fluxes are strongly dependent on wind direction and the footprint of the measurement site. Measuring vertical fluxes allow us to develop accurate and efficient parameterizations (Dorsey et al., 2002; Mårtensson et al., 2006; Martin et al., 2008). Functional relationships between aerosol emissions and CO_2 emissions, which both originates from a common source, suggest the possibility of using carbon dioxide (CO_2) flux as a traffic tracer. The amount of CO_2 produced from vehicle combustion and fossil fuel is well known and varies much less than the aerosol (Vogt et al., 2010, Bayerisches Landesamt fuer Umwelt). The later change not only in concentration but also in size and internal external mixing. Measurement results shown below will be used to calculate size resolved EFs in the size range of (Dp, 0.25–2.5 µm).

2 Measurement site and instrumentation

The measurements were made in Stockholm (Sweden), from the top of a telecommunication tower in the southern central part of the city. The tower is built in concrete, 105 m tall and located 28 m above the sea level. (Latitude North 59°1′0.43″ and Longitude: East 18°5′53.17″). On the top of the tower there is an elevator machine room and on top of that there is a 11 m high metal frame with a 2.5×2.5 m platform at the top. This platform enable us to extend the flux measurements far enough from the bulkier concrete construction to avoid flow distortion caused by the tower. Central Stockholm, with high traffic activity, is located north of the tower. A wide forest area dominates in the easterly direction. Significant green sectors can also be found to the east through to the south-west mixed with residential areas.

Because the focus of this study is road traffic aerosol emissions, more details are provided on the larger streets lying in the footprint area of interest. The communication tower is located just south of Hammarby Fabriksväg, a local road with around 9700 vehicles per day, which merges into Södra Länken , one of the most heavily trafficked

ACPD
10, 21521–21545, 2010

roads in the neighborhood of the tower, with around 50 000 vehicles per day. Södra Länken, is an underground freeway tunnel with one exit located in the Northeast of communication the tower (see Fig. 1). The site has been previously described by Mårtensson et al. (2006) and Vogt et al. (2010)

2.1 Instruments and measurement setup

The instrumentation consists of a Gill (R3) ultrasonic anemometer, an open path infrared CO_2/H_2O analyzer LI-COR 7500 (LI-COR, Inc., Lincoln, Nebraska 68504, USA), and two identical Optical Particle Counters (OPC) (Model, 1.109, Grimm Ainring, Bayern, Germany) in a housing with a system to heat and dry the sampled air (Grimm Model 265, special version going up to 300 °C). The sample air was dried by 1:1 dilution with 0% humidity particle free air, which minimizes the risk of unwanted loss of semi-volatile compounds, compared to simply heating the air in order to dry it. (Detailed information can be found in Vogt et al., 2010)

2.2 Eddy covariance method, data processing corrections and errors

The vertical aerosol number flux was calculated using the eddy covariance technique (EC). For this study the flux $\overline{w'N}$ was calculated over periods of 30 min. The fluctuations w' and N' were separated from the mean by linear de-trending, which also removes the influence of low frequency trends.

The validity of the EC technique at the measurement location was confirmed in earlier studies (Mårtensson et al., 2006; Vogt et al., 2010). The fluxes have been corrected due to the limited time response of the sensor and attenuation of turbulent fluctuations in the sampling line. The response time constant τ_c for both OPC and sampling line was estimated to be 1.5 s by using transfer equations for damping of particle fluctuations in laminar flow (Lenschow and Raupach, 1991) and in a sensor (Horst et al., 1997). The typical magnitude of these corrections varied, resulting in an underestimation of between 12 to 32%, depending on wind speed and stability conditions.

ACPD
10, 21521–21545, 2010

CO_2 has been corrected for variations in air density due to fluctuation in water vapor and heat fluxes in accordance with Webb et al. (1980). This resulted in a maximum increase around noon for CO_2 of ~37%.

In addition the aerosol fluxes and concentrations were corrected for tube losses in the sampling line, which resulted in particle losses of ~5% for the largest size class in the OPC (Dp = 2 µm to 2.5 µm).

3 Results

The measurements in this study were performed from 1st of April 2008 to the 15th of April 2009. About 45% of the data has been removed due to instrumental problems, mostly due to rain. An open path infrared CO_2/H_2O analyzer was used which resulted in large spikes in the data set associated with rain events. In addition to the spike removal, half hourly data were rejected when the atmosphere was not turbulent (u_* <0.1 m s^{-1}) and from 12.12.2008 to 21.1.2009 no LICOR data were available.

