

MEASUREMENTS OF AIR POLLUTION FROM A DANISH HIGHWAY

Research Notes from NERI No. 254

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Abstract: This report presents the results from a measurement campaign carried out at the Holbæk High-

> way during 2008. The objective of the campaign was to determine the emission factors for PM2.5 and PM₁₀ due to highway traffic. The campaign included measurements of NO_x, NO, NO₂, TEOM PM_{2.5}, TEOM PM₁₀, O₃, particle size distribution and local meteorology. The emission factors for PM_{2.5} and PM₁₀ were determined to 45 and 155 mg/(vehicle km), respectively. This is comparable to the emission factors previously determined for H. C. Andersens Boulevard in

Copenhagen and somewhat higher than found at Jagtvej, Copenhagen.

Emission factors, PM₁₀, PM_{2.5}, highway traffic, measurement campaign. NO_x, NO₂, NO, meteor-Keywords:

logical measurements, particle size distribution.

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Research Notes from NERI

Preface

This report was prepared as part of a cooperative project with the Danish Road Directorate on development of a GIS based version of the Danish air quality model OML-Highway, which is going to be used as a tool for estimation of air pollution of nitrogen oxides and particles (PM₁₀ and PM_{2.5}) from Danish highways. This new project was a follow up on a previous project were the specifications for the GIS based version of OML-Highway were made.

The model calculations using OML-Highway are among others based on input of emission factors for nitrogen oxide (NO_x) and particle mass (PM_{10} and $PM_{2.5}$) for highway traffic. However, the emission factors for particle mass are uncertain and at present only few measurements of the emission factors for particle mass from highway traffic have been carried out. Therefore a measurement campaign with focus on direct measurement of emission factors of $PM_{2.5}$ and PM_{10} from Danish highway traffic was included as part of the project. This report describes the measurement campaign and the results from the campaign.

The project was funded by the Danish Road Directorate.

Bjarne Jensen, Tom Rasmussen, Hans Nielsen, Morten Hildan, Henrik W. Madsen, Jane Søfting, Keld Mortensen and Axel Egeløv are acknowledged for their technical work and their valuable contributions to the project.

Summary

This report summarises the results from a measurement campaign at the Holbæk Highway during March and April 2008. The aim of the measurement campaign was to determine the emission factors for $PM_{2.5}$ and PM_{10} from highway traffic. The project was carried out in cooperation with the Danish Road Directorate which also financed the project.

During the campaign one measurement station was placed directly at the north side of the highway and one measurement station was placed about 150 m south of the highway. In this way it was possible to determine the air pollution from the highway traffic during periods with southerly wind direction. The measurements included NO_x, NO, NO₂, O₃, PM_{2.5} (TEOM), PM₁₀ (TEOM) and particle size distribution. Moreover, the meteorological conditions were also measured at the background station.

The concentrations of NO_x , NO, and NO_2 at the highway station (Station 1) were at the same levels as measured at H. C. Andersens Boulevard in Copenhagen during the same time period. The levels measured at the background station (Station 2) were similar to the levels at the urban background station in Copenhagen (H.C. Ørsteds Institute, Copenhagen). Moreover, the results showed that more than 75% of NO_x , NO, and NO_2 at the highway station came directly from the highway traffic.

The concentrations of PM_{2.5} at the highway station (Station 1) were comparable to the levels measured at the urban background station in Copenhagen (H.C. Ørsteds Institute, Copenhagen). The levels measured at the background station (Station 2) were comparable to the levels measured at the rural background station at Lille Valby.

When the results for $PM_{2.5}$ and PM_{10} are corrected for loss of volatile particle mass, then the particle mass of $PM_{2.5}$ and PM_{10} at the highway station originating from the highway traffic were estimated to about 10% and 45%, respectively. This is in agreement with other studies, which have shown that the long range transported fraction of the particle mass is relatively high. Consequently, the fraction of $PM_{2.5}$ and PM_{10} originating from the highway traffic is low.

The measurements of the particle size distribution showed, that the highway traffic is a quite strong source of ultrafine particles with diameters below 100 nm. For the entire measurement campaign the average particle number per cm³ was a factor of four higher at the highway station compared to the background station. The number size distribution showed that it was mainly the smallest particles (10 -50 nm) which came from the highway traffic. However, the highway traffic contributed also significantly to the particles in the range 50 - 150 nm typical for soot.

The average emission factors for $PM_{2.5}$ and PM_{10} were calculated to 45 and 155 mg/(vehicle km), respectively. This is similar to the emission factors previously determined for H. C. Andersens Boulevard in Copenhagen and somewhat higher than found at Jagtvej, Copenhagen. This indicates that the high velocity at the highway do not have a strong influence on the emissions of particles.

Sammenfatning

Denne rapport sammenfatter resultaterne fra målinger udført i forbindelse med en målekampagne ved Holbæk Motorvejen i perioden marts til april 2008. Målet med kampagnen var at bestemme emissionsfaktorerne for PM_{2.5} og PM₁₀ fra motorvejstrafik. Projektet blev udført i samarbejde med Vejdirektoratet, som ligeledes finansierede arbejdet.

I forbindelse med målekampagnen blev der opstillet en målestation direkte på nordsiden af motorvejen (Station 1) og en baggrundsmålestation omkring 150 m syd for motorvejen (Station 2). Hermed er det muligt at bestemme luftforureningen fra motorvejstrafikken når vinden blæser fra sydlig retning. Målingerne inkluderede NO_x, NO, NO₂, O₃, TEOM PM_{2.5}, TEOM PM₁₀ og partikelstørrelsesfordeling. De meteorologiske forhold blev ligeledes målt ved baggrundsmålestationen (Station 2).

Koncentrationerne af NO_x, NO, og NO₂ ved motorvejsstationen (Station 1) var på niveau med niveauerne målt på H. C. Andersens Boulevard, København for den samme tidsperiode. Niveauerne målt på baggrundsmålestationen (Station 2) svarede til niveauerne målt på bybaggrundsmålestationen (H. C. Ørsteds Institutet, København). Mere end 75% af NO_x, NO og NO₂ ved motorvejsmålestationen (Station 1) kom direkte fra motorvejstrafikken.

Koncentrationerne af $PM_{2.5}$ ved motorvejsmålestationen (Station 1) svarede til niveauerne målt på bybaggrundsmålestationen (H. C. Ørsteds Institutet, København). Niveauerne målt på baggrundsmålestationen (Station 2) svarede til niveauerne målt på landbaggrundsmålestationen ved Lille Valby.

Når resultaterne for $PM_{2.5}$ og PM_{10} blev korrigeret for tab af partikelmasse i TEOM-instrumenterne var andelen af partikelmassen, som kom fra motorvejstrafikken, henholdsvis omkring 10% og 45%. Dette er i overensstemmelse med resultater fra andre studier, som har vist, at en relativ stor del af partikelmassen generelt er langtransporteret. Dermed bliver bidraget fra motorvejstrafikken relativ lille.

