Aerosol Particle Sources Affecting the Swedish Air Quality at Urban and Rural Level.

Kristensson, Adam

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Real-world traffic emission factors of gases and particles measured in a road tunnel in Stockholm, Sweden

Adam Kristensson[a,*], Christer Johansson[b,c], Roger Westerholm[e], Erik Swietlicki[a], Lars Gidhagen[b,d], Ulla Wideqvist[b], Vaclav Vesely[b]

[a] *Division of Nuclear Physics, Lund University, P.O. Box 118, SE-221 00 Lund, Sweden*
[b] *Institute of Applied Environmental Research, ITM, Air Pollution Laboratory, SE-106 91 Stockholm, Sweden*
[c] *Air Quality and Noise Analysis, Stockholm Environment and Protection Agency, P.O. Box 38024, SE-100 64 Stockholm, Sweden*
[d] *Swedish Meteorological and Hydrological Institute (SMHI), SE-601 76 Norrköping, Sweden*
[e] *Department of Analytical Chemistry, Stockholm University, SE-106 91 Stockholm, Sweden*

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Abstract

Measurements in a road tunnel in Stockholm, Sweden give the real-world traffic emission factors for a number of gaseous and particle pollutants. These include 49 different polycyclic aromatic hydrocarbons (PAH), CO, NO_X, benzene, toluene, xylenes, aldehydes, elements and inorganic/organic carbon contained in particles, the sub-micrometer aerosol number size distribution, $PM_{2.5}$ and PM_{10}. The exhaust pipe emission factors are divided with the help of automated traffic counts into the two pollutant sources, the heavy-duty vehicles (HDV) and light-duty vehicles (LDV). The LDV fleet contains 95% petrol cars and the total fleet contains about 5% HDV. When data permitted, the emission factors were further calculated at different vehicle speeds. The current work shows that average CO, NO_X and benzene emission factors amounted to 5.3, 1.4 and 0.017 g veh^{-1} km^{-1}, respectively. Since the mid-90s CO and benzene decreased by about 15%, carbonyls by about a factor 2, whereas NO_X did not change much. PAH emission factors were 2–15 times higher than found during dynamometer tests. Most particles are distributed around 20 nm diameter and the LDV fleet contributes to about 65% of both PM and particle number. In general, the gaseous emissions are higher in Sweden than in USA and Switzerland, foremost due to the lower fraction catalytic converters in Sweden. The PM and number emissions of particles are also slightly higher in the Swedish tunnel.

Keywords: $PM_{2.5}$; Polycyclic aromatic hydrocarbons; Particle size distribution; NO_X; Aldehydes

1. Introduction

The motorized vehicles are major contributors of polluted air in many urban areas. To be able to characterize to what extent the traffic or different vehicle types contribute to the city pollution detailed information of the emissions is needed. Emission factors can be used for this purpose and they may be expressed as the amount of species emitted per vehicle km driven or per volume of fuel consumed. So far, considerable information of the gaseous exhaust pipe emission factors and some of particulate matter is available from the 90s of individual vehicles from so-called laboratory dynamometer tests. These include gasoline/diesel light-duty vehicle (LDV) and heavy-duty vehicle (HDV) emission factors (see e.g., Westerholm and Egebäck, 1994; Burman, 1999; Sjögren et al., 1996; Hall et al., 2001, Färnlund et al., 2001; Andersson and Wedekind, 2001; Tobias et al., 2001, etc.). To simulate more realistic driving patterns and conditions found in real-world

*Corresponding author. Tel.: +46-46-2227635.
E-mail address: adam.kristensson@pixe.lth.se (A. Kristensson).

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traffic situations, so-called "car-chasing" experiments can be performed directly behind moving vehicles to obtain emission factors of individual vehicles (Kittelson et al., 2000). Another way to obtain emission factors from single vehicles in a traffic environment is to use the FEAT-technique (Sjödin and Lenner, 1995). These dynamometer and on-road methods provide information about the effect of fuel brands/additives, acceleration influence, engine make, temperature, dilution, etc. on the emissions.

However, when information about the contribution from traffic to pollution in an urban area is required, results for individual vehicles need to be complemented. Emission factors from the entire fleet during real-world conditions at a site typical of the urban area must be examined, since individual vehicles have such diverse emission profiles and cannot be representative of a specific fleet. Such information can basically be obtained in three ways:

1. Via road tunnel measurements (see e.g. Sagebiel et al., 1996).
2. Via "inverse" dispersion modeling of street-canyon measurements (see e.g. Ketzel et al., 2003).
3. Via mass conservation box models of open-road measurements (see e.g. Jamriska and Morawska, 2001).

So far, the most extensive work has been done in USA and Switzerland, where specific emission factors from the LDV and HDV fleet are available for a number of hydrocarbons, CO, NO_X, some polycyclic aromatic hydrocarbon (PAH), particle number and mass, etc. (e.g. Sagebiel et al., 1996; Kirchstetter et al., 1999; Staehelin et al., 1998; Weingartner et al., 1997). However, to be able to obtain the emissions in other countries where the vehicle composition and fuel brands are different, the measurements must be repeated.

There are already a limited number of emission factors for some gaseous species, PAH and particle number size distribution presented for Danish and Swedish conditions (Sjödin et al., 1996; Wingfors et al., 2001; Ketzel et al., 2003; Wåhlin et al., 2001). In this study, real-world emission factors are calculated from measurements during the 1998/1999 winter in a traffic tunnel in Stockholm, Sweden. The sub-micrometer particle number size distribution is measured, as well as $PM_{2.5}$, PM_{10}, particle elemental composition, organic and inorganic (C_{org} and C_{ele}, respectively) carbon particulate fraction, 49 different PAH, carbonyls, CO, CO_2, NO_X, aldehydes, benzene, toluene and finally xylenes. Whenever possible, the emission factors are divided into the two types HDV and LDV. Since they are also speed dependent, a separation for speed is made. Finally, the emission factors obtained from the tunnel measurements are compared with typical values derived from emission models representative for Swedish conditions. These results may be used as input in air quality dispersion modeling of urban air in e.g. Stockholm, Sweden.

2. Method

2.1. Sampling and analysis

The measurements were conducted in a densely trafficked road tunnel ("Söderledstunnel") in central Stockholm, Sweden from December 1998 to February 1999. Two sites were used, 370 and 965 m inside the tunnel, respectively, to quantify the emissions of gaseous and particulate pollutants from traffic. The tunnel is 1.5 km long. There are two lanes in the tunnel bore with traffic moving in a northerly direction towards the city center. First 735 m of the bore has a positive slope of 0.1%, the following 370 m has a slope of 1% and the rest of the tunnel is sloping 2.6% downwards.

Tunnel air was drawn via Hi-volume PM_{10}-inlets into a space housing the instruments at both sites. In January 1999 a $PM_{2.5}$-inlet was installed in the flow to achieve a change of sampling to particles below 2.5 μm diameter. Gaseous species measured in the tunnel were carbonyls, 49 different semi volatile and particulate PAH, NO_X, CO, CO_2 and BTX (benzene, toluene and xylenes). Particle measurements included the size distribution (3–900 nm), PM_{10}, $PM_{2.5}$, and $PM_{2.5}$ elemental composition and elemental and organic carbon. Measurements and methods and their resolution and availability at the 370 m site are summarized in Table 1. All measurements are available at the 965 m site.