3.1 Wind direction and sector selection

The wind direction dependency of the particle number concentration and flux and the CO_2 flux and concentration is shown in Fig. 2. The data has been sorted into mean values in 10° bins for the incoming wind direction. The CO_2 and particle fluxes show similar wind direction dependencies. The highest values for the CO_2 and particle flux are found to the Northeast (40 to 80°). This maximum coincides with the densest traffic within the footprint area. A minimum in the fluxes is found in the East to South (90 to 180°). The CO_2 flux shows negative values within this sector indicating that the photosynthetic activity from the urban forest located in the East dominates surface carbon exchange in this area. The particle fluxes also show negative values (120 to 200°) indicating that deposition of particles is the most important particle surface exchange process in this wind sector. Particle fluxes as well as CO_2 fluxes increase from SW to N (200 to 360°).

ACPD
10, 21521–21545, 2010

Unlike the flux results, the maximum in particle concentration was found in the East to South (90 to 200°), due to long range transport. The CO_2 concentrations show maximum values for northerly winds (270 to 90°).

The wind direction dependence of concentration suggests that for particles in the size range of 0.25–2.5 µm, air masses coming from the eastern and southern part of Europe are dominating most of the particle concentration above Stockholm, which has typically been seen earlier (Areskoug et al., 2000; Tunved et al., 2005). The local emissions from Stockholm of particles in this size range have much less influence on the mean particle concentrations than long-range transport. This phenomenon is true except for dry spring days, where there highest number concentrations can be found. On the other hand, the CO_2 concentrations are higher in the Northern sector indicating that Stockholm is a major net source of CO_2.

3.2 Diurnal cycles

Figure 3 shows a comparison of the diurnal cycles of aerosol particle flux (Dp, size range 0.25–2.5 µm) and CO_2 fluxes for northerly winds (270 to 90°). The aerosol fluxes are low in magnitude during nighttime and high during daytime. The CO_2 fluxes show the same diurnal pattern with minimum values at night and highest during daytime with the maximum around midday being related both to increased human activity and more turbulent conditions. The median particle and CO_2 fluxes are always positive, which indicates that the city is mostly a net source for these parameters.

Aerosol and CO_2 fluxes start to increase rapidly between 5 to 8 a.m. which correlates well with the morning traffic rush hour. Both fluxes stay high during daytime. The decline in the evening around 6 p.m. is caused by a drop in traffic activity, which is main source for both CO_2 and aerosol within the footprint area. These observations are consistent with earlier studies in cities (Nemitz et al., 2002; Valesco et al., 2005; Coutts et al., 2007; Vogt et al., 2005; Järvi et al., 2009). The diurnal and seasonal cycles of this data have been previously described in detail by Vogt et al. (2010).

ACPD
10, 21521–21545, 2010

3.3 Emission factors

Particle EFs can be derived with different units depending on the available data and intended use. For example certain distance driven by a vehicle [g/km], or the amount of fuel burned [g/l], can be the fundament to quantify emissions of a certain substance. In our case CO_2 is used as a tracer for road traffic combustion.

A linear correlation between particle number flux and CO_2 flux was used to determine an emission factor (Dp, size range 0.25–2.5 µm) in units of particles/mmol CO_2. Figure 4 shows the linear fit to the data. The data has been divided into 15 concentration intervals. The interval width was chosen so that in each interval at least 20 or more half hour values were present. The linear fit was made to the median value of each size bin. The slope of this fit has the units of [particles/mmol CO_2], which can be considered an emission factor. By converting mol into mass, EFs with units of [particle/g CO_2] can also be calculated. Note that the linear fit has been made only to the part of the data set with positive CO_2 fluxes, as combustion does not consume CO_2.

