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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 in addition, 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 to derive aerosol emission factors based on carbon dioxide (CO_2) emission fluxes. Aerosol fluxes were measured over one year using the eddy covariance method at the top of a 105 m high communication tower in Stockholm, Sweden. Maximum CO_2 and particle fluxes were found when the wind direction coincided with the area of densest traffic within the footprint area. Negative fluxes (uptake of CO_2 and deposition of particles) coincided with periods of sampling from 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 be directly related to the amount of fuel burnt by vehicles in the footprint area. The estimated emission factor for the fleet mix in the measurement area was, in number 1.8×10^{11} particle $\mathrm{veh}^{-1}\,\mathrm{km}^{-1}$ (for 0.25–2.5 µm size range). Assuming spherical particles of density $1600\,\mathrm{kg\,m}^{-3}$ this corresponds to $27.5\,\mathrm{mg\,veh}^{-1}\,\mathrm{km}^{-1}$. For particles (0.8–2.5 µm) the emission factors were $5.1 \times 10^{9}\,\mathrm{veh}^{-1}\,\mathrm{km}^{-1}$ for number and $11.5\,\mathrm{mg\,veh}^{-1}\,\mathrm{km}^{-1}$ for mass. But a wind speed dependence was noted for high wind speeds. Thus, for wind speeds larger than $9\,\mathrm{m\,s}^{-1}$, as measured in the tower at 105 m (U_{105}), the emission factor for particle number and mass was parameterised as: $E_\mathrm{f}(\mathrm{Number},\ 0.8-2.5\ \mu\mathrm{m}) = (6.1\pm1.7)10^{9}U_{105} - 50 \pm 188$ and $E_\mathrm{f}(\mathrm{Mass},\ 0.8-2.5\ \mu\mathrm{m}) = (20\pm12)U_{105} - 171 \pm 122$.

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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 (Harrison et al., 2000), but PM_{10} and $PM_{2.5}$ are the usual metrics used in regulations of air pollution within Europe. This is despite the fact that information about bulk particle mass concentrations or emissions of PM_{10} or $PM_{2.5}$ are of limited value for assessing climate and health effects of aerosol pollution. Particle size resolved information on emissions is urgently required to better understand the processes controlling emissions and their importance for health and climate. In order to make accurate air quality models, traffic emissions and 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 approaches have been used to quantify road traffic.

These include:

– *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).

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– *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 and Buchmann, 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., 2009; 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 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 combustion of vehicle fuels is well known and varies much less than the production of particles (Vogt et al., 2011).The latter varies not only in total amount produced but also in size and is not only dependent on fuel combustion but also on mechanical processes such as road, brake and tyrewear. In this paper we present size resolved fluxes of particles in the size range of 0.25–2.5 µm and by using simultaneously measured CO_2 fluxes we estimate emission factors that are representative of the net emission from the city of Stockholm.

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 was built in concrete, 105 m tall and located 28 m above the sea level. (Latitude North 59°18′0.43″, Longitude: East 18°5′53.17″). On the top of the tower is an elevator machine room and on top of that there is an 11 m high metal frame with a 2.5×2.5 m platform at the top. This platform enables 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 within 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 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. (2011).

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 semivolatile compounds, compared to simply heating the air in order to dry it (detailed information can be found in Vogt et al., 2011).

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., 2011). The fluxes have been corrected for 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 1.0 s 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 prior to the correction of between 12 to 32 %, depending on wind speed and stability conditions. The median relative counting error in the flux was 15 % at the smallest and largest OPC sizes and peaked at $D_p = 0.7$ µm) with 35 %, see Vogt et al. (2011).

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

CO_2 has been corrected for variations in air density due to fluctuations 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 ($D_p = 2$ µm to 2.5 µm).