The BTX species were sampled using a BTX monitor (CP-7001 BTX-Multi Component Monitor, Chrompack, The Netherlands). The aldehydes and ketones were collected on 2,4-dinitrophenylhydrazine (Sep-Pak® DNPH-Silica Cartridges). The carbonyls react with the hydrazine and form the corresponding hydrazone, which was eluted with acetonitrile (ACN), tetrahydro furane (THF) and water, and analyzed by high-performance liquid chromatography (HPLC, Perkin Elmer 200). Forty-nine particulate PAH were collected on Munktell glass fiber filters (MG 160, Munktell Filter, Sweden) and the semi-volatile PAH were sampled on polyurethane foam plugs (PUF) (75 mm diameter × 50 mm, 60 PPI; Specialplast AB, Sweden) (Westerholm et al., 1991). The samples were analyzed by gas chromatography-mass spectrometry (GC-MS), for quantification as described in detail by Westerholm et al. (2001). NO_X was measured using an AC 30M, Environment SA, France, which is based on the reaction of NO with ozone and detection of chemiluminescence of NO_2. NO_2 was also measured with the AC 30M by first converting NO_2 to NO using a Molybdenum converter at 350°C. The

Table 1
Species measured in the tunnel and associated methods

Species measured	Method	Time resolution	Available 370 m
Benzene, toluene, xylenes	BTX (continuous Chrompack) GC/FID detection	1 h	—
Aldehydes and ketones	DNPH-cartridge, HPLC analysis	1 h (every 2 h)	X
49 PAH	Glassfibre filters, polyurethane foam plugs, GC/MS analysis	1 h (3 times daily)	—
Elements Al, Si, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br, Sr, Zr, Mo and Pb.	PIXE analysis ($PM_{2.5}$ and PM_{10})	1 h (every 3 h)	X
Aerosol size distribution	DMPS	15 min	—
NO_X	Chemiluminescence	15 min	X
CO	Non-dispersive infrared technique	15 min	X
$PM_{2.5}$ and PM_{10}	TEOM	15 min	X
Elemental and organic carbon	ACPM	1 h	—

Measurements that are available at the 370 m site beside the one at 965 m are marked with an X.

Thermo Electron Corporation (USA) Model 48 instrument was used to measure CO. This instrument is based on a non-dispersive infrared technique.

On line measurements of $PM_{2.5}$ and PM_{10} were conducted using the Tapered Element Oscillating Microbalance particle monitor (TEOM®, Series 1400a, Rupprecht & Patashnick Co., USA) (Patashnick and Rupprecht, 1999). On line measurements of the organic and inorganic carbon mass fraction for $PM_{2.5}$ was accomplished with the Ambient Carbon Particulate Monitor (ACPM, Series 5400, Rupprecht & Patashnik Co., USA) (Rupprecht et al., 1995). Finally, $PM_{2.5}$ and PM_{10} particles where collected on filter strips with the Stationary Aerosol Monitor (SAM, Hansson and Nyman, 1985) and subsequently analyzed with the Particle Induced X-ray Emission (PIXE) method (Johansson and Campbell, 1988) for elemental composition of elements Al and heavier. A Differential Mobility Particle Sizer (DMPS), described in detail by Zhou (2001) measured the particle size distribution on-line in the size range 3–900 nm diameter.

Categorized traffic volumes and speed, relative humidity, temperature and wind speed were measured continuously with 15 min time resolution in the tunnel. The LDV were defined to be those with a wheel pair distance smaller than 5.5 m and vice versa for the HDV (generally this translates to the situation where LDV are lighter than 3.5 t).

2.2. Derivation of average emission factors

The ventilation system was closed during the measurements, meaning that the vehicles pushed the air forwards in the tunnel, hence creating the airflow through the tunnel. In an earlier study in the tunnel (Johansson et al., 1996), wind speed, v has been correlated with the total airflow, V measured using SF_6 as a tracer. V is obtained from $V = 79.5v + 15\,(\mathrm{m^3\,s^{-1}})$. The correlation was used in this study to obtain the total emission from the vehicle fleet:

$$E_{\mathrm{tot}} = \Delta C \cdot V/D, \tag{1}$$

where D is the distance between the two measurement sites (595 m) in the tunnel and ΔC the corresponding difference in concentration.

Not all species were measured at 370 m (see Table 1), but were only available at the 965 m site and consequently ΔC in Formula (1) could not be calculated straightforwardly in those cases. For this reason, an attempt was made to use the levels measured during street measurements in Stockholm as the concentrations in the tunnel entrance (0 m), since the levels in street canyons and in the tunnel entrance should be about the same. Then, by employing the measured values at 965 and 0 m, respectively, and with $D = 965$ m, it should be possible to calculate E_{tot}. However, this was not the feasible, since the air entering the tunnel was mixed with air exiting the parallel tunnel bore with emissions from vehicles driving in the opposite direction. Therefore, another approach was tried to deduce the concentrations at the tunnel entrance. Namely, from a comparison between the $PM_{2.5}$-levels and the organic carbon levels at 965 m ($C_{\mathrm{m}}^{\mathrm{PM_{2.5}}}$ (965 m)and $C_{\mathrm{m}}^{\mathrm{org}}$ (965 m), respectively) it became clear that these correlated well with each other ($R^2 = 0.61$). Thereby there was no reason to assume that they did not correlate also if measured at the tunnel entrance. Hence, a linear extrapolation was made to 0 m of the $PM_{2.5}$ levels ($C_{\mathrm{m}}^{\mathrm{PM_{2.5}}}$ (0 m)), using the 965 and 370 m sites and the levels of the organic carbon at 0 m (C^{org} (0 m)) were then calculated as

$$C^{\mathrm{org}}\,(0\,\mathrm{m}) = C_{\mathrm{m}}^{\mathrm{org}}\,(965\,\mathrm{m}) \cdot C_{\mathrm{m}}^{\mathrm{PM_{2.5}}}\,(0\,\mathrm{m})/C_{\mathrm{m}}^{\mathrm{PM_{2.5}}}\,(965\,\mathrm{m}) \tag{2}$$

for each data point. The missing species might as well have been calculated at 370 m, though 0 m was used to

be able to compare to street levels and see if the concentrations were reasonable. C_{ele} was also used in conjunction with $PM_{2.5}$ to calculate the levels at 0 m, whereas BTX was based on CO and the particle size distribution on NO_X. The motivation for this approach was again the high correlations of these species with each other measured at the 965 m site (particle size-NO_X, $R^2 = 0.74$, benzene-CO, $R^2 = 0.65$, C_{ele}-$PM_{2.5}$, $R^2 = 0.62$). The levels at 0 m compared to 965 m varied between 0% and 80% depending on species and time of the day (higher at night). The average error of the emission factor resulting from these assumptions is on the order of 20% for particle number, 40% for BTX and 50% for C_{org} and C_{ele}. PAH were only measured during daytime and the levels at 0 m were set to 15% of the ones at 965 m. This was based on levels of NO_X at 0 m compared to 965 m giving an error of about 20% for the PAH emission factors.

To calculate the emission factor per vehicle km of the combined fleet of LDV and HDV (E), the total emission is divided by the number of vehicles (N) passing by in a certain time interval:

$$E = E_{tot}/N. \tag{3}$$

The derivation of the total emission, E_{tot} and the emission factor, E is explained in detail by Chang et al. (1981).

The total emission can also be divided into its two parts, which are tailpipe emissions:

$$E_{tot} = E_{LD} \cdot NLD + E_{HD} \cdot NHD, \tag{4}$$

where NLD and NHD is the number of LDV and HDV, respectively, and E_{LD} and E_{HD} their respective tailpipe emission factors expressed per vehicle km. If (4) is rewritten, linear regression can be used to split between LDV and HDV. With the specific fleet composition at hand, formula (5) yielded the most stable regression result for E_{HD} with NHD/NLD as the independent variable and with E_{tot}/NLD as the dependent:

$$E_{tot}/NLD = E_{HD} \cdot NHD/NLD + E_{LD}. \tag{5}$$

For E_{LD}, (6) proved most appropriate with NLD/NHD and E_{tot}/NHD as the independent and dependent variables, respectively:

$$E_{tot}/NHD = E_{LD} \cdot NLD/NHD + E_{HD}. \tag{6}$$

Method (6) will not work for E_{HD} when the species is mainly emitted from gasoline vehicles and E_{LD} is close to E, e.g. for CO and BTX. For those species only E will be presented, since the HDV fleet anyway contributes much less to the pollutants compared to the LDV fleet. C_{org} and C_{ele} emission factors could not be divided into E_{LD} and E_{HD} for unknown reasons and $PM_{2.5}$ since the time pattern of the traffic flow and $PM_{2.5}$ are different. $PM_{2.5}$ is influenced by other sources, namely road dust, tire wear and possibly also break wear.