Målinger af partikelstørrelsesfordelingen viste, at motorvejstrafikken er en betydelig kilde til ultrafine partikler med diameter under 100 nm. Som gennemsnit for hele måleperioden var det gennemsnitlige partikelantal fire gange højere ved motorvejsmålestationen (Station 1) end ved baggrundsmålestationen (Station 2). Partikelstørrelsesfordelingen viste, at det navnlig var de mindste partikler (10 – 50 nm), som kommer fra motorvejstrafikken. Dog bidrager motorvejstrafikken også betydeligt til partikler i størrelsesorden fra 50-150 nm, som er typisk for sodpartiklerne.

Emissionsfaktorerne for $PM_{2.5}$ og PM_{10} blev beregnet til henholdsvis 45 og 155 mg/(køretøjs km). Det svarer til det der tidligere er blevet målt for H. C. Andersens Boulevard i København. Emissionsfaktorerne målt for Jagtvej, København er lidt mindre end målt ved motorvejen. Dette viser, at den høje hastighed på motorvejen tilsyneladende ikke spiller en stor rolle for mængden af partikler udledt fra trafik.

1 Introduction

In recent decades the traffic intensity on the Danish highways has increased significantly and there is therefore a need for a tool, which can be used to estimate the air pollution of nitrogen dioxide (NO₂) and particles (PM_{2.5} and PM₁₀) from highway traffic. On this background NERI carried out a cooperative project in 2005-2006 together with and funded by the Danish Road Directorate. This first project was the first step (phase 1) towards development of a GIS-based version of NERI's air quality model OML-Highway. In phase 1 the overall specifications for the GIS based version of OML-Highway were made. The results from this project are reported in Berkowicz et al., (2007).

In 2007 the Danish Road Directorate funded phase 2 with the objective to make the operative GIS-based version of OML-Highway. As part of this project model calculations for two examples was carried out in order to demonstrate the abilities of the GIS-based version.

One of the conclusions from phase 1 was that the emission factors for particle mass emitted due to highway traffic are very uncertain (Berkowicz et al., 2007), because the emission of particles not only originates from exhaust but also from dust from road, wear of tires and brakes, and from salting of the highway during winter time. The effective emission factors for particles have until now only been measured in Danish cities, where the emissions of particles may be different from emissions due to highway traffic. To overcome this problem phase 2 included a measurement campaign with the objective to measure the effective emission factor for $PM_{2.5}$ and PM_{10} for Danish highway traffic.

This measurement campaign was carried out during April and March 2008 at a site on the Holbæk Highway (route No. 21) north of Tåstrup. This report describes the measurement campaign, the used methods and present the results from the campaign.

2 Measurement Campaign

The measurement campaign took place during the period from 08-03-2008 to 07-05-2008. The strategy behind the measurement campaign was to measure the particle mass (PM $_{2.5}$ and PM $_{10}$) and the concentrations of NO $_{x}$ simultaneously at two measurement sites close to a Danish highway. In this way we could isolate the contribution from the highway traffic and determine the emission factors for PM $_{2.5}$ and PM $_{10}$ relative to the emission factor for NO $_{x}$ (see explanation below).

During the campaign one measurement site (Station 1) was placed directly at the northern side of the highway which most often is the downwind side of the highway (Figure 2.1). The other measurement site (Station 2) was placed 250 m southwest from Station 1 and about 150 m south of the highway. Station 2 is most often on the upwind side of the highway. During southerly wind subtraction of the results from the two sites can be used to calculate the concentrations resulting from the highway traffic alone. This requires that no other local sources are present within the measurement area. This strategy has been used with success in a series of other studies and is also one of the main principles behind the selection of measurement sites in the Danish Air Quality Monitoring Programme (Kemp et al., 2008).



Figure 2.1 The two white measurement containers at the measurement site. One station was placed at the northern side and the other station was placed about 150 m south of the highway. The picture is taken from the bridge at Bondehøjvej.

The emission factor for PM can be determined relative to the emission factor for NO_x . This is based on the simple assumption, that both PM and NO_x are diluted in the same way. Based on this assumption the ratio between the concentration of PM and the emission factor of PM is equal to the ratio between the concentration of NO_x and the emission factor of NO_x :

$$PM/E_{PM} = C_{NO_{\pi}}/E_{NO_{\pi}}$$

where E_{PM} and E_{NOx} are emission factors for particles and NO_x , respectively. PM is the particle mass measured either as $PM_{2.5}$ or PM_{10} , and C_{NOx} is the concentration of NO_x . PM and C_{NOx} are the concentrations resulting from the highway traffic (Station 1 minus Station 2) for southerly winds. This can be rearranged to:

$$E_{PM} = E_{NO_{x}}(PM/C_{NO_{x}})$$

The total emission factor for NO_x from traffic are well known (e.g. the European COPERT 4; EEA 2007) and depend only on traffic intensity, composition of traffic and speed of traffic. Hence the total emission factor for PM_{10} and $PM_{2.5}$ can be calculated.

This method has previously been used by Ketzel and coworkers to determine the emission factors for particle mass at busy streets in Copenhagen (Ketzel et al., 2007).

The applied NO_x emission factors have been validated in a previous measurement and model project for Danish highway conditions (Jensen et al., 2004).

The particle mass is measured with a TEOM instrument that has a known artefact. Due to the measurement principle part of the particle mass evaporates, because the particles are heated to 50 °C during the measurements. Measurements have shown that the loss is the same at kerb site and urban background (Palmgren et al., 2003). It is therefore mainly the non local particle mass that evaporates. This means, that the PM measurements at both Station 1 and 2 will be affected equally since the particle mass due to non local sources are equal at the two stations. Evaporation of part of the particle mass is off course a disadvantage with the TEOM instruments. However, at present it is the only instrument that can measure particle mass with high time resolution (½ hour), and this is needed in this measurement campaign. Calculation of the contributions to particle mass due to the highway traffic can only be done with high time resolution due to frequent changes in wind direction during a day.

The measurements of particle mass and nitrogen oxides were supplemented with measurements of ozone and particle size distribution for the fine particles (range from 10 nm up to 700 nm).

2.1 Selection of measurement sites

There was a series of criteria for selection of the measurement sites:

- 1. High traffic density in order to ensure that the air pollution due to the highway traffic was relatively high compared to the background air pollution.
- 2. Few other local sources than the highway traffic.
- 3. The direction of the highway should preferable be east-west due to high frequency of wind from southerly directions.
- 4. Sufficient power supply.
- 5. Possibility and permission to position the measurement station at the site of the highway.
- 6. Easy access to the measurement sites and not too far from NERI.
- 7. Availability of traffic information.
- 8. Representative traffic composition.
- 9. Representative diurnal traffic pattern.
- 10. Simple terrain.

Based on inspection of maps and aerial photos a number of possible measurement sites were selected. After a visit to these sites it was decided, that the best site for the measurement campaign was at Highway 21 where Bondehøjvej via a bridge crosses the highway north of Taastrup (Figure 2.2 and 2.3). The Danish Road Directorate measures the traffic flow and speed at this site (measurement station No. 41). Criteria 1-9 were fulfilled here. The only disadvantages were with respect to criteria 10.

The terrain (criteria 10) is quite complex due to the high trees and the slopes down to the highway (Figure 2.1). This would not give direct problems for the measurement of the emission factor for particle mass. The only problem will be during comparisons of measurements and model results, because the model may not be able to give good results due to this quite complex terrain.