The combined linear fits to each of the 15 size channels of the OPC (Dp, size range 0.25–2.5 µm) gives size resolved EFs. By assuming a particle density of 1600 kg/m^3 (Pitz et al., 2003) and using the particle sizes from the OPC, mass related emission factors [μg/g CO_2] may also be calculated (see Fig. 5). Even though Pitz et al. (2003) investigated that aerosol density showed pronounced diurnal pattern both in summer and in winter and also on weekdays and weekends, this hasn't be taken into account to keep the approach simple. Details of the EFs in mass and number can be found in Table 2. The number based emission factors have their highest values for the smallest aerosol sizes, while the mass based emission factors have the largest values in the super-micrometer range. Mass emissions below 0.75 µm Dp are close to constant with size, but show an exponential increase above this size.

ACPD
10, 21521–21545, 2010

3.4 Comparable measurements and annual variation in emission factors

Comparable measurements have been made at a densely trafficked site (Hornsgatan with around 30 000 vehicles per day) in the city centre, in the northwest direction from of the tower. The measurements at Hornsgatan (a street canyon site previously described in detail by e.g. Gidhagen et al., 2005) were made using an OPC (Grimm Technologies, Model 1.109), i.e. the same measurement technique and instrument as for the particle fluxes on the tower. The EFs obtained at Hornsgatan were calculated by using the NO_x scaling method. Thereby NO_x is used as a tracer. Detailed description can be found in Omstedt et al. (2005).

Figure 6 shows the annual variation in the EFs [mg veh^{-1} km^{-1}] derived from the tower and the Hornsgatan measurements. To convert the EFs to the units of [veh^{-1} km^{-1}] it was assumed that 90% of the cars in Stockholm run on gasoline fuel and 10% on diesel fuel and 95% of the cars are light duty traffic and 5% heavy duty. In addition we assumed that light duty cars consume 0.1 l km^{-1} of gasoline, 0.07 l km^{-1} of diesel and heavy duty vehicles 0.3 l km^{-1} of diesel (SCM, Bayerisches Landesamt fuer Umwelt).

The EFs calculated from the communication tower correlate very well with those from Hornsgatan. Emission rates are generally higher in spring and early summer, than the rest of the year. The estimated emission factors and the variability of those for the tower-site and street-site overlap in most of the months except for July. Reasons for that might be less traffic, which might lead to less break wear production. In addition the amount of heavy duty traffic drops a lot during this period. The mean annual emission factor for Hornsgatan is 29.7 [mg veh^{-1} km^{-1}] and 27.5 [mg veh^{-1} km^{-1}] for the tower measurements.

ACPD
10, 21521–21545, 2010

3.5 Relevant source processes and their influence on the emission factor

Gillette et al. (1982) showed the importance of turbulence for the suspension of course mode particles in deserts. As this is a process one could suspect it would apply on dust on roads as well, we attempted to investigate the effect of different turbulent conditions on number EFs (Dp, size range 1–2.5 µm). Turbulence is produced either from heat driving convection or wind shear near the ground shown in the friction velocity. Number EFs for seven different turbulence scenarios were calculated (friction velocity at the tower from 0.1 to 1.1 m s^{-1}, see Fig. 7b). Figure 7b shows that the number emission factor is not significantly affected by turbulence for friction velocity conditions below 0.8 m s^{-1}. Turbulent conditions over 0.8 m s^{-1} show a large increase in the emission factor as the suspension of particles larger than 1 µm starts to become more effective. The same analysis can be applied for the horizontal wind speed. Nemetz et al. (2001) found a dependence of the flux of coarse particles with horizontal wind speed similar to that shown in (7a). This indicates that coarse particles on roads or other ground surfaces may be suspended at high wind speeds. This is a well know phenomena and has been shown for deserts (Fratini et al., 2007). Particles fluxes with a diameter smaller than 1.0 µm do not show this clear wind speed dependence (Nemetz et al., 2001). Because the mass EFs are dominated by particles in the size range (Dp, 1.0–2.5 µm), it is anticipated that high winds will affect the particle mass flux. To test this hypothesis, the number EFs size range (Dp, 1.0–2.5 µm) were binned based on wind speeds between 1 and 13 m s^{-1}. We divided the wind speed into 11 bins to ensure that in each bin at least 20 values were present in each bin (see Fig. 7a).