2.3. Derivation of speed dependent emission factors

The average emission factor for the total fleet, E is calculated by Eq. (3) for the speed intervals, 35–47.5, 47.5–52.5, 52.5–57.5,…,87.5–90 km h^{-1} for CO, BTX, NO_X, total particle number, $PM_{2.5}$, C_{org} and C_{ele}. This results in a 10 interval speed dependence. Speed dependent E_{HD} and E_{LD} can on the other hand only be calculated for four different speed intervals using Eqs. (5) and (6), respectively, and is calculated for NO_X and the particle size distribution. To be able to see how well these four bins speed dependent emission factors, E_{HD} and E_{LD} re-create the true concentrations at 965 m, formula (7) can be used, which is derived from (1) and (4):

$$C\,(965\,\mathrm{m}) = C_m\,(0\,\mathrm{m}) + D/V \cdot (E_{LD} \cdot NLD + E_{HD} \cdot NHD), \tag{7}$$

where C (965 m) is the calculated concentration at 965 m and C_m (0 m) the measured at 0 m. Then insertion of the speed dependent E_{HD} and E_{LD} of NO_X and the particle size distribution in formula (7) is made. This does however not give significantly higher resemblance with the true measured concentration at 965 m than do speed independent emission factors, since the speed dependence is not strong. Nevertheless, the four interval binning suggested that the speed dependence of E_{LD} and E_{HD} should be fitted with a 3rd degree polynomial to further improve the speed dependent emission factors (vehicle speed between 35 and 90 km h^{-1}), i.e.

$$E_{HD} = a \cdot v^3 + b \cdot v^2 + c \cdot v + d, \tag{8}$$

$$E_{LD} = e \cdot v^3 + f \cdot v^2 + g \cdot v + h. \tag{9}$$

By using the four points from the regression a first guess could be found for the eight parameters. The improvement is achieved reducing the difference between C (965 m) (from (7)) and the true measured concentration (C_m (965 m)) by minimizing ξ^2 with a non-linear least-squares fitting method and varying parameters $a-h$ in Eqs. (8) and (9):

$$\xi^2 = \frac{\sum_{i=1}^{n}([C_i\,(965\,\mathrm{m}) - C_{m,i}\,(965\,\mathrm{m})]/\bar{C}_i)^2}{n}, \tag{10}$$

where $\bar{C}_i$ is the average of C_i (965 m) and $C_{m,i}$ (965 m) for each data point i and n the number of measurements ($\sim$550) available for the minimizing routine. The ξ^2-value could be reduced by about a factor 2 for both NO_X and the particle number size distribution using these speed dependent emission factors compared to speed independent emission factors. However, the emission factors of the smallest particles, <30 nm diameter could not be calculated for speeds lower than 70 km h^{-1} as will be seen later. For these particle sizes it was sufficient to use a two-degree polynomial to fit for speeds between 70 and 90 km h^{-1}. Again, Eq. (10) was

minimized. The resulting ξ^2-value for NO_X was about 30×10^{-3} and for the particle size distribution it varied between 8.7×10^{-3} and 23×10^{-3} depending on particle size.

3. Diurnal profiles

During a typical working day more than 30,000 vehicles pass through the tunnel in one direction. About 95% of these are LDV and this type accounts for the morning and afternoon rush-hour peaks depicted in the form of a traffic flow rate in Fig. 1. Out of the LDV vehicles, 60% are catalyst based and most are utilizing 95–98 octane lead-free gasoline, a small fraction being diesel-driven (5% on average, with slightly more diesel-driven taxis in the night). HDV are trucks, big vans and buses. They are most numerous around midday. The average gasoline and diesel fuel sulfur content is about 100 ppm. The average number for the whole period of LDV ($\pm$standard deviation) is $1296 \pm 776\,h^{-1}$ and for HDV $60 \pm 60\,h^{-1}$.

The levels of a species mainly emitted from gasoline vehicles such as carbon monoxide is expected to follow the daily traffic flow trend of the LDV. As can be seen in Fig. 2, the averaged working day profile of CO at the 965 m site closely follows the LDV flow except for the morning rush-hour peak with concentrations reaching $30\,mg\,m^{-3}$. This morning rush-hour peak is due to a regular traffic jam at this time compared to adjacent hours, which is exemplified by the decreased vehicle speed in Fig. 1. In fact, the total emission of CO is anti-correlated with vehicle speed ($R^2 = 0.65$). The BTX species have high concentrations in the afternoon as for the species typical for the gasoline LDV such as CO, but with no exceptional increase in emission caused by low vehicle speed during the morning rush hour. NO_X has high concentrations around midday as well (Fig. 2) due

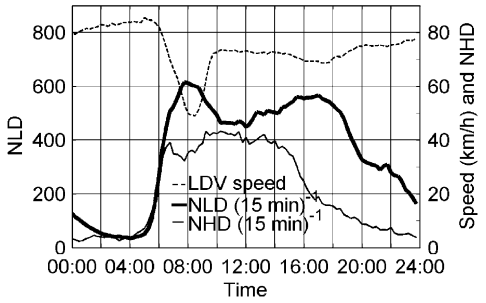

Fig. 1. Working day (Monday–Friday) average diurnal profile of the traffic flow in the Söderledstunnel (LDV—light-duty vehicle, NLD—number of light-duty vehicles, NHD—number of heavy-duty vehicles).

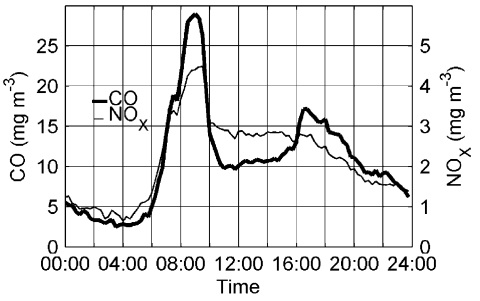

Fig. 2. Working day average diurnal 15 min profile of the CO and NO_X levels at the 965 m site.

to the relatively high emissions from the HDV at this time. An even stronger dependence on HDV is observed for e.g. formaldehyde, 2-butanone, acetaldehyde, methacrolein, etc., which have the highest concentrations around midday.

As for the particles, it can be seen from Fig. 3a that the number concentration of the smallest ones (<20 nm diameter) does not co-vary with typical species coming from vehicle exhaust emissions such as CO or NO_X, respectively. In fact, the levels go down during the morning rush hour and high levels remain in the night. Particles between 20 and 560 nm aerodynamic diameter are on the other hand highly correlated with NO_X (Fig. 3b). This means that these particles should originate from both fleets of LDV and HDV, i.e. have the same source as NO_X. The number of the largest particles from the DMPS measurements shows high resemblance with $PM_{2.5}$ (Fig. 3c), which does not correlate well with the traffic flow, suggesting that road resuspension is involved in the trends for large particles. Finally, C_{org} and C_{ele} are co-varying with $PM_{2.5}$ slightly better than with NO_X, although the explanation for this is not straightforward.

At non-working days, the traffic flow is increasing in the afternoon, however it is still lower than during the working days, especially for the HDV, which generally leads to lower concentrations of all pollutants than at working days.

4. Emission factors

Tailpipe emission factors will be presented in this section for the combined fleet of LDV and HDV (E) and separated for LDV and HDV (E_{LD} and E_{HD}, respectively). During the presentation of E it should be noted that deviations presented as the standard deviation from the mean also include the variability e.g. in the vehicle composition and meteorological conditions and not only the measurement errors themselves.