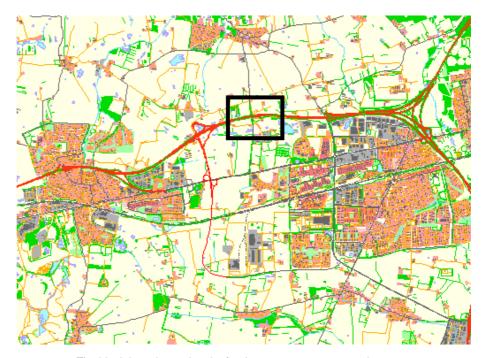


Figure 2.2 The black box shows the site for the measurement campaign.



Figure 2.3 Aerial view of the measurement site. The red dots indicate the position of the two measurement stations (Station 1 and 2) and the blue dot indicates the position of the meteorological mast. Source: Google Earth.

The highway curves slightly at the measurement site. This has to be taken into account when the wind directions usable for determination of the air pollution due to the highway traffic is selected. Figure 2.4 show that at directions between 85° and 245° the air mass arriving at the background measurement station (Station 2) will in principle not be affected by air pollution from the highway. However, due to short term fluctuations of the wind directions it is necessary to reduce the usable wind di-

rections. It is therefore estimated, that the measurements at Station 2 will not be affected by air pollution from the highway at wind directions from 105°-225° at low wind speeds and 95°-235° at strong wind speeds.

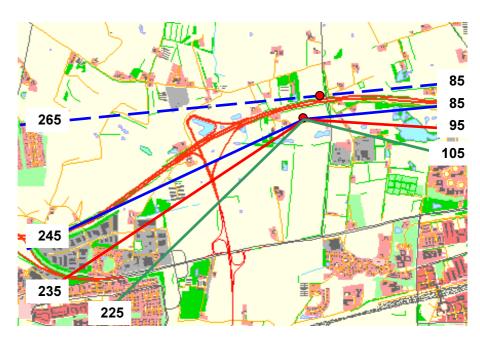


Figure 2.4 The numbers on the map illustrates the wind directions (degrees) where the measurements at Station 2 will not be affected by air pollution due to the highway traffic. At strong wind speeds this is between 105°-225° and at low wind speeds this is between 95°-235°.

2.2 Measurement stations, methods, quality assurance and data storage

The two NERI mobile measurement stations and the meteorological mast can be seen in Figure 2.5 and 2.6. The GPS positions are listed in Table 2.1.

Table 2.1 Position of Station 1, Station 2 and the meteorological mast. UTM 32 is with datum WGS84.

Meteorological mast:	UTM 32 E	UTM 32 N	Længde	Bredde
Station 1 Highway	327248	6172272.5	12° 15 min. 12 sec.	55° 39 min. 56 sec.
Station 2 Background	327125	6172066.5	12° 15 min. 6 sec.	55° 39 min. 49 sec.
Meteorlogical mast	327104	6172062	12° 15 min. 5 sec.	55° 39 min. 49 sec.

The measurement instruments used during the campaigns are listed in Table 2.2. For NO_x and O₃ NERI's accredited methods and quality control procedures were followed. The methods used for TEOM (Tapered-element Oscillating Microbalance) and DMPS (Differential Mobility Particle Sizer) followed standard procedures described by the manufacturer.

The meteorological measurements using a SONIC anemometer (10 Hz acoustic measurements in three dimensions of temperature, wind speed and direction) followed standard procedures described by the manufacturer.

The data from the instruments were send by modem to NERI and stored in NERI's databases. Near real time data from the measurement campaign were available via NERI's intranet. This ensured a fast preliminary quality control of the measurements. This improved the running time for the instruments and the quality of the data.

Moreover, parallel measurements in the laboratory with the two TEOM instruments and the two DMPS instruments were carried out prior and after the measurement campaign. This ensured that systematic errors between the measurements at Station 1 and 2 were as small as possible.



Figure 2.5 The NERI measurement station at Station 1 at the northern side of the highway. The mast and metal box in front of the picture is the Danish Road Directorate's traffic measurement station No. 41.



Figure 2.6 The NERI measurement station and the meteorological mast at Station 2 at the background position about 150 m south of the highway.

Table 2.2 Instruments used during the measurement campaign.

Parameter	Station 1 Highway	Station 2 Background	Instrument	Time resolution	Intake height	Uncertainty
NO _x , NO, NO ₂	Х	Х	API Chemiluminescent NOx Analyzer, Model 200A	½ hour	3.1 m	±10%+3 ppb
$\overline{O_3}$		Х	API Photometric O₃ Analyzer, Model 400A	½ hour	3.1 m	±10%+3 ppb
PM _{2.5} (TEOM)	Х	Х	Rupprecht & Patashnick Co., Inc., TEOM Particulate Mass Monitor Series 1400	½ hour	3.8 m	±1 μg/m³
PM ₁₀ (TEOM)	Х	Х	Rupprecht & Patashnick Co., Inc., TEOM Particulate Mass Monitor Series 1400	½ hour	3.7 m	±1 μg/m³
DMPS	Х	Х	Home build	½ hour	3.4 m	
Sonic Ane- mometer		Х	Model USA-1 from METEK GmbH	10 Hz / ½ hour	7.2 m	

2.3 Traffic at the measurement site

The Danish Road Directorate has delivered traffic data. Traffic data consist of data from an automatic traffic counting station and from short counts (manual counts) (Danish Road Directorate 2006). The traffic data are used to calculate emissions with the emission module of the WinOSPM. It is necessary to estimate the Average Daily Traffic (ADT), the vehicle distribution, speed, the diurnal variation, and traffic data as a time-series. ADT for the six lane highway was 58,291 in 2007.

2.3.1 Automatic traffic counts

Automatic traffic counts have been obtained from a permanent traffic count station as part of the so-called TRIM traffic monitoring system in Greater Copenhagen. The automatic traffic count station is located right next to the location of the air quality measurement equipment (Figure 2.5). Data are available for the full period of the air quality measurement campaign.

The traffic data include the number of vehicles and average speed in 15 minutes intervals subdivided in three vehicle length categories (0m - 5.80m, 5.80m - 12.50m, 12.50m -). The length is the length between axles of the vehicle. Data are available for each of the three lanes in each direction of the motorway.

In principle it should be possible to use the automatic traffic counts to estimate the vehicle distribution. However, there are limitations as the vehicle length 0m - 5.8m includes passenger cars and vans and even small trucks. Analysis of similar data has shown that automatic traffic data are not suitable for estimation of the detailed vehicle distribution needed for emission estimation. Therefore, the automatic traffic counts are only used to estimate Average Daily Traffic (ADT) and the diurnal variation on hourly basis. The average diurnal variation is described for Mondays to Thursdays, Fridays, Saturdays, and Sundays. This subdivision takes into account the differences in diurnal variation between different days of the week. A complete time-series (hour by hour) will also be generated for model calculations.

Former analysis has also revealed that it is not necessary to handle each lane separately but just each direction (three lanes together) to obtain good agreement between modelled concentrations at different distances from the motorway. Therefore, data are aggregated on a direction by direction basis.