3 Results

The measurements in this study were performed from 1 April 2008 to the 15 April 2009. Approximately 45 % of the data has been removed due to instrument problems, primarily 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 ($u_* < 0.1\,\mathrm{m\,s^{-1}}$) and from 12 December 2008 to 21 January 2009 when 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 have been averaged in 10 degree bins with respect to 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 degrees). 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 degrees). 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 degrees) indicating that deposition of particles is the dominant particle surface exchange process in this wind sector. Particle fluxes as well as CO_2 fluxes increase from SW to N (200 to 360 degrees).

Unlike the fluxes, the maximum in particle concentration was found in the East to South (90 to 200 degree). The CO_2 concentrations show maximum values for northerly winds (270 to 90 degree). The wind direction dependence of the particle concentrations is consistent with earlier studies that have shown importance of long range transport from the eastern and southern part of Europe (Areskoug et al., 2000; Tunved et al., 2005). The local emissions of particles in the size range 0.25–2.5 µm from Stockholm itself have much less influence on the mean particle concentrations than long-range transport. For CO_2, the concentrations are higher in the NW-NE sectors in consistence with the net positive fluxes in these sectors.

3.2 Diurnal cycles

Figure 3 shows a comparison of the diurnal cycles of aerosol particle flux (D_p, size range 0.25–2.5 µm) and CO_2 fluxes for northerly winds (270 to 90 degrees). 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 shows that the city is mostly a net source for these parameters.

Aerosol and CO_2 fluxes start to increase rapidly between 05:00 to 08:00 a.m. which correlates well with the morning rush hour traffic. Both fluxes stay high during daytime. The decline in the evening around 06:00 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., 2006; Järvi et al., 2009). The diurnal and seasonal cycles of this data have been previously described in detail by Vogt et al. (2011).

3.3 Emission factors

Particle EFs can be derived with different units depending on the available data and intended use. For example the amount per distance driven by a vehicle [$\mathrm{g\,km^{-1}}$], or the amount of emitted pollutant per amount fuel burned [$\mathrm{g\,l^{-1}}$], can be used to quantify emissions of a certain substance. In our case CO_2

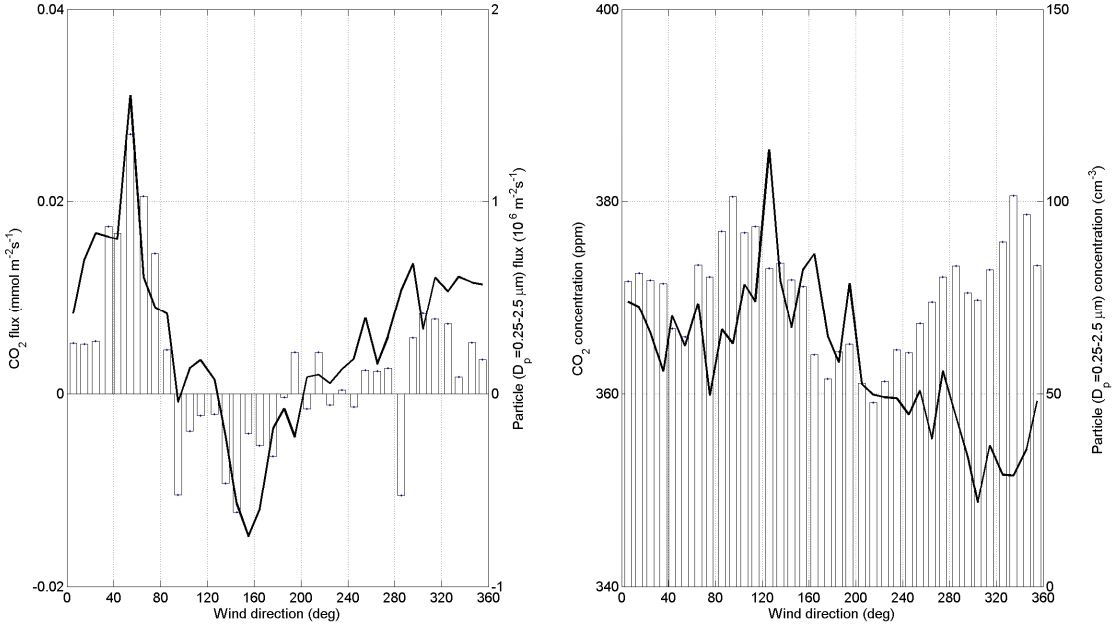

Fig. 2. **(a)** Aerosol number and CO_2 flux and **(b)** particle and CO_2 concentrations in wind sector intervals. Bars represent CO_2 flux and concentrations. Solid line: aerosol number flux and particle concentration.