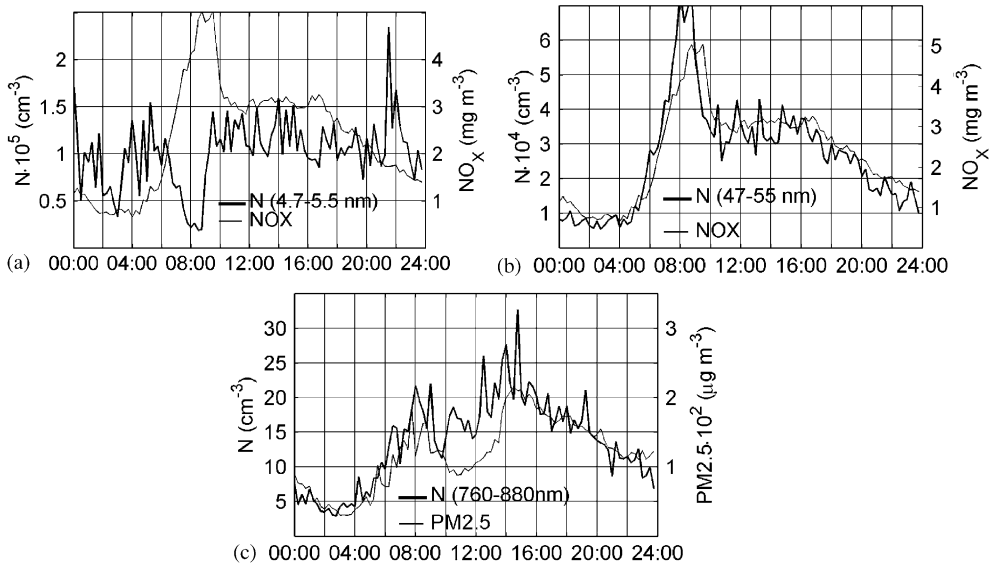

Fig. 3. Diurnally averaged profiles at the 965 m site. Number of particles: (a) 4.7–5.5 diameter with NO_X, (b) 47–55 nm diameter with CO and (c) 760–880 nm diameter with $PM_{2.5}$.

4.1. Gaseous emissions

Table 2 shows the development of the CO emissions between 1993 and 1999. It can be seen that the emissions have decreased by about 10–25% since the early/mid-90s to a value of $5.3 \pm 0.2\,g\,km^{-1}$ due to the increased fraction of catalytic converters among gasoline fueled LDV. The value is close to earlier measurements in other tunnels in Europe/Asia/North America from the first half of the 90s (Sjödin, 1999). The reason for the relatively high emission factors in the Söderledstunnel compared to other measurements is foremost the higher fraction of catalytic converters in those countries (see e.g. John et al., 1999). Diesel LDV/HDV contribution to the CO levels are negligible compared to gasoline LDV and therefore not considered in the discussion (Siegl et al., 1999; Palmgren et al., 1998). As for the speed dependent emissions, the most interesting observation is the large CO emission factor at speed intervals below $70\,km\,h^{-1}$ during congestion (Fig. 4a). It reaches $9\,g\,km^{-1}$ at $45\,km\,h^{-1}$, compared to the average value of $5.3\,g\,km^{-1}$.

The development for NO_X from 1993 to 1998/1999 shows that the combined average emission factor of both fleets ($E = 1.36 \pm 0.03\,g\,km^{-1}$) has not decreased significantly (Table 2). It is E_{LD} that remains nearly unchanged, while E_{HD} has decreased by about 30% since the mid-90s (c.f. Johansson et al., 1996). The EVA and COPERT model that are based on laboratory studies show that the emission factors for NO_X have decreased by about 30% for both vehicle types and that E_{LD} in the tunnel is higher compared to the models (Table 2). This might indicate the importance of emissions from relatively few old non-catalyst LDV present in the vehicle fleet. Fig. 5 shows that the emission factors of NO_X increase at high speeds ($>75\,km\,h^{-1}$) and are also higher, especially for the HDV during more congested type of traffic ($<70\,km\,h^{-1}$) when more accelerating and decelerating conditions are experienced. The parameters for the 3rd degree fit of NO_X emission factors are (Eqs. (8) and (9)); $a = 9.59 \times 10^{-4}\,g\,h^3\,km^{-4}$, $b = -0.186\,g\,h^2\,km^{-3}$, $c = 11.6\,g\,h\,km^{-2}$, $d = -221\,g\,km^{-1}$, $e = 3.62 \times 10^{-5}\,g\,h^3\,km^{-4}$, $f = -0.00638\,g\,h^2\,km^{-3}$, $g = 0.369\,g\,h\,km^{-2}$, $h = -6.05\,g\,km^{-1}$. Kirchstetter et al. (1999) show in an intercomparison between different tunnel studies that the NO_X emissions calculated on a $g\,kg^{-1}$ fuel consumption basis is invariant with roadway grade/speed. This means that when more fuel is consumed (as for accelerating situations ($v < 70\,km\,h^{-1}$) and high speeds ($v > 75\,km\,h^{-1}$)), the emission factors calculated per vehicle km should be higher as in this study. Generally, the contribution of the HDV fleet to NO_X levels is on the order of 20–50% (Fig. 5).

For the highly volatile species of BTX, it seems that the emission factors have decreased by abut 10% since 1995/1996 (Table 3), which is again a smaller decrease than predicted from dynamometer studies (ca. 50%, Table 2). The total emissions are dominated by LDV. The values of the BTX emission factors are about 70% lower than given in another Swedish tunnel study as indicated in Table 3, but that tunnel is subject partly to uphill/downhill driving as well as more mixed driving which might give the higher values. In general, the

Table 2
CO, NO_X and benzene average emission factors from different studies in Sweden given in g km^{-1} (Benzene mg km^{-1})

Study	Söderledstunnel 1993–1999 (winter, total fleet)[a,b]				Söderledstunnel 1998/1999[b]		EVA + COPERT model[c]											
Species	Winter 1993	Autumn 1994	Winter 1995/1996	Winter 1998/1999	LDV	HDV	1990			1995			1997			1999		
							tot	LDV	HDV	tot	LDV	HDV	tot	LDV	HDV	tot	LDV	HDV
CO	6.2±0.2	6.3±0.1	5.3±0.1	5.27±0.10														
NO_X	1.3±0.1	1.1±0.1	1.5±0.1	1.36±0.03	1.07±0.03	8.0±0.8	2.1	1.7	12.8	1.7	1.2	11.7	1.4	1.0	10.7	1.2	0.8	9.3
Benzene			20.6±1.7	17.3±0.3			83			53			36			27		

The LDV emissions are based on a 5% diesel share.
tot—total fleet.
[a] Johansson et al. (1997).
[b] This study. Average speed 75 km h^{-1}, stdv 8 km h^{-1}, NLD = 1296 h^{-1}, stdv 776 h^{-1}, NHD = 60 h^{-1}, stdv 60 h^{-1}.
[c] Ahlvik (1998) and Johansson et al. (2000). Average urban emission factors derived from exhaust lab dynamometer studies in Sweden and EVA and COPERT models.

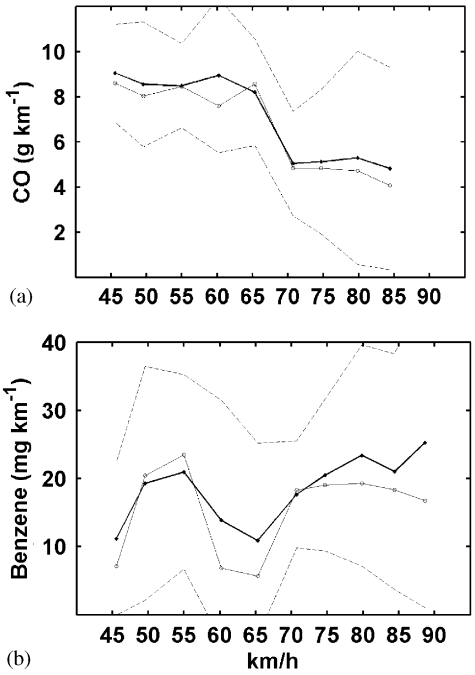

Fig. 4. CO (a) and benzene (b) emission factors. The thick line denotes average and the thin the median, the upper and lower dashed lines are values within standard deviation. Speed intervals are 35–47.5, 47.5–52.5, 52.5–57.5 km h^{-1}, etc.

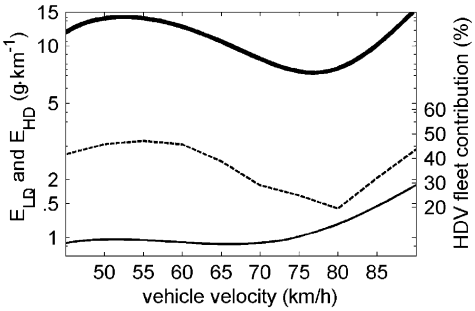

Fig. 5. NO_X E_{LD} (thin line) and E_{HD} (thick line) emission factors as a function of speed. The emission contribution of the entire HDV fleet to the total emission is denoted by a dashed line.