The emission module of the WinOSPM model is used to generate emissions based on hourly means of traffic data.

2.3.2 Manual traffic counts

Manual traffic counts are only carried out at selected locations. Manual counts provide detailed information about the vehicle categories and the diurnal variation. Manual counts are carried out for one day hour by hour during 6:00 am to 6:00 pm. The vehicle categories counted are: passenger cars, vans, truck(>3,5t), truck with trailer, semi-trailer, busses, and motorcycles. Counts are carried out for each direction separately.

The locations that are closest to the location of the air quality measurement site have been chosen to represent the vehicle distribution at the site. Two manual traffic count sites were identified (Table 2.3). The locations of the two manual traffic counts are shown in Figure 2.7.

Table 2.3 The two manual traffic count sites.

Location	Description	Date
11-17/811	Holbæk motorway north of Vallensbæk Mose	March 20, 2007
11-17/811	Holbæk motorway north of Vallensbæk Mose	Sept. 9, 2007
11-47/106	Holbæk motorway at Kirke Såby East of Roskilde	March 20, 2007
11-47/106	Holbæk motorway at Kirke Såby East of Roskilde	Sept. 5, 2007
11-47/106	Holbæk motorway at Kirke Såby East of Roskilde	April 3, 2008





Figure 2.7 Locations of the two manual traffic count sites. Top: Overview. Middle and bottom: The two stations at higher spatial resolution.

3 Meteorology

The meteorological measurements were primarily established to determine wind direction in order to be able to select situations where the air pollution measurements at the background station (Station 2) were not affected by the highway emissions. Secondly, data were used to demonstrate that the local meteorological conditions were not abnormal. Thirdly, data will be used for detailed analysis of the air pollution measurements and will be processed for possible OML-Highway model calculations. The meteorological instrument and the position are described in Chapter 2.

The 10 Hz sonic measurements have been averaged to 10 minutes values. In Figure 3.1 the wind directions for the entire campaign are shown. The read dashed lines marks the wind direction sector where the highway emissions are not expected to have impacts on the measurements of the background concentration.

Wind speed and temperature are shown in Figure 3.2 and 3.3. Precipitation at Lille Valby 9 km to the west-north-west of the highway is presented in Figure 3.4.

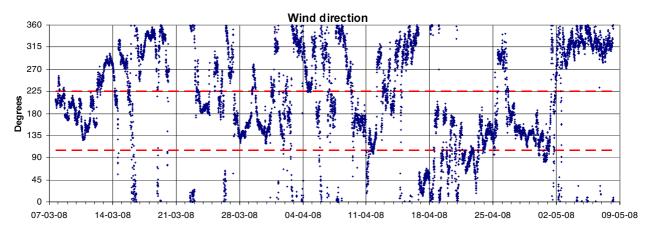


Figure 3.1 Wind directions during the campaign (10 min. averages). The red dashed lines mark the wind direction sector (105-225°) where the highway emissions have no impact on the background concentration.

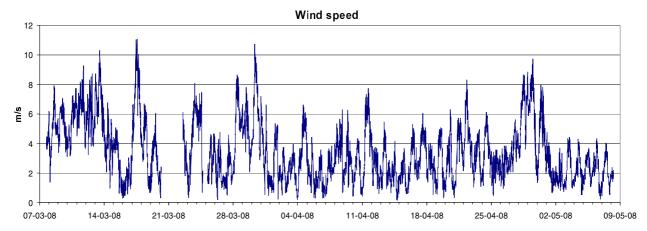


Figure 3.2 Wind speed during the campaign (10 min. averages).

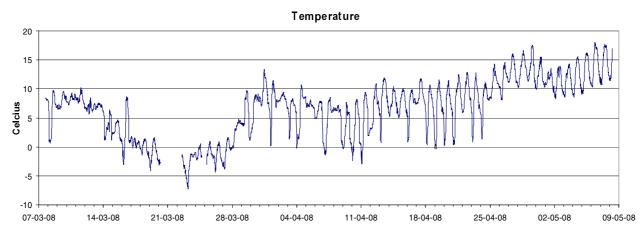


Figure 3.3 Temperature (10 min. averages).

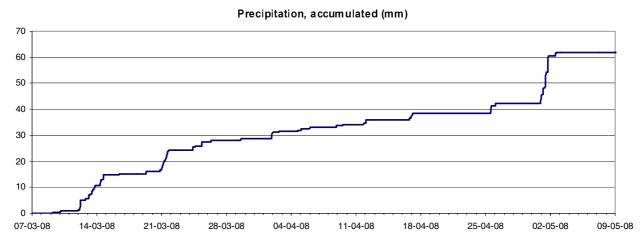


Figure 3.4 Accumulated precipitation measured at Lille Valby 9 km to the west-north-west of the highway. Data measured by Risø DTU.

One step in checking and preparing the meteorological data is to correct the raw sonic measurement for tilt (apparently non-horizontal mean flow). Tilt can be due to non-ideal circumstances in the physical placement of the mast and instrument, the terrain inclination and the minor flow distortion around the sonic. The correction is not vital for the parameters already shown, but for the turbulence parameters accounting for the dilution of air pollution the correction is essential. The tilts are shown in Figure 3.5. Here the approximately sinusoidal 'mean'-shape is due to the combined tilt of the mast/sonic and the ground. The offset of about 1 degree is due to flow distortion around the sonic.

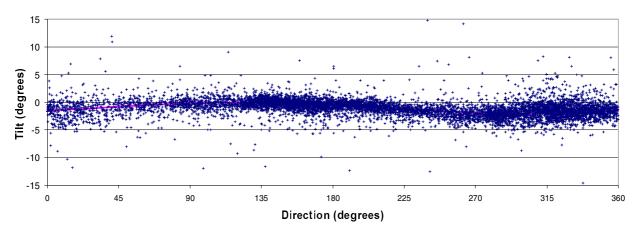


Figure 3.5 The tilt of the flow relative to the sonic (10 min. averages).

The main turbulence parameters accounting for the dilution of air pollution have been calculated and are shown in the next figures. Figure 3.6 shows the time variation of the friction velocity (u*). It is a parameter describing the mechanical turbulence generated due to the friction from the surface. In Figure 3.7 is shown the sensible heat flux from the ground to the atmosphere. It is a measure for the so-called thermic turbulence.

The aerodynamic roughness length, z0, is a parameter describing the roughness of the ground in the surroundings. It is calculated for each 10 minutes period and is primarily based on the friction velocity. In Figure 3.8 z0 is shown as a function of the wind directions. The tall trees at the highway in the northerly sector give rise to the relative high values of z0.

Over all, the meteorological measurements seem to behave as expected for the weather conditions during the campaign confirming that the quality of the results are acceptable.

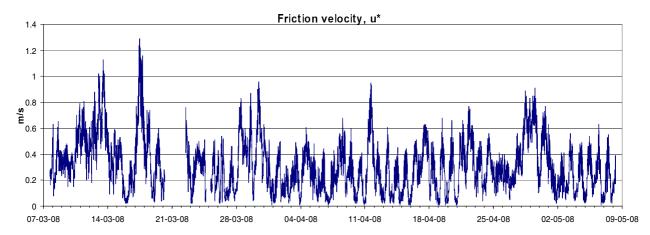


Figure 3.6 Friction velocity is a measure of mechanical turbulence.