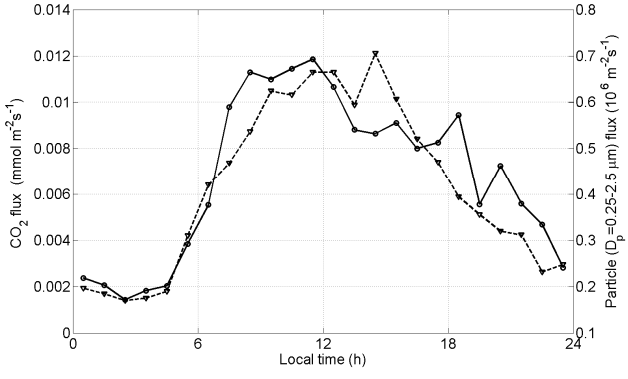

Fig. 3. Median diurnal cycles of aerosol number flux within the total OPC size range (solid line) and CO_2 flux (dashed line) for the North sector.

is used as a tracer of combustion by road traffic. A linear correlation between particle number flux and CO_2 flux was used to determine an emission factor (D_p, size range 0.25–2.5 µm) in units of particles mmol^{-1} CO_2. Hence here we only include the North eastern sector (0–90 degrees), the most traffic intense sector, to minimize the effect of other sources of both CO_2 and particles, for example house warming, industry cooking etc. In the emission database for the footprint area the relative contribution for different sources is listed which are the following: for CO_2 emissions within the footprint area energy contributes 8 %, road traffic = 80 %, sea traffic = 9 %, industry = 1 %, other = 2 % (Johansson and Eneroth, 2007).

Figure 4 shows the linear fit to the CO_2 and aerosol fluxes. The data has been divided into 15 intervals with different flux magnitude. The interval width was chosen such 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^{-1} CO_2]. 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 (D_p, 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 may also be calculated. Possible variations of aerosol densities, as shown by Pitz et al. (2003), were not considered due to lack of information. EFs for particle mass and number for each size bin is shown in Table 1. The number based emission factors have their highest values for the smallest aerosol sizes, while the mass based emission factors are highest for the super-micrometer particle sizes. Number emission factors range from 0.082 to 8.16×10^{10} particles veh^{-1} km^{-1}, i.e. by almost 2 orders of magnitude. Mass emission factors range from 0.23 to 7.36 mg veh^{-1} km^{-1}, a factor of around 30.

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 bin (µm)	Mass [mg veh^{-1} km^{-1}]	Number 10^{10} [particle veh^{-1} km^{-1}]
0.25–0.28	1.12 (0.68; 1.59)	8.16 (4.97; 11.57)
0.28–0.30	0.83 (0.51; 1.29)	4.34 (2.70; 6.73)
0.30–0.35	0.79 (0.49; 1.32)	2.94 (1.83; 4.92)
0.35–0.40	0.75 (0.50; 1.43)	1.82 (1.23; 3.46)
0.40–0.45	0.58 (0.42; 1.06)	0.96 (0.70; 1.76)
0.45–0.50	0.23 (0.15; 0.40)	0.27 (0.18; 0.47)
0.50–0.58	0.58 (0.38; 0.85)	0.47 (0.31; 0.69)
0.58–0.65	0.46 (0.32; 0.72)	0.24 (0.17; 0.41)
0.65–0.70	0.32 (0.27; 0.47)	0.13 (0.11; 0.19)
0.70–0.80	0.58 (0.31; 0.93)	0.17 (0.10; 0.28)
0.8–1.0	0.76 (0.56; 1.15)	0.13 (0.09; 0.20)
1.0–1.3	1.17 (0.86; 2.27)	0.098 (0.07; 0.19)
1.3–1.6	1.57 (1.45; 1.86)	0.066 (0.06; 0.07)
1.6–2.0	3.10 (2.29; 4.12)	0.067 (0.05; 0.09)
2.0–2.5	7.36 (6.94; 8.80)	0.082 (0.07; 0.09)