Swedish fuels and vehicle fleet tend to give higher emission rates than in the USA and other European countries since the fraction catalytic converters among the LDVs in gasoline cars is much lower in Sweden as mentioned before (Switzerland 1993: 60% fraction catalytic converters, Sweden 1998: 60%, and USA 1999: about 90%). Also, the introduction of methyl tert-butyl ether (MTBE) in gasoline in USA during 1995/1996 reduced the emissions of aromatic hydrocarbons (Kean et al., 2001 and Kirchstetter et al., 1996).

Table 3
BTX average emission factors from different studies given in mg km^{-1}

Species	Söderledstunnel winter 1995/1996[a]	Söderledstunnel winter 1998/1999[b]	Tingstad tunnel, Sweden, winter 1994/1995[c]	Gubrist tunnel, Switzerland, September/1993[d]		Allegheny tunnel, USA, September/1979[e]	Tuscarora tunnel, USA, June/1992[f]		Fort McHenry tunnel, USA September/1992[g]	
	Average	Average	Average	GV	DV	Average	LDV	HDV	LDV	HDV
Benzene	20.6±1.7	17.3±0.3	52±9	10.3±6.3	21±35		9.2±0.9	8.5±3.0	14.7±1.1	12.1±5.7
Toluene	76.5±5.9	67.3±1.0	120±21	20.4±7.0	33±36	38±1.5	14.2±2.3	13.7±7.6	28.5±2.6	17±14
m-xylene		33.4±0.6								
p-xylene		15.3±0.3								
m+*p*-xylene		48.6±0.8		10.8±3.1	27±16		10.5±2.2	10.7±7.2	23.9±4.9	41±27
o-xylene		19.6±0.4					4.0±0.9	4.3±2.9	8.8±1.6	14.0±8.6
Toluene/benzene	3.7	3.9	2.3	2.0	1.6	—	1.5	1.6	1.9	1.4

[a] Johansson et al. (1997).
[b] This study. Average speed 75 km h^{-1}, stdv 8 km h^{-1}, NLD = 1296 h^{-1}, stdv 776 h^{-1}, NHD = 60 h^{-1}, stdv 60 h^{-1}.
[c] Sjödin et al. (1996). Average vehicle speed, 70 km h^{-1}, down/up-hill up to 4% slope, highly mixed driving.
[d] Staehelin et al. (1998). +1.3% slope, 32–97 km h^{-1} vehicle speed, average 90 km h^{-1}. 60% catalyst fleet of GV = Gasoline Vehicles, DV = Diesel Vehicles.
[e] Hampton et al. (1983). 53% of the fleet catalyst based, −0.5% slope.
[f] Sagebiel et al. (1996). Very small slope, average speed 90 km h^{-1}.
[g] Sagebiel et al. (1996). Up/down-hill ±3.8%.

It could be mentioned that the emissions in Söderledstunnel of benzene in Fig. 4b show no clear speed dependence, although E at 45, 60 and 65 km h^{-1} seems to be lower.

The emission factors for aldehydes and ketones are presented in Table 4. It is evident that benzaldehyde and acetaldehyde have decreased substantially from 1995/1996, by about 70% and 90%, respectively. A large reduction is anticipated since the fraction catalytic converters among LDV has increased by about 10% during this time and non-catalytic cars release up to 100 times the amount of e.g. acetaldehyde compared to catalytic cars (Johansson et al., 1997). Other studies agree with the fact that the HDV have much higher emission factors than the LDV according to Table 4, although the contribution of the entire fleet of the respective vehicle types is of the same order since HDV are much fewer. The exception is crotonaldehyde, where the HDV fleet seem to emit 50 times more than the LDV fleet, which is not the case for the Tuscarora tunnel in 1999. The emission factors of LDV are comparable with other studies for formaldehyde and acetaldehyde, although the fraction catalytic converter is lower in Sweden. This is reasonable, since the addition of MTBE in gasoline in USA in 1995/1996 levelled out the cleaning effect of catalysts for these species (Kean et al., 2001). For benzaldehyde and the tolualdehydes, on the other hand, the emission factors from this study are much higher than in USA, since toluene and xylenes are precursors for their production (Kean et al., 2001), which are much less abundant in USA.

4.2. PAH emissions

Average PAH emission factors are shown in Fig. 6. PAH such as 9,10-dimethyl anthracene, dibenzo(a,l)pyrene, dibenzo(a,i)pyrene and dibenzo(a,h)pyrene were found to be below the detection limit more than 50% of the time, i.e. $<0.1\,\mu g\,km^{-1}$. As shown in the figure, low molecular weight PAH is predominantly associated with the gaseous phase, which is expected. The five most abundant PAH are: phenanthrene, fluorene, pyrene, fluoranthene and cyclopenta(cd)pyrene which are in the range of 30–75 $\mu g\,km^{-1}$. The emission factor of benzo(a)pyrene (B(a)P) was determined to be about 4.6 $\mu g\,km^{-1}$. Emission factors of B(a)P for a gasoline fueled light-duty gasoline car with three-way catalytic converter (TWC) based on dynamometer emission tests is 0.3 $\mu g\,km^{-1}$ and corresponding value for a heavy-duty diesel truck is $<0.1\,\mu g\,km^{-1}$ (Boström et al., 2002). This indicates that real world PAH emissions from vehicles are higher compared to emissions reported from dynamometer emission tests, possibly a factor of 15. On the other hand, using the phenanthrene emission factor in Fig. 6 (74 $\mu g\,km^{-1}$) and the corresponding

Table 4
Carbonyls average emission factors from different studies given in mg km^{-1}

Species	Söderledstunnel winter 1995/1996[a]	Söderledstunnel winter 1998/1999[b]			Tingstad tunnel, Sweden, winter 1994/1995[c]	Gubrist tunnel, Switzerland, Sep 1993[d]		Caldecott tunnel, USA, May 1999[e]	Tuscarora tunnel, USA, June 1992[f]		Tuscarora tunnel, USA Summer 1999[g]	
	Average	Average	LDV	HDV	Average	GV	DV	LDV	LDV	HDV	LDV	HDV
Formaldehyde	18±3	13±3	4.9±4.6	215±137	20	11±9	68±37	3.7±0.5	3.9±1.4	27±5	2.6	6.7
Acetaldehyde	28±8	3.1±0.8	1.4±1.8	50±35		2.3±1.2	15±6	0.96±0.01			0.64	4.0
Acetone		1.4±0.6	1.5±1.8	14±27		1.1±1.9	8.0±8.2				1.7	2.5
Acrolein		tfd	tfd	tfd		1.3±0.4	7.2±1.7				0.11	0.72
Propanal		0.47±0.20	0.58±0.46	6.9±10.4		0.18±0.45	3.2±2.1				0.12	0.63
Crotonaldehyde		1.8±0.5	0.04±0.79	49±15							0.15	1.1
2-butanone		0.2±0.5	—	—		0.08±0.44	1.2±2.1				0.12	0.75
Methacrolein		0.29±0.07	−0.20±0.15	6.6±4.5							0.10	0.69
m+*i* butanal		0.41±0.10	0.30±0.37	4.4±8.8								
Benzaldehyde	9.5±0.7	3.8±0.7	1.8±1.3	63±31							0.091	0.86
Cyklohexanone		tfd	tfd	tfd								
Isovaleraldeh.		tfd	tfd	tfd								
Valeraldehyde		tfd	tfd	tfd								
o-tolualdehyde		0.70±0.14	0.56±0.45	0.1±13.4							0.023	0.31
m+*p*-tolualdeh.		2.6±0.4	1.6±1.1	22±27							0.083	1.6
Hexanal		0.46±0.08	0.44±0.22	2.9±6.7				0.027±0.0015			0.014	0.54

tfd—too few data points.
The LDV diesel share is 5%.
[a] Johansson et al. (1997).
[b] This study. Average speed 75 km h^{-1}, stdv 8 km h^{-1}, NLD = 1296 h^{-1}, stdv 776 h^{-1}, NHD = 60 h^{-1}, stdv 60 h^{-1}.
[c] Sjödin et al. (1996). Average vehicle speed, 70 km h^{-1}, down/up-hill up to 4% slope, highly mixed driving.
[d] Staehelin et al. (1998). +1.3% slope, 32–97 km h^{-1} vehicle speed, average 90 km h^{-1}. 60% catalyst fleet of GV = gasoline vehicles, DV = diesel vehicles.
[e] Kean et al. (2001), +4.2% slope, only LDV, 94% equipped with 3-way catalytic converter, 2% diesel driven. Accelerated driving, from 52 to 71 km h^{-1}.
[f] Zielinska et al. (1996), 0.5% slope towards the center of the tunnel.
[g] Grosjean et al. (2001), 0.5% slope towards the center of the tunnel. Average speed 55 km h^{-1}. 52% LDV.