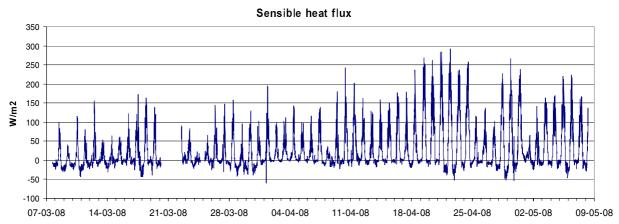


Figure 3.7 Sensible heat flux from ground to atmosphere (positive) responsible for generation of thermic turbulence.

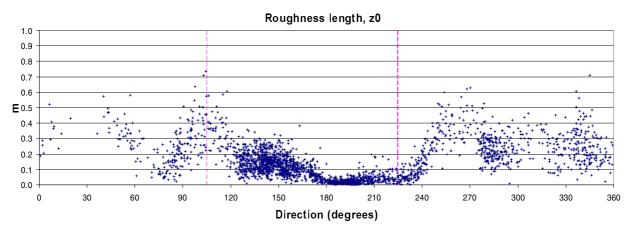


Figure 3.8 Aerodynamic roughness lengths, z0. The red dashed lines marks the wind direction sector where the highway emissions have no impact on the background concentration. The values above 0.05 m are due to vegetation (trees and bushes) in the surroundings at about 150 m distance.

4 Nitrogen Oxides, NO_x

Figure 4.1 to 4.3 presents the results from measurement of NO_x , NO, and NO_2 during the measurement campaign. Table 4.1 presents the number of measurements, average concentration and the 98 percentile. The average concentration of NO_x at the highway is about 5 times higher at the highway (Station 1) compared to the background (Station 2) clearly showing the influence of NO_x from the highway traffic. Similarly NO and NO_2 are 10 and 3 times higher, respectively. This pattern is as expected since NO_x from traffic is primarily emitted as NO.

The results from the measurements at the highway has been compared to the average concentrations of NO_x , NO, and NO_2 at selected monitoring stations around Copenhagen in the Danish Air Quality Monitoring programme (Kemp et al., 2008) for the period of the measurement campaign (Table 4.2). The concentrations measured at the highway (Station 1) are comparable to the concentrations measured at H. C. Andersens Boulevard, where the daily traffic intensity is similar to the traffic intensity at the highway station. The concentrations measured at the background (Station 2) are comparable to the concentrations measured in urban background (H.C. Ørsteds Institute). The NO concentration at the background station (Station 2) are relatively high compared to the urban background, however, this reflects simply that the background station was placed only 150 m south of the highway.

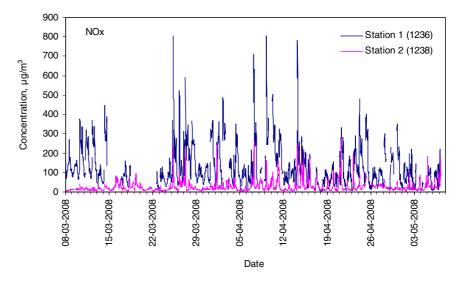


Figure 4.1 ${\rm NO}_{\scriptscriptstyle x}$ concentrations measured during the measurement campaign.

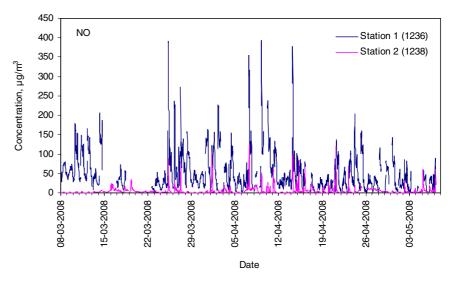


Figure 4.2 NO concentrations measured during the measurement campaign.

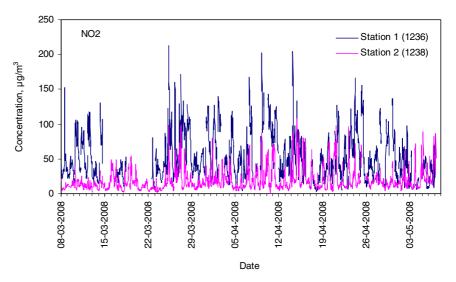


Figure 4.3 NO₂ concentrations measured during the measurement campaign.

Table 4.1 Average concentrations and 98 percentiles of NO_x , NO and NO_2 at Station 1 and 2. "Number" is the number of half hours with accepted data. The total number of half hours during the measurement campaign corresponds to 2928. Note that the NO_x concentration is calculated as NO_2 , and hence the sum of NO and NO_2 does not add up to NO_x

			NO _x as NO ₂	NO	NO ₂
Station 1	Number		2276	2276	2276
Otation	Average	μg/m³	131	50	53
	98 percentile	μg/m ³	390	178	135
Station 2	Number		2772	2772	2772
	Average	μg/m³	27	5	19
	98 percentile	μg/m³	114	32	72

Table 4.2 Average concentrations of NO_x , NO and NO_2 at selected monitoring stations in the Danish Air Quality Monitoring programme (Kemp et al., 2008) around Copenhagen during the period from 08-03-2008 to 07-05-08. Note that the NO_x concentration is calculated as NO_2 , and hence the sum of NO and NO_2 does not add up to NO_x .

Monitoring station	Type of station	NO _x as NO ₂	NO	NO ₂
		μg/m³	μg/m³	μg/m³
H.C. Andersens Boulevard, Copenhagen	Street	113	36	58
Jagtvej, Copenhagen	Street	92	30	46
H.C. Ørsteds institute, Copenhagen	Urban background	23	2	20
Lille Valby, Roskilde	Rural background	13	1	11

Figure 4.4 to 4.6 present the results from measurements of NO_x , NO, and NO_2 during periods with wind direction between 105° and 225°. This is the wind directions where the contribution from the highway traffic can be calculated by subtraction of the concentrations at Station 2 from the concentrations at Station 1. Table 4.3 presents the number of measurements, average concentration and the 98 percentile for Station 1 and for the contribution due to the highway traffic. 87%, 95% and 74% of the concentrations of NO_x , NO, and NO_2 at Station 1, respectively, originates from the highway traffic during periods with southerly wind.

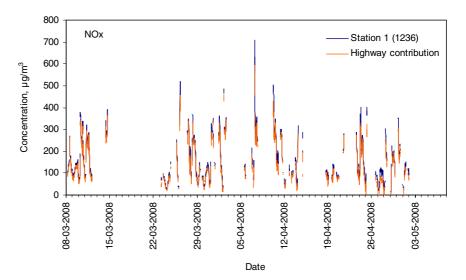


Figure 4.4 Total NO_x concentration at Station 1 and NO_x concentration due to highway traffic (highway contribution) at wind directions between 105° and 225°.