From Fig. 7a, it appears that high wind speeds have a huge impact on the mass emission factor but the frequency of 30 min periods with wind speeds greater than 8 m s^{-1} appearing in our data set is around 5%, which means that these high wind speed events do not strongly impact the monthly and annual estimated mass emission factor. For locations where were high wind speeds are more frequent, the high EFs related to high wind speed should be taken into account. But also the high EFs related

ACPD
10, 21521–21545, 2010

to high wind speed may influence extreme values and the amount of days that exceed critical levels especially if wind conditions coincide with high traffic counts. This effect should therefore be taken into account in air quality models. In conclusion, wind speed and friction velocity make a contribution on the aerosol flux and this is likely related to the fact that at higher wind speeds (or high u^*) particles sizes larger than 1.0 µm may become suspended. Since these high wind speeds were not common over the measurement period, vehicle induced turbulence and suspension of particles was likely to be the dominant process for large particle emissions.

3.6 Comparison of different methods to estimate emission factors

Table 2 gives an overview of $PM_{2.5}$ EFs in [g veh^{-1} km^{-}1]. Emission factors range from 0.01 to 0.3 mg/vkm. Highest value is reported by Keogh. Differences between studies are to different contribution from exhaust and non-exhaust PM. Exhaust maybe due to different fleet mix HDV/LDV and different fuel mix (diesel/gasoline). Evaluating emission trends by comparing data with (Ketzel et al., 2007) is not very significant, as the contribution of exhaust emission, which may have decreased due to better catalyst in cars from 2004 to 2008 is relatively small in Stockholm (Dp<0.6 µm). The major contribution to the total EFs of $PM_{2.5}$ here is mechanically produced particle matter (Dp>0.6 µm). Norman and Johansson discuss the fact that meteorology, and in particular road wetness is the main parameter which controls $PM_{2.5}$ emissions (Norman et al., 2006). That being the case, the higher annual emission factor observed by Ketzel in 2004 is likely due to a dry spring period.

Notwithstanding this point, the emission factor for $PM_{2.5}$ determined in this study is within a factor of 2 to 3 of previous studies (Kristensson et al., 2004; Keogh et al., 2009 and Ketzel et al., 2004).

ACPD
10, 21521–21545, 2010

4 Summary and conclusion

Size-resolved vertical aerosol number fluxes of particles with Dp, = 0.25–2.5 µm were measured with the eddy covariance method from a 105 m high communication tower over the city Stockholm, Sweden. In this study, size resolved number and mass EFs have been calculated and compared with other published results. In addition meteorological and special factors that may influence the EFs have been discussed.

The key findings are

1. The highest values for the CO_2 and particle flux are found to the Northeast.
2. Unlike the flux results, the maximum in particle concentration was found in the East to South (90 to 200°), due to long range transport.
3. Particle and CO_2 fluxes show the same diurnal pattern with lows at night and highs during daytime with the maximum around midday being related both to increased human activity and more turbulent conditions.
4. Emission factors were determined by a linear correlation between particle number flux and CO_2 flux.
5. Annual emission mass emission factor obtained with the eddy covariance method at the communication tower is slightly lower than that determined with the NO_x method in the street canyon (28, 30 [mg veh^{-1} km^{-1}]).
6. Emission rates are generally higher in spring than in summer.
7. Turbulent conditions over 0.8 m s^{-1} show a large increase in the emission factor as the suspension of particles larger than 1 µm starts to become more effective.
8. Extreme values can be found if horizontal wind speeds are large (>8 m s^{-1}).

ACPD
10, 21521–21545, 2010

In conclusion CO_2 fluxes in combination with particle fluxes can be used to derive EFs. The EFs in the size range of Dp, = 0.25–2.5 µm are affected by high turbulence and high wind speed conditions due to resuspension of particles.

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ACPD

10, 21521–21545, 2010

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ACPD
10, 21521–21545, 2010

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ACPD

10, 21521–21545, 2010

The relationship between 0.25–2.5 µm aerosol and CO_2 emissions

M. Vogt et al.

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ACPD

10, 21521–21545, 2010

Table 1. Median mass and number emission factor for each of the 15 OPC size bins. Values in brackets represent the 95% confidence interval.