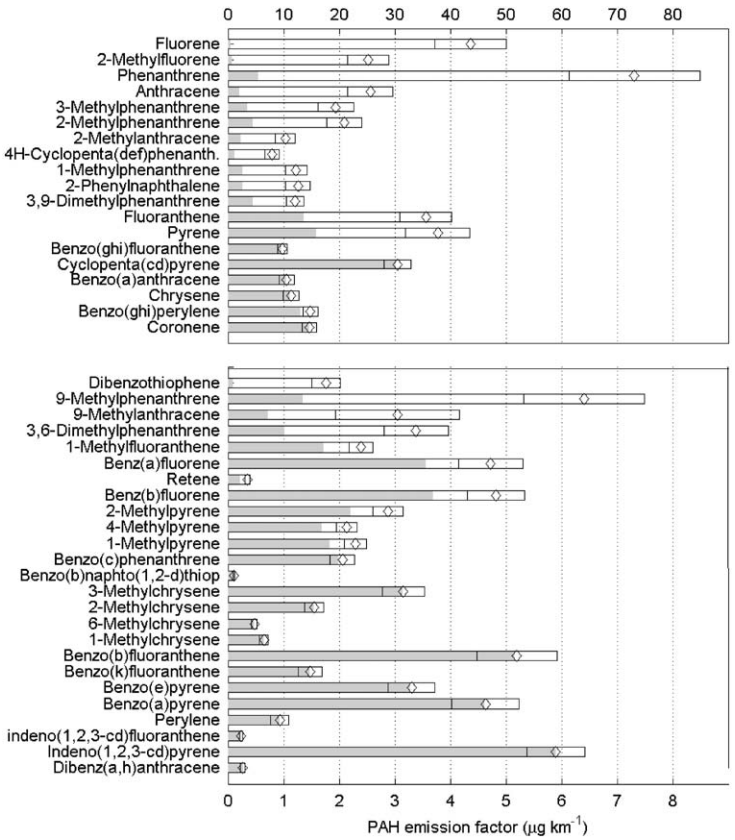

Fig. 6. Average PAH emission factors (diamonds) within standard deviation (vertical limits) of the combined fleet of LDV and HDV. The particle fraction is shaded.

emission factor reported by Boström and co-workers from a gasoline fueled TWC vehicle and from a heavy-duty diesel truck, i.e. 28 and 40 $\mu g\,km^{-1}$, respectively, reveals only a factor of about 2 higher emissions during real world driving. This discussion is based on assuming only exhaust PAH from vehicles in the tunnel. There could also be some contribution from PAH present in bitumen and tires.

By comparing PAH emission factors from this study with those reported by Wingfors et al. (2001) in Fig. 7, which are also measured in a road tunnel (Gothenburg, Sweden), it is evident that the emission factors of the more volatile PAH are comparable in magnitude. The emission factors for the less volatile PAH on the other hand are much higher in this study. The B(a)P emission factor, e.g. is a factor of 8 higher than reported by Wingfors. In the Wingfors-study, the amount of HDV was 8–24% of the total traffic volume. Wingfors et al. (2001) found that most of the particle bound carcinogenic PAH were not associated with HDV, which might explain the discrepancy between the studies depicted in Fig. 7, since the HDV share in this study is lower (about 5%).

4.3. Particle emissions

An earlier study has been conducted on the data used in this study to model the aerosol dynamic processes (Gidhagen et al., 2003). It has shown that about 40% and 80% particle number is lost between 10–29 nm diameter and 3–10 nm diameter, respectively, during the morning rush hour ($<70\,km\,h^{-1}$) due to coagulation and dry deposition to tunnel walls. The loss is too large to be able to calculate emission factors for the number size distribution and the size distribution will only be presented for particles larger than 30 nm diameter at speeds $<70\,km\,h^{-1}$. For higher speeds, the entire size distribution will be calculated. However, a significant number of particles are still lost during other times of the day due to deposition. This loss might possibly even reach a factor of 1.3 and 1.6 for particles 10–29 and 3–10 nm diameter, respectively (Gidhagen, unpublished data).

In Table 5, the emission factors of the total particle number between 70 and $85\,km\,h^{-1}$ are presented and they are seen to increase from 2.7×10^{14} to $11\times10^{14}\,km^{-1}$. The average emission factor of the

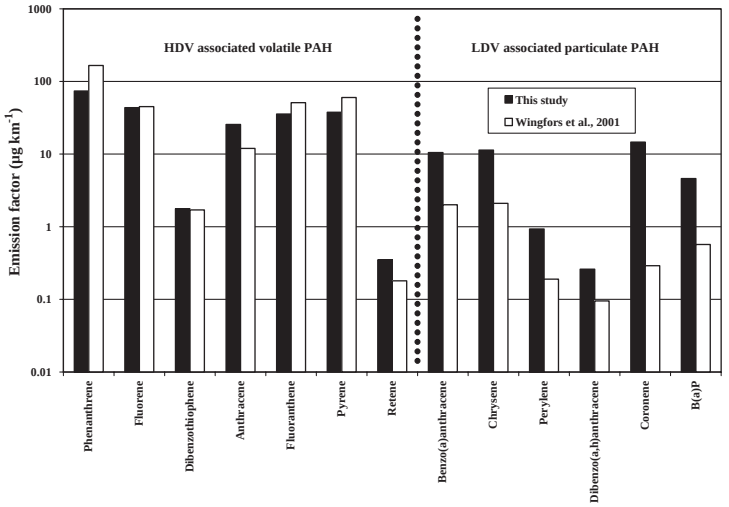

Fig. 7. Emission factors of PAH of the combined fleet of LDV and HDV from this study and from Wingfors et al. (2001) for some of the important PAH.

Table 5
Speed dependent particle number emission factors of the combined fleet of LDV and HDV and parameters of the E_{LD} and E_{HD} log-normal fitted size distribution emission factors

Parameter	Speed ($km\,h^{-1}$)						Average (70–90 $km\,h^{-1}$)
	50	60	70	75	80	85	
Total number of particles ($\times 10^{14}$ km^{-1})	—	—	2.7±1.1	3.3±1.4	5.4±3.4	11±5.7	4.6±1.9
NLD (h^{-1})[a]	2421	2224	2065	1489	606	335	1179
NHD (h^{-1})[b]	136	141	92	67	23	23	55
$dE_{LD}/d\log dp$[c]							
N_1 ($\times 10^{14}\,km^{-1}$)	—	—	0.035	0.045	0.12	0.32	
N_2 ($\times 10^{14}\,km^{-1}$)	—	—	1.7	1.9	2.7	4.0	
N_3 ($\times 10^{14}\,km^{-1}$)	0.17	0.15	0.15	0.17	0.18	0.20	
CMD_1 (nm)	—	—	3	3	3	3	
CMD_2 (nm)	—	—	20	16	16	16	
CMD_3 (nm)	77	77	77	77	77	77	
σ_1	—	—	1.25	1.25	1.25	1.3	
σ_2	—	—	1.9	2.1	2.1	2.1	
σ_3	1.75	1.75	1.75	1.75	1.75	1.75	
$dE_{HD}/d\log dp$[c]							
N_1 ($\times 10^{14}\,km^{-1}$)	—	—	0.80	1.0	2.0	4.0	
N_2 ($\times 10^{14}\,km^{-1}$)	—	—	20	17	30	50	
N_3 ($\times 10^{14}\,km^{-1}$)	3.7	3.0	2.0	2.3	3.5	6.3	
CMD_1 (nm)	—	—	3.2	3.2	3.2	3.2	
CMD_2 (nm)	—	—	17	17	17	17	
CMD_3 (nm)	75	75	75	75	75	75	
σ_1	—	—	1.7	1.7	1.7	1.7	
σ_2	—	—	1.85	1.85	1.85	1.85	
σ_3	1.75	1.75	1.75	1.75	1.75	1.75	

LDV fleet contains 5% diesel vehicles.
[a] Number of light duty vehicles.
[b] Number of heavy duty vehicles.
[c] See formula (11).