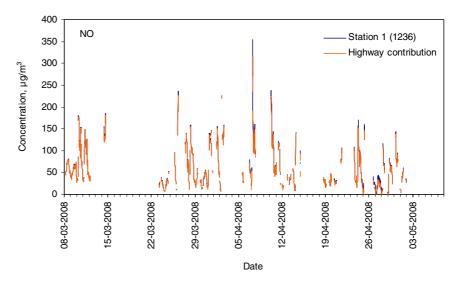


Figure 4.5 Total NO concentration at Station 1 and NO concentration due to highway traffic (highway contribution) at wind directions between 105° and 225°.

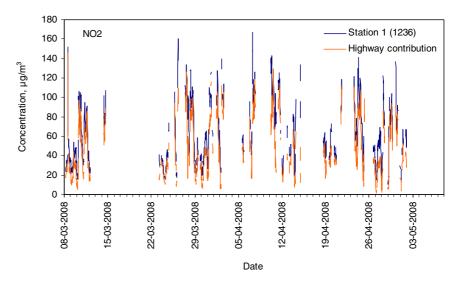


Figure 4.6 Total NO₂ concentration at Station 1 and NO₂ concentration due to highway traffic (highway contribution) at wind directions between 105° and 225°.

Table 4.3 Average concentrations and 98 percentiles of NO_x , NO and NO_2 at Station 1 and the contribution from the highway traffic to NO_x , NO and NO_2 at Station 1. Only data measured when the wind directions were between 105° and 225° . "Number" is the number of half hours with accepted data. The total number of half hours during the measurement campaign corresponds to 2928. Note that the NO_x concentration is calculated as NO_2 , and hence the sum of NO and NO_2 does not add up to NO_x .

			NO _x as NO ₂	NO	NO ₂
Station 1	Number		960	960	960
	Average	μg/m³	160	62	64
	98 percentile	μg/m³	410	186	137
Station 2	Number		960	960	960
	Average	μg/m³	139	60	48
		%	87	95	74
	98 percentile	μg/m³	345	178	109
		%	84	96	79

Figures 4.7 to 4.9 show the diurnal variations of the concentrations of NO_x NO, and NO₂ for the entire measurement campaign and at wind directions between 105° and 225°. The diurnal variations at Station 1 (highway) shows the typical pattern with very low concentrations during night, high concentrations during morning and afternoon rush hours and a little lower concentration at noon compared to rush hour. The diurnal variations at Station 2 (background) shows clearly a different pattern with at maximum concentration during early morning, lowest concentration during noon and afternoon, and a some what higher concentration during night time. The diurnal variations at Station 2 can be explained by a combination of influence of local sources and dilution due to increase of wind speed and mixing layer during the day.

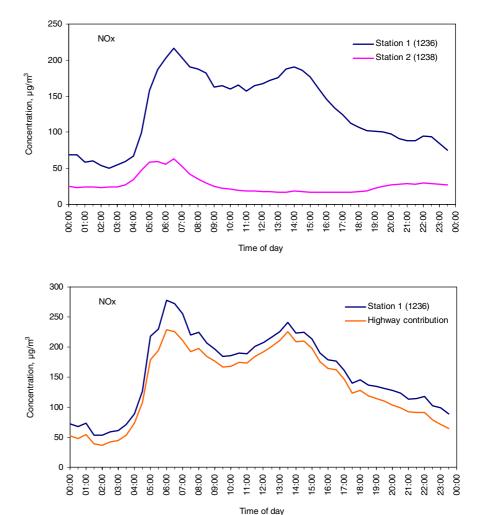


Figure 4.7 Average diurnal variation of NO_x concentration at Station 1 and 2 during the entire measurement campaign (top) and at wind directions between 105° and 225° (bottom).

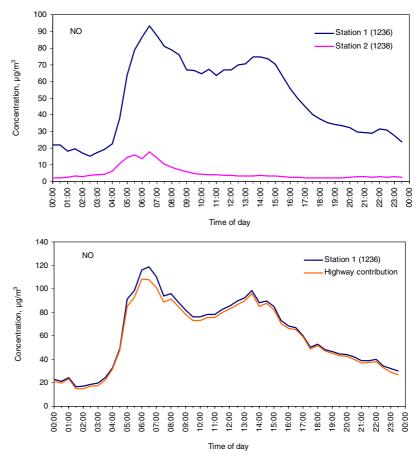


Figure 4.8 Average diurnal variation of NO concentration at Station 1 and 2 during the entire measurement campaign (top) and at wind directions between 105° and 225° (bottom).

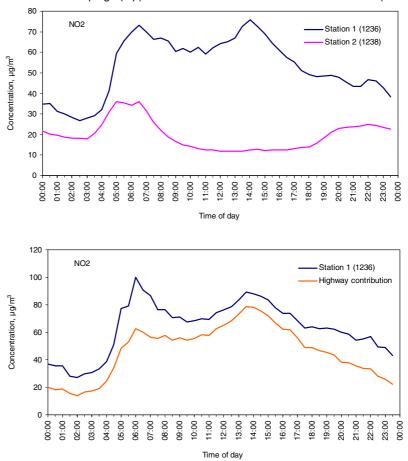


Figure 4.9 Average diurnal variation of NO_2 concentration at Station 1 and 2 during the entire measurement campaign (top) and at wind directions between 105° and 225° (bottom).

5 Ozone, O₃

The ozone concentration was measured at the background station (Station 2) in order to improve the evaluation of the NO_x measurements and for model calculations of NO_2 . Figure 5.1 presents the results of the ozone measurements at Station 2.

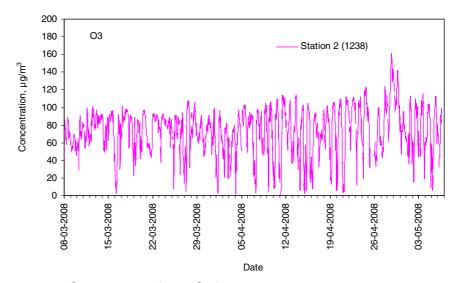


Figure 5.1 Ozone concentrations at Station 2.

6 $PM_{2.5}$ and PM_{10} , TEOM

Figure 6.1 and 6.2 presents the results from measurements of $PM_{2.5}$ and PM_{10} using TEOM during the measurement campaign. Note that the TEOM do not give the real $PM_{2.5}$ and PM_{10} due to evaporation of part of the sampled particle mass. Typically about 8-9 $\mu g/m^3$ is lost at Danish conditions (Kemp et al., 2005; Palmgren et al., 2003).

Table 6.1 presents the number of measurements, average particle mass and the 98 percentile. The average $PM_{2.5}$ is about 20% higher at the highway (Station 1) compared to the background (Station 2) while PM_{10} is about 60% higher at the highway compared to the background. This is as expected since it is well known that only a small fraction of $PM_{2.5}$ and PM_{10} originates from local sources. If the $PM_{2.5}$ and PM_{10} are corrected for the artefact due to the use of TEOM (see chapter 2) then the amount of PM originating from the highway traffic is even smaller; probably about 10% and 45% for $PM_{2.5}$ and PM_{10} , respectively (based on the assumption that about 9 $\mu g/m^3$ of particle mass evaporates during TEOM measurements of both $PM_{2.5}$ and PM_{10}).