Size [µm]	Mass [mg veh^{-1} km^{-1}]	Number 10^{10} [particle veh^{-1} km^{-1}]
0.2600	0.70 (0.80;0.52)	5.73 (6.51;4.24)
0.2900	0.48 (0.65;0.36)	2.80 (3.77;2.07)
0.3250	0.42 (0.57;0.33)	2.00 (2.68;1.54)
0.3750	0.43 (0.81;0.30)	1.27 (2.40;0.89)
0.4250	0.39 (0.76;0.25)	0.78 (1.52;0.50)
0.4750	0.20 (0.30;0.12)	0.28 (0.42;0.17)
0.5400	0.38 (0.73;0.21)	0.38 (0.74;0.21)
0.6200	0.54 (0.87;0.28)	0.25 (0.41;0.13)
0.6800	0.33 (0.43;0.21)	0.12 (0.16;0.08)
0.7500	0.69 (0.9;0.40)	0.17 (0.22;0.10)
0.9000	0.90 (1.40;0.67)	0.12 (0.18;0.08)
1.1500	2.03 (2.41;1.25)	0.12 (0.14;0.07)
1.4500	1.40 (1.64;1.28)	0.04 (0.05;0.04)
1.8000	3.31 (3.99;2.72)	0.05 (0.06;0.04)
2.2500	7.80 (10.77;5.28)	0.06 (0.09;0.04)

ACPD
10, 21521–21545, 2010

Table 2. Published emission factors for particulate mass fractions on road studies.

Authors	Vehicle type	Particle Mass Fraction	Emission factor [g/veh km^{-1}]	Uncertainty	Type of road
Kirchstetter et al. (1999)	HD vehicles	$PM_{2.5}$	0.99	Sd 0.08	Road tunnel
	LD vehicles	$PM_{2.5}$	0.0098	Sd 0.0009	
Kristensson et al. (2004)	All vehicles	$PM_{2.5}$	0.067	0.005	Road tunnel
Harrison et al. (2006)	HD vehicles	$PM_{2.5}$	0.179	Sd 0.022	City street
	LD vehicles	$PM_{2.5}$	0.01	Sd 0.004	
Grieshop et al. (2006)	All vehicles	$PM_{2.5}$	0.022 (213)[a]	Sd 31	
Cheng et al. (2009)	All vehicles	$PM_{2.5}$	0.131	Sd 0.0369	
Ketzel et al. (2007)	All vehicles	$PM_{2.5}$	0.054[b]	Not available	
	All vehicles	$PM_{2.5}$	0.067[c]	Not available	City street
	All vehicles	$PM_{2.5}$	0.029[d]	Not available	City street
	All vehicles	$PM_{2.5}$	0.033[e]	Not available	City street
Keogh et al. (2009)	HD vehicles	$PM_{2.5}$	0.302		
	LD vehicles	$PM_{2.5}$	0.033		
This study	All vehicles	$PM_{2.5}$	0.030[f]	(0.018,0.48)	Street Canyon
	All vehicles	$PM_{2.5}$	0.028[g]	(0.013,0.45)	Tower(EC)

[a] Emission factor in unit [mg (kg fuel^{-1})] (original value).
[b] Measurements made at H.C Andersens Blvd in in Copenhagen, Denmark (2003–2004).
[c] Measurements made at Hornsgatan in Stockholm, Sweden (2002–2004).
[d] Measurements made at Merseburger Strasse, Halle, Germany (2003–2004).
[e] Measurements made at Runbergkatu, Helsinki, Finnland (2003–2004).
[f] Measurements made at Hornsgatan in Stockholm, Sweden (2008–2009).
[g] Measurements made at Communication Tower Stockholm, Sweden (2008–2009).

ACPD
10, 21521–21545, 2010

Fig. 1. Shows the location of the tower in Stockholm and its surroundings. Blue = open water surfaces, green = forest/park areas, brown = built-up areas (mainly residential areas), orange = Public buildings (schools, sport arenas etc), white = roads.