3.4 Annual variation in emission factors and comparison with street canyon data

TEOM-$PM_{2.5}$ measurements and emission factors have also been calculated for a densely trafficked site (Hornsgatan with around 30 000 vehicles per day) in Stockholm's 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). The EFs ($PM_{2.5}$) obtained at Hornsgatan were calculated by using the NO_x scaling method which assumes that the dispersion of the emitted particles is similar to NO_x dispersion. This is justified by the fact that the timescale for deposition of particles is several hours, which is much longer than the timescale for mixing and dilution making it reasonable to assume that differences in deposition of NO_x and particles have a minor influence. This method has been successfully used in several earlier studies (Ketzel et al., 2003; Gidhagen et al., 2004, 2005; Omstedt et al., 2005).

Figure 5 shows the annual variation in the EFs ($PM_{2.5}$) [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 ran on gasoline fuel and 10 % on diesel fuel and 95 % of the cars were 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 (Bayerisches Landesamt fuer Umwelt; http://www.lfu.bayern.de/index.htm).

The EFs (D_p, size range 0.25–2.5 µm) calculated from the communication tower correlate very well with those from

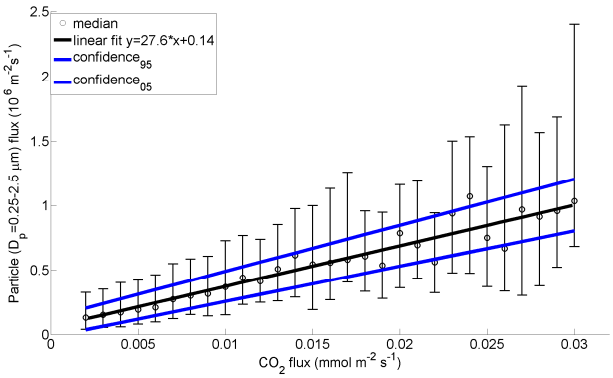

Fig. 4. Median values for particles particle flux with $D_p = 0.25$–2.5 µm within constant intervals of CO_2 flux. The vertical bars represent 25 and 75 percentiles. The blue line represents the 95 confidence interval of the linear regression line.

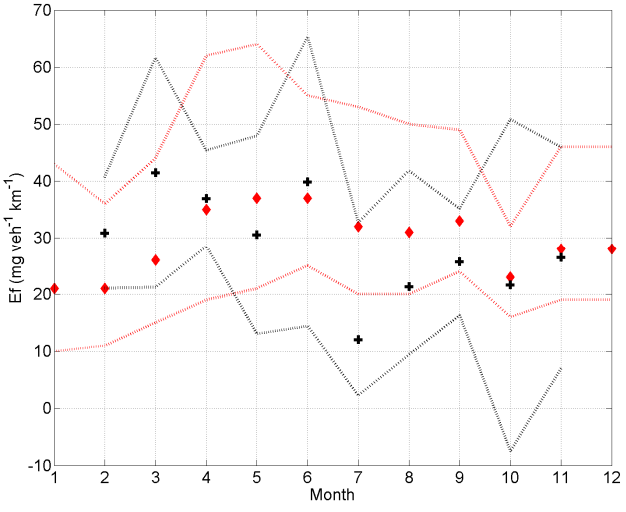

Fig. 5. Monthly median emission factors (red, triangle) street canyon, using the NO_x method and (black, plus) tower using the eddy covariance method. The red dashed lines are the 25, 75 percentiles. The black dashed lines are the variability using the 95 confidence interval of the linear fit as shown in Fig. 4.