Söderledstunnel in this interval is $4.6 \times 10^{14}\,\mathrm{km}^{-1}$ and on the order of 1.5–15 times higher than in American outdoor and tunnel studies between 70 and 100 km h^{-1} (Kirchstetter et al., 1999; Kittelson et al., 2001; Abu-Allaban et al., 2002; Jamriska and Morawska, 2001). The higher emissions in Sweden might be due to the higher share diesel LDV, lower fraction catalytic converters and/or lower ambient temperature during measurements favoring nucleation (Kittelson et al., 2000). However, it is comparable to the emission factors at street levels, $3 \pm 0.5 \times 10^{14}\,\mathrm{km}^{-1}$ found in Copenhagen (Denmark), where there is a similar vehicle fleet composition, but measured at lower speeds ($\sim$50 km h^{-1}) and at higher temperature (on average 15.6°C in the Danish study and 3.2°C in the tunnel) (Ketzel et al., 2003).

The number size distribution emission factor is presented in Fig. 8 for three speeds, 50, 70 and 85 km h^{-1}. The distribution basically contains two fully discernible log-normal modes, one around 20 nm diameter (named 2) from nucleated cooling vapors containing e.g. organic carbon, sulfuric acid, etc. (Kittelson, 1998) and the other around 80 nm diameter (named 3) consisting of aggregates of less volatile condensed hydrocarbons and soot (Fig. 8). A fresh secondary nucleation mode below 10 nm diameter is appearing (called mode 1) together with the onset of a resuspended mode above 300 nm diameter. In general, the number size distribution could not be divided between LDV and HDV for sizes above 500 nm diameter, since a large part is resuspended dust which is correlated to other parameters than the exhaust pipe emissions (see diurnal profile section). Modes 1, 2 and 3 are parameterized with the following log-normal model;

$$\mathrm{d}E/\mathrm{d}\log dp = \sum_{1}^{j=3} N_j \frac{\mathrm{e}^{-(\log(d/\mathrm{CMD}_j)^2/\log(\sigma_j)^2)}}{\sqrt{2\pi}\log(\sigma_j)}, \qquad (11)$$

where j are the different modes, d the diameter, CMD is the count median diameter, σ the width of the modes and finally, N the number of particles (km^{-1}) in the modes. Fitting parameters for $\mathrm{d}E_{\mathrm{LD}}/\mathrm{dlog}\,dp$ and $\mathrm{d}E_{\mathrm{HD}}/\mathrm{dlog}\,dp$ are given in Table 5. The log-normal fits are also presented in Fig. 8 and it can be seen that the shape of the size distribution is nearly independent with speed and almost the same for LDV and HDV.

Absolute LDV diesel emissions are nearly invariant with speed and dominating over gasoline LDV emissions for particles in mode 3 according to dynamometer studies (Andersson and Wedekind, 2001). This might explain why mode 3 does not change in absolute amount with speed in this study (c.f. Fig. 8 and Table 5). For the

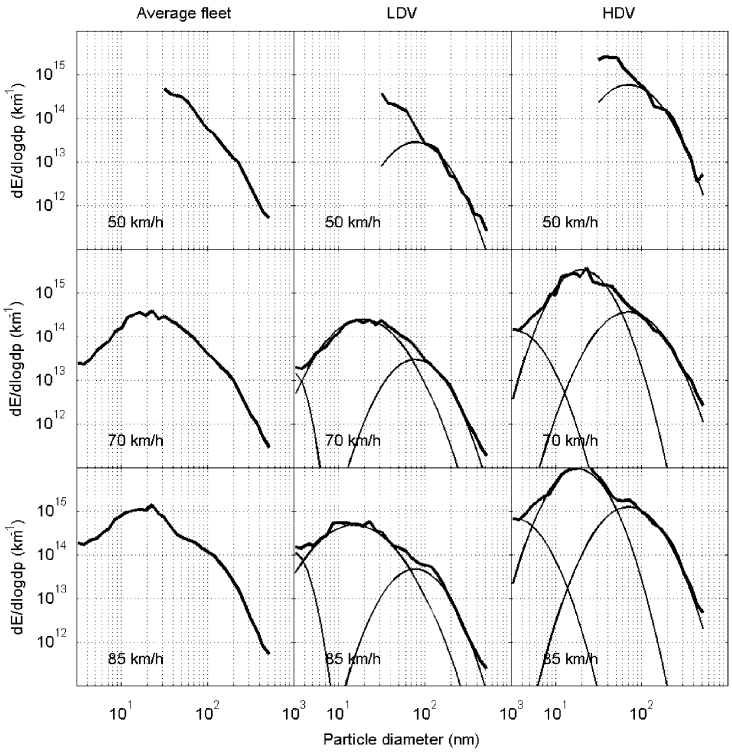

Fig. 8. The emission factor size distribution (thick line) of the average fleet, HDV and LDV (5% diesels), respectively, for three speeds. Fitted log-normal modes are drawn with thin line.

smallest particles in mode 1 and 2 on the other hand, it is shown by the same authors that gasoline LDV emission factors increase several orders of magnitude when going from 50 to 120 km h^{-1} and becoming similar to emission factors of diesel LDV. Since the amount of gasoline LDV is about 20 times higher than the diesel LDV share in this study, it is probable that they dominate emissions above 70 km h^{-1} for particles <30 nm diameter and that emission factors at lower speeds (~ 50 km h^{-1}) should be much lower, which is inferred from the study of Gidhagen et al. (2003) (about a factor 50). However, any speculation about the contribution to particle levels from the diesel and gasoline LDV, respectively, cannot be judged as the definite truth, since these vehicles are not counted separately. The shape of the HDV size distribution is similar to that measured by Kittelson et al. (2000) in car-chasing experiments. In that paper it is shown that the emission factor increases for all sizes during accelerating conditions compared to steady driving. The same tendency is observed in this study for the speeds <70 km h^{-1} during rush-hour like situations. Also, during stronger fuel consumption at higher speeds, the emission factor goes up (c.f. Fig. 8 and Table 5). The total HDV contribution to particle number varies between about 20% at 70–75 km h^{-1} to about 45% at 90 km h^{-1}.