ACPD
10, 21521–21545, 2010

The relationship between 0.25–2.5 µm aerosol and CO_2 emissions

M. Vogt et al.

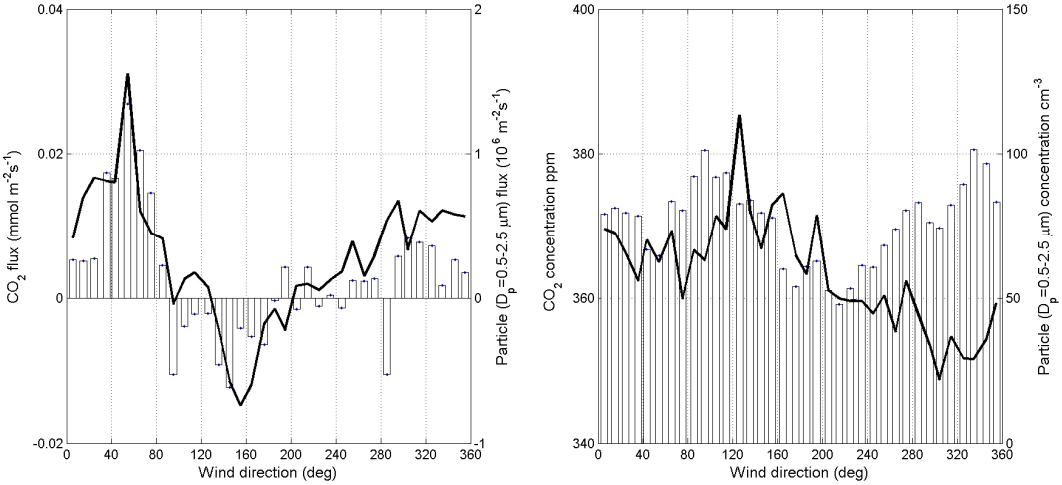

Fig. 2. **(a)** Aerosol number and CO_2 flux and **(b)** particle and CO_2 concentrations in constant wind sector intervals. Bars represent CO_2 flux and concentrations. Solid line: aerosol number flux and particle concentration.

ACPD
10, 21521–21545, 2010

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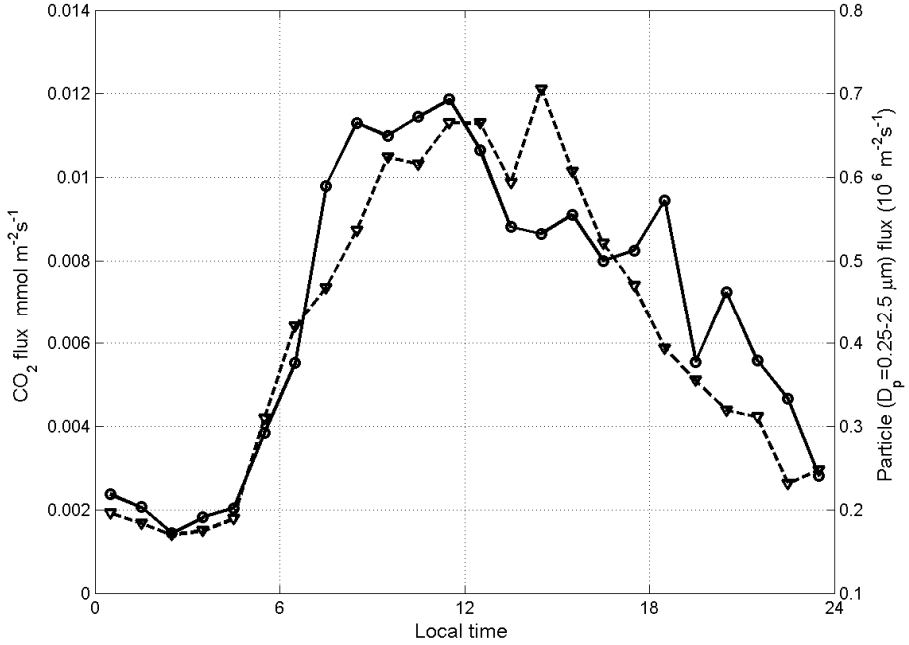

Fig. 3. Median diurnal cycles of aerosol number flux within the total OPC size range (solid line) and CO_2 flux (dashed line) for the North sector.