Hornsgatan. Emission rates are generally higher in spring and early summer than the rest of the year. We find reasonable overlap between the estimated emission factors and their associated variability from the tower-site and street-site in most months, except July. A reason for this disagreement may be less traffic volumes in July, which would lead to less brake wear. In addition the amount of heavy duty traffic drops significantly during this period due to less professional traffic during the main vacation month. The mean annual emission factor ($PM_{2.5}$) for Hornsgatan is 29.7 [mg veh^{-1} km^{-1}] and 27.5 [mg veh^{-1} km^{-1}] for the tower measurements.

3.5 Relevant source processes and their influence on the emission factor

Wind speed could be important for suspension of dust on roads. In Fig. 6 the number EFs for the size range (D_p, 0.8–2.5 µm) have been binned based on wind speeds between 1 and 12 m s^{-1} with 11 bins to ensure that in each bin at least 20 values were present in each bin. Figure 6 shows the effect of different wind speeds on the number EFs (D_p, size range 0.8–2.5 µm). For wind speed larger than 9 m s^{-1}, the EF increase as wind speed increase. Below 9 m s^{-1} there is no influence of wind speed. Nemitz et al. (2001) found a similar dependence of the flux of super micron particles with horizontal wind speed. This indicates that super micron 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; Gilette et al., 1980). Nemitz et al. (2001) found no clear wind speed dependence for sub-micron particle fluxes. This can be seen in our data as well. The EF (D_p, size range 0.25–0.8 µm) were constant for all wind speeds wit 1.74×10^{11} veh^{-1} km^{-1}] in number and 6.5 [mg veh^{-1} km^{-1}] in mass.

Even though high wind speeds have a significant impact on the emission factor, only 5 % of the dataset have wind speeds greater than 9 m s^{-1}. This means that these high wind speed events do not strongly impact the monthly and annual estimated emission factor. By excluding wind speeds over 9 m s^{-1} the annual emission factor was reduced to 26.9 [mg veh^{-1} km^{-1}]. For locations where high wind speeds are more frequent, the high EFs related to high wind speed should be taken into account. In addition, the high EFs related 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 volumes. This effect should therefore be taken into account in air quality models. We attempted to parameterize the EF (D_p, size range 0.8–2.5 µm) using wind speed as parameter. Equations (1) and (2) show the influence of wind speed larger than 9 m s^{-1} on the EFs:

$$E_{\mathrm{f}}(\mathrm{Number},\ 0.8-2.5\ \mu\mathrm{m}) = (6.1\pm1.7)10^{9}U_{105} - 50 \pm 188 \quad (1)$$

$$E_{\mathrm{f}}(\mathrm{Mass},\ 0.8-2.5\ \mu\mathrm{m}) = (20\pm12)U_{105} - 171 \pm 122. \quad (2)$$

where U is the horizontal wind speed at tower level. Modelers may prefer a standard reference height such as 10 m, but it is not straightforward to define such a height in a complex urban terrain. While the surrounding buildings have an average height of 12 m, it varies largely. If using forest canopies as an analog system, one could perhaps define a displacement height as a function of the building height and define a reference wind speed height from that, but the urban canopy is less well described than forests.

Since these high wind speeds were not common over the measurement period, vehicle induced turbulence and suspension of particles is likely to be the dominant process for large particle emissions.

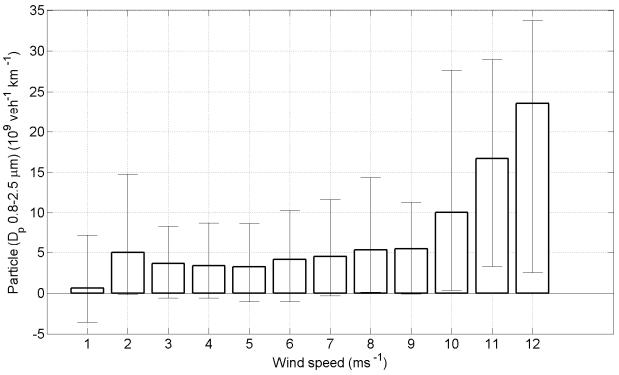

Fig. 6. Median number emission factor for particle size ($D_p = 0.8$–2.5 µm) within constant wind speed intervals Vertical bars represent the 25, 75 percentiles.