The average $PM_{2.5}$ has been compared to results from TEOM measurements at selected monitoring stations around Copenhagen in the Danish Air Quality Monitoring programme (Kemp et al., 2008) for the period of the measurement campaign (Table 6.2). The $PM_{2.5}$ measured at the highway (Station 1) are smaller than the $PM_{2.5}$ measured at the street station at H. C. Andersens Boulevard, and about the same level as $PM_{2.5}$ measured at the urban background station at H. C. Ørsteds Institute. The concentrations measured at the background (Station 2) are comparable to the concentrations measured at the rural background station at Lille Valby, Roskilde.

Table 6.1 Average and 98 percentiles of TEOM $PM_{2.5}$ and PM_{10} at Station 1 and 2. "Number" is the number of half hours with accepted data. The total number of half hours during the measurement campaign corresponds to 2928.

			TEOM PM _{2.5}	TEOM PM ₁₀
Station 1	Number	·	2564	2497
	Average	μg/m³	11	22
	98 percentile	μg/m³	24	60
Station 2	Number	·	2546	2555
	Average	μg/m³	9	14
	98 percentile	μg/m³	22	36

Table 6.2 Average TEOM PM_{2.5} at selected monitoring stations in the Danish Air Quality Monitoring programme (Kemp et al., 2008) around Copenhagen during the period from 08-03-2008 to 07-05-08. The TEOM measurements at H.C. Ørsteds Institute and Lille Valby are carried out by NERI in a project for the Danish Environmental Protection Agency.

Monitoring station	Type of station	TEOM PM _{2.5} µg/m ³	
H.C. Andersens Boulevard, Copenhagen	Street	<u>μg/m</u> 15	
H.C. Ørsteds institute, Copenhagen	Urban background	13	
Lille Valby, Roskilde	Rural background	8	

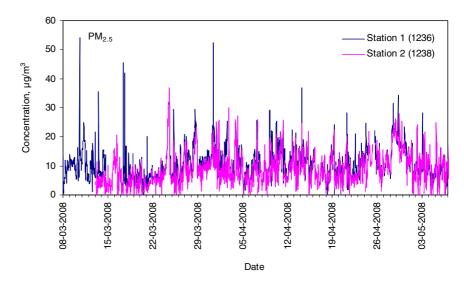


Figure 6.1 TEOM PM_{2.5} measured during the measurement campaign.

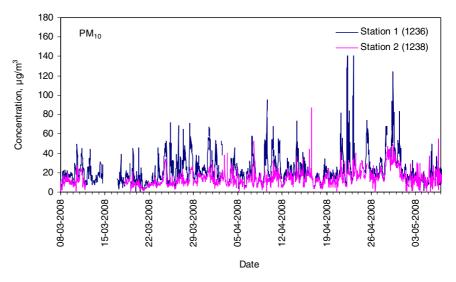


Figure 6.2 TEOM PM₁₀ measured during the measurement campaign.

Figures 6.3 and 6.4 present the results from measurement of TEOM PM_{2.5} and PM₁₀ during periods with wind direction between 105° and 225°. This is the wind directions where the contribution from the highway traffic can be calculated by subtraction of the concentrations at Station 2 from the concentrations at Station 1. Table 6.3 presents the number of measurements, average and 98 percentile for PM_{2.5} and PM₁₀ for Station 1 and for the contribution due to the highway traffic. Only 20% and 45% of TEOM PM_{2.5} and PM₁₀, respectively, originates from the highway traffic during periods with southerly wind. If TEOM PM_{2.5} and PM₁₀ are corrected for the loss of particle mass due to evaporation during the TEOM measurement, then the percentages will be even lower. The lost mass corresponds approximately to 9 μ g/m³ for both PM_{2.5} and PM₁₀ (Kemp et al., 2005). Based on this, the amount of PM_{2.5} and PM₁₀ originating from the highway traffic are estimated to only 12% and 34% for PM_{2.5} and PM₁₀, respectively.

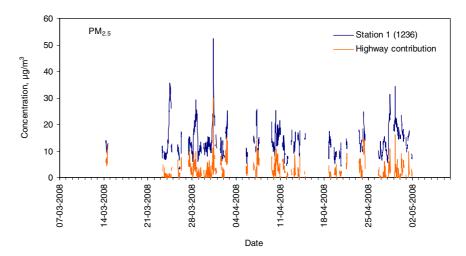


Figure 6.3. TEOM PM $_{2.5}$ at Station 1 and PM $_{2.5}$ due to highway traffic (highway contribution) at wind directions between 105° and 225°.

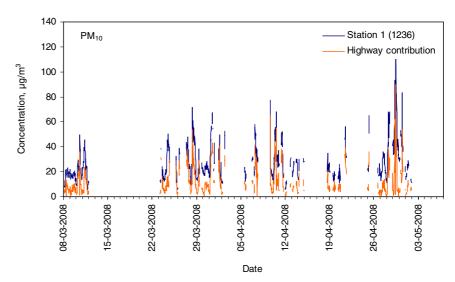


Figure 6.4 TEOM $PM_{2.5}$ at Station 1 and $PM_{2.5}$ due to highway traffic (highway contribution) at wind directions between 105° and 225° .

Table 6.3 Average and 98 percentiles of TEOM PM2.5 and PM10 at Station 1 and the contribution from the highway traffic to TEOM PM2.5 and PM10 at Station 1. Only data measured when the wind direction was between 105° and 225°. Number gives the number of half hours with accepted data. The total number of half hours during the measurement campaign corresponds to 2928.

			TEOM PM _{2.5}	TEOM PM ₁₀
Station 1	Number	·	800	935
	Average	μg/m³	14	27
	98 percentile	μg/m ³	28	65
Contribution		·	800	935
from highway	Average	μg/m³	3	12
		%	20	44
	98 percentile	μg/m³	11	41
		%	40	62

Figures 6.5 and 6.6 show the diurnal variations of TEOM $PM_{2.5}$ and PM_{10} for the entire measurement campaign and at wind directions between 105° and 225°. The diurnal variation at Station 1 (highway) shows higher

particle mass during the day compared to night time. However, the relative influence of rush hour traffic is less evident for particle mass compared to the nitrogen oxides. This is simply explained by the fact that the amount of long range transported particle mass is much higher than for the nitrogen oxides (NO and NO₂). The diurnal variation at Station 2 (background) shows a diurnal pattern that looks similar to the diurnal pattern for nitrogen oxides. The diurnal variation at Station 2 can be explained by a combination of influence of local sources and increase in dilution due to increase of wind speed and mixing layer during the day.

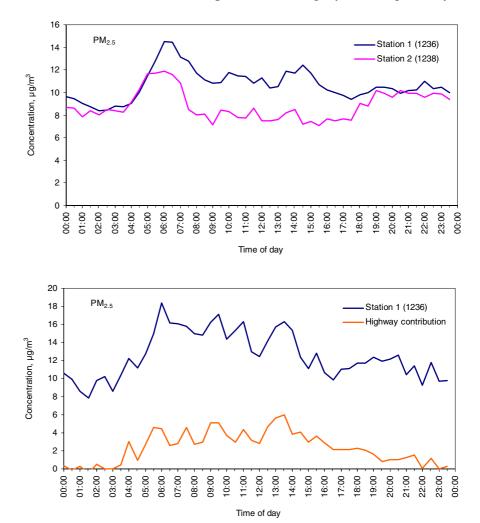


Figure 6.5 Average diurnal variation of TEOM PM_{2.5} at Station 1 and 2 during the entire measurement campaign (top) and at wind directions between 105° and 225° (bottom).