ACPD
10, 21521–21545, 2010

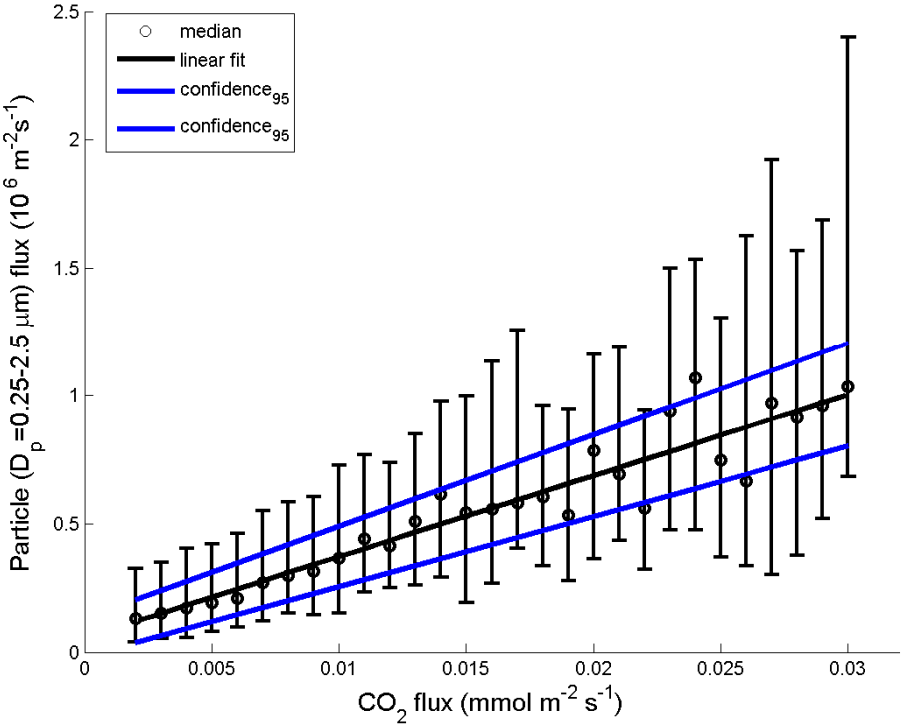

Fig. 4. Median values for particles particle flux with D_p = 0.25–2.5 µm within constant intervals of CO_2 flux. The vertical bars represent 25 and 75 percentiles. The blue line represents the linear fit the 95 confidence interval for the linear fit.

ACPD
10, 21521–21545, 2010

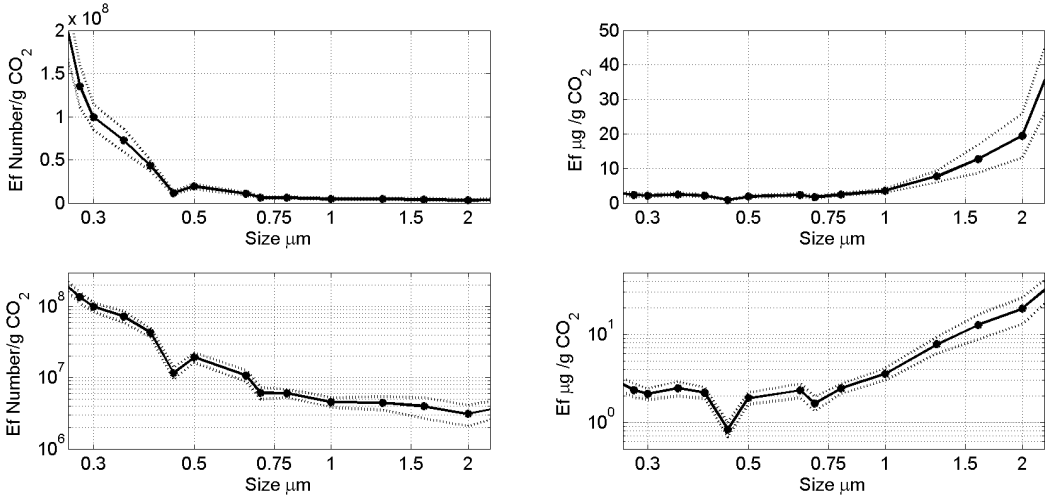

Fig. 5. Median size resolved **(a)** number and **(b)** mass emission factor and **(c)** in logarithmic y-axis for number **(d)** in logarithmic y-axis for mass. The dashed line represents the variability using the 95 confidence interval of the linear fit similar to the one shown in Fig. 4.