3.6 Comparison of different methods to estimate emission factors

Table 2 gives an overview of $PM_{2.5}$ mass EFs in [g veh^{-1} km^{-1}] as reported in different studies. Emission factors range from 0.01 to 0.99 g veh^{-1} km^{-1}. Highest value is reported by Kirchstetter. But it is difficult to compare the results from different studies due to different conditions and trends. Ketzel et al. (2007) present emission factors for Hornsgatan in Stockholm based on the NO_x method. They assumed that particles smaller than 0.6 µm Diameter are mainly originating from exhaust emissions and this may have decreased from 2004 to 2008 due to better particle filters in cars, but the major contribution to the total EFs of $PM_{2.5}$ is the mechanically produced particulate matter with a particle diameter larger than 0.6 µm (Ketzel et al., 2007). As shown by Ketzel et al. (2007) for Stockholm winter and spring months is a big source of mechanical produced particles presumably from studded tires used on Scandinavian roads. Norman and Johansson (2006) discuss the fact that meteorology, and in particular road wetness is the main parameter which controls $PM_{2.5}$ emissions. That being the case, the higher annual emission factor observed by Ketzel for measurements taken 2004, compared to our study, could be due to a dry spring period. Comparing the other studies shown in Table 2 it is noticeable that there is a relative wide range in the emission factors, which can be associated with the variability of the contribution from exhaust and non-exhaust PM, and/or the type of method. No clear bias was found which would explain the variety in emission factors.

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).

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

Authors	Vehicle type	Emission type Exhaust/non exhaust	Method [g veh km^{-1}]	Emission factor	Uncertainty	Type of road
Kirchstetter et al. (1999)	HD vehicles	N/A	NO_x	0.99	Sd 0.08	Road tunnel
	LD vehicles	N/A		0.0098	Sd 0.0009	
Kristensson et al. (2004)	All vehicles	Total	NO_x	0.067	0.005	Road tunnel
	All vehicles	Non exhaust	NO_x	0.027		
Harrison et al. (2006)	HD vehicles	N/A	NO_x	0.179	Sd 0.022	City street
	LD vehicles	N/A	NO_x	0.01	Sd 0.004	
Grieshop et al. (2006)	All vehicles	Exhaust	NO_x	0.022 (213)[a]	Sd 31	
Cheng et al. (2009)	All vehicles	N/A	Gradient	0.131	Sd 0.0369	Road tunnel
Ketzel et al. (2007)	All vehicles	Total	NO_x	0.054[b]	Not available	
	All vehicles	Total	NO_x	0.067[c]	Not available	City street
	All vehicles	Total	NO_x	0.029[d]	Not available	City street
	All vehicles	Total	NO_x	0.033[e]	Not available	City street
Keogh et al. (2009)	HD vehicles	exhaust		0.302		
	LD vehicles	exhaust		0.033		
This study	All vehicles	Total	NO_x	0.030[f]	(0.018,0.48)	Street Canyon
	All vehicles	Total	(EC)	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 MerseburgerStrasse, 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).

Table 3. Comparison between the different attempts to find a source parameterisation (F) for aerosol particles larger than 0.8 µm D_p. The traffic activity used is the mean daily traffic activity from (Monday-Friday, 24 values) in the NE sector over an area of 1 km × 1 km, (TA, traffic activity, LDV, light duty vehicles and HDV, heavy-duty vehicles).