The average $PM_{2.5}$ and PM_{10} emissions are 67 ± 4 and 236 ± 12 mg km^{-1}, respectively (Table 6). They are comparable to other American highway and Swiss tunnel studies, although slightly higher (c.f. Allen et al., 2001; Kirchstetter et al., 1999; Weingartner et al., 1997). It appears that the $PM_{2.5}$ emissions go up considerably for speeds above 70 km h^{-1} (Fig. 9a). C_{ele} and C_{org} follow these trends clearly (Fig. 9b and c) suggesting that the increase in $PM_{2.5}$ with speed is in some parts explained by a simultaneous increase in the emission of particulate hydrocarbons and soot particles, respectively. On average, though, inorganic carbon and organic carbohydrates seem to cover only about 50% of the exhaust part of the $PM_{2.5}$ emissions ($PM_{0.6}$), assuming that the exhaust part of $PM_{2.5}$ is $PM_{0.6}$ (c.f. Table 6 and Section 3). Similar importance of the inorganic and organic carbon in the $PM_{2.5}$ emissions was found in the Van Nuys tunnel (Fraser et al., 1998). These authors also showed that ammonium and nitrate species responsible for secondary production and condensational growth of particles in the exhaust plume cover large parts of the rest of the $PM_{2.5}$ mass. This is possibly also the case for the exhaust part of $PM_{2.5}$ ($PM_{0.6}$) in this study, though no evidence exists, since no measurements of those species are made. On the other hand, three other studies, two different in the Caldecott

Table 6
Emission factors of PM_{10} and $PM_{2.5}$ and the fractionation of $PM_{2.5}$ (mg km^{-1})

Species	Mass average			45–70 km h^{-1}			70–75 km h^{-1}			75–90 km h^{-1}		
PM_{10} (total)	236 ± 12			91			154			329		
$PM_{2.5}$ (total)	67 ± 5			33			41			100		
	tot	LDV	HDV	tot	LDV	HDV	tot	LDV	HDV	tot	LDV	HDV
$PM_{0.9}$ (total, LDV and HDV fleet contribution)	49	63%	37%	44	66%	34%	40	68%	32%	61	67%	33%
$PM_{0.6}$[a] (total)	40			36			31			51		
$PM_{2.5}$–$PM_{0.6}$ (resuspended) (total)[a]	27			−3			10			49		
Elemental carbon (total)	6.7			5.5			5.2			8.5		
Carbohydrates (total)[b]	11.2			7.4			8.2			16		
Al_2O_3 (total)[c]	5.7											
SiO_2 (total)[c]	10.0											
Fe_2O_3 (total)[c]	1.0											
NaCl (total)[d]	5.6											
SO_4^{2-}[e] (total)	1.0											
Fraction resuspended of $PM_{2.5}$	40%			≈0%			24%			51%		
Fraction oxides and NaCl of resuspended $PM_{2.5}$[c]	33%											
Fraction carbohydrates + elemental carbon of $PM_{0.6}$[a]	45%			36%			43%			48%		

The LDV fleet contains 5% diesel vehicles.

[a] Calculated from the DMPS and based on the assumption that all particles above 560 nm diameter are resuspended and that the particle density equals 1×10^3 kg m^{-3}.

[b] Based on the assumption that all carbohydrate mass is 1.2 times C_{org} (Schauer et al., 1999).

[c] Based on the assumption that all Fe, Al and Si are oxides and resuspended (soil origin).

[d] Based on the approximation that all Cl is on the form NaCl (salting of roads).

[e] Based on the assumption that all S is dissolved as SO_4^{2-}.

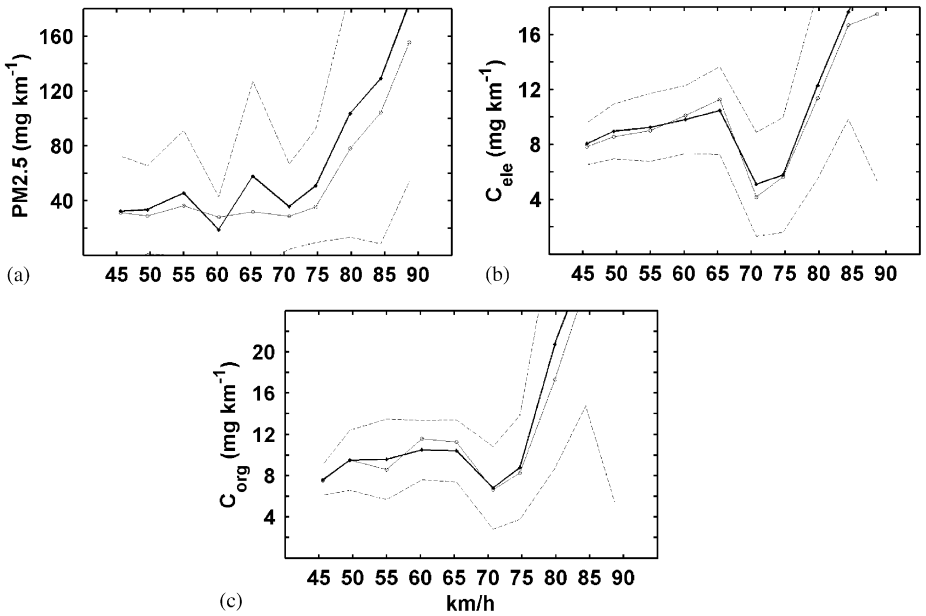

Fig. 9. $PM_{2.5}$ (a), C_{ele} (b) and C_{org} (c) emission factors. The thick line denotes average and the thin the median, the upper and lower dashed lines are the values within standard deviation. The speed intervals are 35–47.5, 47.5–52.5, km h^{-1}, etc.

tunnel (Allen et al., 2001; Kirchstetter et al., 1999) and one in the Gubrist tunnel in Switzerland (Weingartner et al., 1997), show that almost all $PM_{2.5}$ mass is covered by C_{ele} and hydrocarbons, giving less room for any other species such as ammonium and nitrate. For the exhaust part of the $PM_{2.5}$ emissions in this study, it can moreover be mentioned that from a schematic calculation presented in Table 6, it seems that the HDV fleet contributes to about 35% of the particle mass emissions. The remaining part of the $PM_{2.5}$ emissions in this study ($PM_{2.5}$–$PM_{0.6}$) is probably mainly consisting of non-exhaust emissions, namely road dust. This fraction is higher at higher speeds, reaching a value of about 50% of the $PM_{2.5}$ above 75 km h^{-1} (Table 6), which is expected due to the increased turbulence created by the vehicles at higher speeds. Road surface material and salts analyzed in Table 6 explain a part of the re-suspension, but more is unexplained. Another part of the resuspension might be brake and tire wear, though only one species of the heavy elements (Zn) was high enough to account for any significant mass (Table 7) and still several orders of magnitude lower than the most important soil metal.

Table 7
Average (±stdev) emission factors of PIXE elements in the Söderledstunnel

Species (μg km^{-1})	f	Δf	No. of points
$PM_{2.5}$			
Al	2991.143	685.6874	35
Si	4477.707	1224.703	48
S	343.2089	69.71872	48
Cl	3402.589	908.6835	48
K	422.7323	132.8906	48
Ca	438.8359	143.5541	48
Ti	92.20528	20.26397	47
Cr	0.14109	1.56453	48
Mn	17.11581	4.97778	48
Fe	696.9147	258.9942	48
Co	4.90862	2.19462	29
Ni	0.14555	0.10023	42
Cu	0.2136	4.92296	48
Zn	23.65736	5.7654	48
Br	0.21002	0.50585	48
Sr	5.67983	2.02913	14
Zr	2.95101	2.66575	12
Mo	1.33673	1.16189	17
Pb	4.84289	2.2553	45

5. Conclusions

Although the fraction catalytic converters of gasoline vehicles is increasing in Sweden and the emission factors for gaseous species have decreased since the mid-90s, the emission factors are still lower (though of the same size order) in USA mainly due to an even higher fraction catalytic converters. A dependency on speed of the emission factors was also found and it varied differently depending on species. The CO and benzene emissions are dominated by the LDV fleet, whereas NO_X and aldehydes have contributions from both the LDV and

HDV fleets. Each HDV vehicle, however, emits much larger quantities of NO_X and aldehydes than an LDV. For PAH it has become clear from this study that emission factors during real-world conditions are higher than during dynamometer studies of individual vehicles.

Particle number emission factors are comparable between Denmark and Sweden and three modes could be clearly discerned in the number size distribution. The emission factors are increasing from 70 to 85 km h^{-1} both for mass and number. A larger emission of organic particle species, elementary carbon and re-suspended material explains a large fraction of this increase with speed. However, on average only 50% of the exhaust particle mass ($PM_{0.6}$) and only about 30% of the non-exhaust particle fraction ($PM_{2.5}$–$PM_{0.6}$) could be explained by the species measured in this study. Emissions due to wear of brake and tire is about 100 times smaller compared to the soil part of the re-suspension.

The emission factors from the traffic derived from this study are valid for the Swedish vehicle fleet of late 1990s and can be used during air quality modeling (AQM). It was however, not possible to asses the relative importance of LDV diesel and gasoline vehicle emissions since no separate registration of the two vehicle types was made.

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