ACPD
10, 21521–21545, 2010

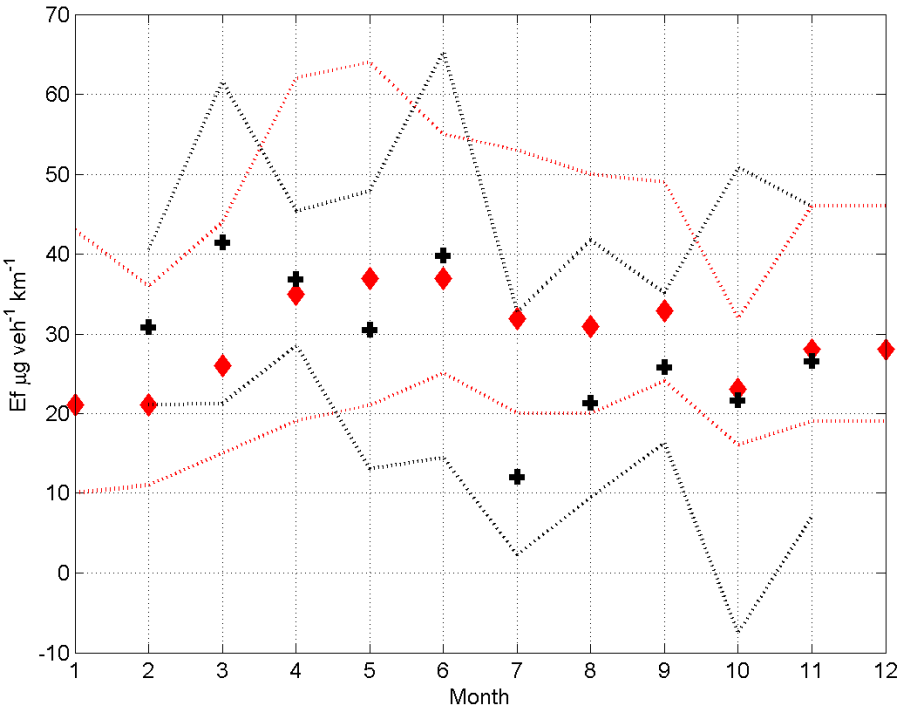

Fig. 6. Monthly median emission factors (red, triangle) street canyon, using the NO_x method and (black, plus) tower using the eddy covariance method. The red dashed lines are the 25, 75 percentiles. The black dashed lines are the variability using the 95 confidence interval of the linear fit as shown in Fig. 4.

ACPD
10, 21521–21545, 2010

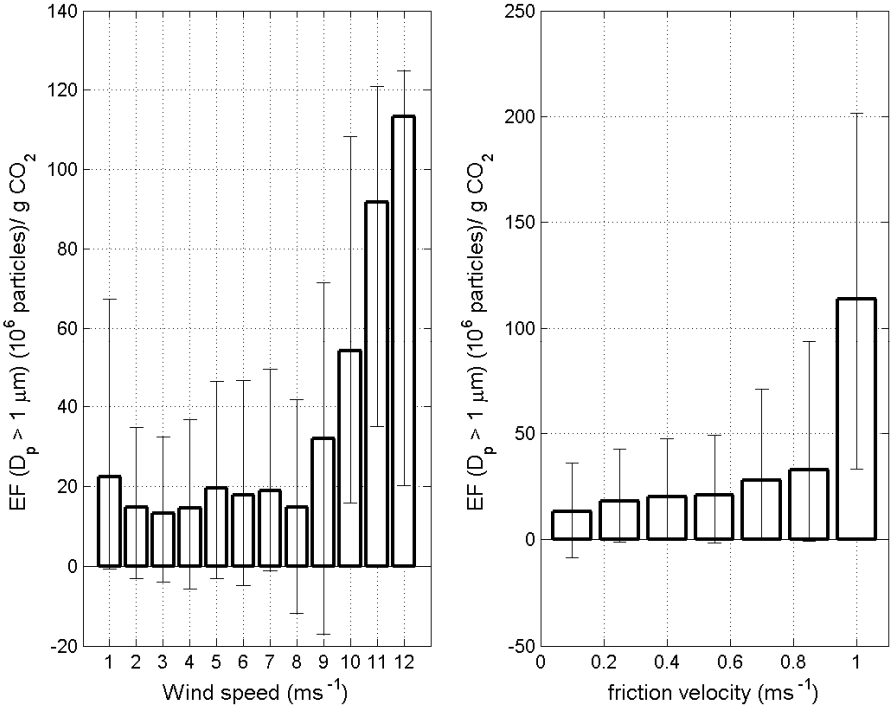

Fig. 7. Median number emission factor for coarse particles within **(a)** constant wind speed intervals and **(b)** constant friction velocity intervals. Vertical bars represent the 25,75 percentiles.

ACPD
10, 21521–21545, 2010

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M. Vogt et al.