Method	Statistics	Emission factor, (10^9 veh^{-1} km^{-1})		Intercept, (10^6 m^{-2} s^{-1})
Linear regression	R^2	EF, with 95 % confidence intervals		F_0, with 95 % confidence intervals
$F = \mathrm{EF}_{\mathrm{Fleet\ mix}}\mathrm{TA} + F_0$	0.70	8.2 ± 2.4		0.0005 ± 0.021
Multiple linear regression		$\mathrm{EF}_{\mathrm{LDV}}$	$\mathrm{EF}_{\mathrm{HDV}}$	
$F = \mathrm{EF}_{\mathrm{LDV}}\mathrm{TA}_{\mathrm{LDV}} + \mathrm{EF}_{\mathrm{HDV}}\mathrm{TA}_{\mathrm{HDV}} + F_0$	0.79	2.7 ± 4.3	72.7 ± 45	-0.0012 ± 0.021
EF based on CO_2 emission fluxes	0.55			
$F = \mathrm{EF}_{\mathrm{Fleet\ mix}}\mathrm{TA}$		4.4 ± 1.4		

3.7 Uncertainty of the calculated emission factors

In Table 3 we have calculated the emission factor for particles larger than 0.8 µm in diameter using the traffic database and the CO_2 method. It can be seen that the emission factors obtained by the CO_2 method are slightly lower than the one obtained using the traffic count.

Even though we used only the Northern sector to minimize the influence of other sources than traffic for particles and CO_2, there might be still some small sources which contribute to the CO_2 flux which would result in a lower emission factor. Another uncertainty is the conversion of the CO_2 flux into veh^{-1} km^{-1}. This involves assumptions regarding vehicle fleet and fuel consumption and that the other sources and sinks of CO_2 can be neglected. The slightly higher emission factors could also due to the fact that the emission database was not updated for the last years. A possible increase in traffic activity in this area would cause on over prediction of the emission factors using the Traffic counts. In addition it is noticeable that the EF for HDV is about 30 times higher than for LDV. These higher EF for HDV compared to LDV indicates that the trucks and buses are more efficient in suspending supermicron road dust particles than light duty vehicles. The diurnal variation in the particle flux is more

in agreement with HDV than LDV. One possible explanation might be that this coincidence of the diurnal variation of the particle flux and the HDV could also be due to influence of meteorological factors on the particle flux. One such factor is that road surfaces may be more frequently wet in the early morning which tend to suppress the particle suspension and leading to the lack of morning rush hour peak in the particle flux. But this needs further studies to be resolved. In summary, these emission factors are useful for atmospheric models where sources are introduced at the top of the surface layer, but do not represent the actual amount of particles at street level for human exposure.

4 Summary and conclusion

Size-resolved vertical aerosol number fluxes of particles with $D_p = 0.25$–$2.5\,\mu m$ were measured with the eddy covariance method from a 118 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 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 for the sector with most dense traffic (NW).
2. Unlike the flux results, the maximum in particle concentration was found in the East to South (90 to 200 degree), likely due to influence of long range transport.
3. Particle and CO_2 fluxes show the same diurnal pattern with low values at night and high values 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. The annual mass emission factor obtained with the eddy covariance method ($D_p = 0.25$–$2.5\,\mu m$) at the communication tower is similar to that determined with the NO_x method ($PM_{2.5}$) in the street canyon (28, 30 [mg veh^{-1} km^{-1}]) respectively.
6. The mass emission factors are higher in spring early summer than in late summer, autumn and winter.
7. The emission of particles ($D_p = 0.8$–$2.5\,\mu m$) increase with wind speed for wind speeds $>9\,m\,s^{-1}$. Below $9\,m\,s^{-1}$ no wind speed dependence was observed.
8. HDV and LDV Emission factors were determined by a using traffic counts and compared with EFs obtained by CO_2-method, which resulted in good agreement for the fleet mix.
9. EFs for HDV are 30 times larger than for LDV which indicates that the trucks and buses are more efficient in suspending road dust particles ($D_p = 0.8$–$2.5\,\mu m$).

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