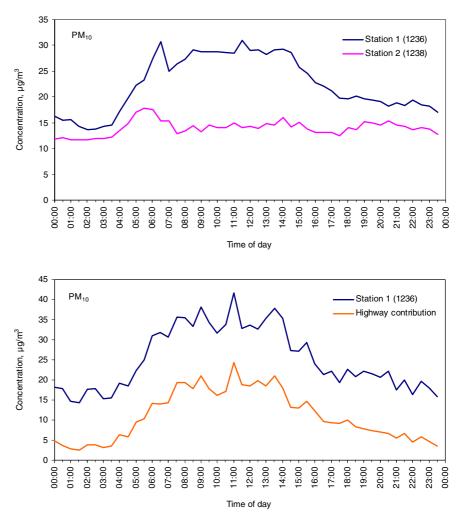


Figure 6.6 Average diurnal variation of TEOM PM_{10} at Station 1 and 2 during the entire measurement campaign (top) and at wind directions between 105° and 225° (bottom).

7 Particle number, volume and number size distribution

Figure 7.1 presents the results of the particle number concentration measurements obtained by the DMPS (sub micrometer particles, 10 – 700nm) at Station 1 and Station 2 during the entire measurement campaign. The figure shows, that the concentration at the highway station always was higher than at the background station. The mean concentration at the highway station amounts to about 23400 particles/cm³ in comparison to about 6300 particles/cm³ at the background site.

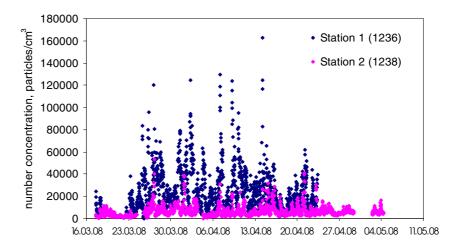


Figure 7.1 Total particle number concentration (size range: 10 – 700nm) at Station 1 and Station 2 as function of time during the entire measurement period.

The total particle volume concentrations during the entire experiment at both stations (Figure 7.2) shows only small differences in particle volume between the highway and the background site. Here, the total mean volume concentration at the highway station was about 9.7 $\mu m^3/cm^3$ (the volume of particles in μm^3 per cm³ of air) compared to about 7.1 $\mu m^3/cm^3$ at the background site. This large difference in particle count and small difference in particle volume concentration suggests that most of the difference in particle number between the two stations was caused by very small particles emitted from the highway traffic.

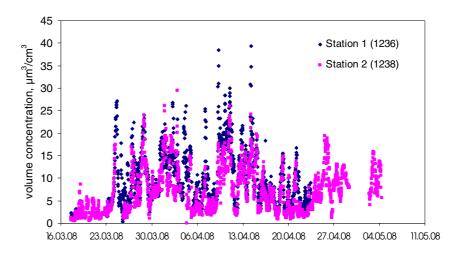


Figure 7.2 Total particle volume concentration (size range: 10 - 700nm) at Station 1 and Station 2 as function of time during the entire measurement period.

Table 7.1 Number of measurements, mean number of particles and particle volume concentrations for Station 1 and Station 2 during the entire experiment. Number gives the number of half hours with accepted data. The total number of half hours during the measurement campaign corresponds to 2928.

	Station 1 (1236)	Station 2 (1238)
Number of observations during the entire measurement period	1566	2046
Mean particle number concentration in particles/cm ³	23400	6300
Mean particle volume concentration in μm³/cm³	9.7	7.1

In Figure 7.3 the results for particle number concentrations obtained by DMPS measurements $(10-700\,\mathrm{nm})$ during periods with wind direction between 105° and 225° are illustrated. These are the wind directions where the contribution from the highway traffic was calculated by subtraction of the concentrations at Station 2 from the concentrations at Station 1. The highway contribution to the number of sub micrometer particles was very large (79%) compared to the number of particles generally detected at Station 1 (Table 7.2).

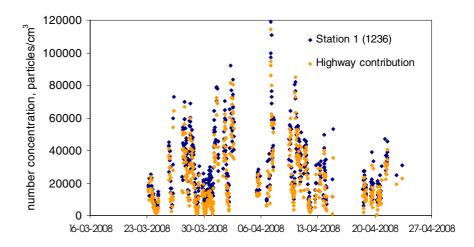


Figure 7.3 Total particle number concentration (size range: 10 – 700nm) at Station 1 and the highway contribution as function of time at southerly wind directions (105° and 225°).

A different picture is seen for the total particle volume concentrations of sub micrometer particles for south-westerly wind directions measured at the highway site (Figure 7.4). The highway contribution only amounts to about 28% in total particle volume at this station in the sub micrometer size range (Table 7.2). At Station 1 most of the particle volume was caused by particles generally present at Station 1 and hence the particle volume did not originate from the vehicles passing on the highway. This indicates that the particles originating from the highway were of small size.

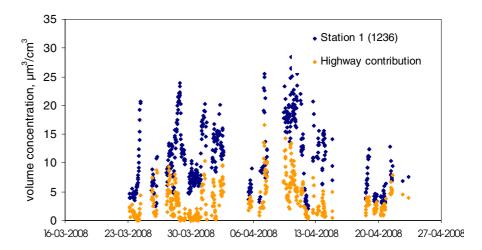


Figure 7.4 Total particle volume concentration (size range: 10-700nm) at Station 1 and the highway contribution as function of time at southerly wind directions (105° and 225°).

Table 7.2 Number of measurements, mean particle number and particle volume concentrations for Station 1 and for contribution due to highway traffic during southerly wind directions (105° and 225°). Number gives the number of half hours with accepted data. The total number of half hours during the measurement campaign corresponds to 2928.

	Station 1 (1236)	Highway contribution
Number of observations during south-westerly wind directions	568	568
Mean particle number concentration in particles/cm³	29200	23100
Mean particle volume concentration in μm³/cm³	11.1	3.1

In Figure 7.5 the particle number size distribution determined by the DMPS (10-700 nm) is illustrated for Station 1 and 2 for the entire measurement period. At the background station the data shows the maximum value in the range from 50-100 nm (the Aitken mode). At the highway station this peak is not so evident due to the large increase in particle number at the smaller particle sizes. At the highway station the maximum particle number is measured to about 45000 particles/cm³ at about 10 nm (detection limit of DMPS).

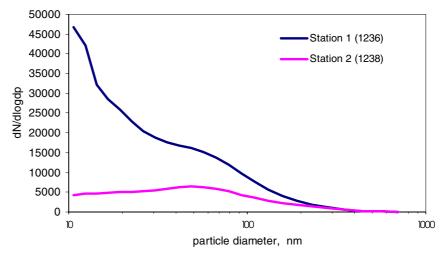


Figure 7.5 Mean particle number size distribution (10 - 700nm) for Station 1 and 2 for the entire measurement period.

In Figure 7.6 the particle number size distribution determined by the DMPS (10-700 nm) is shown for Station 1 and 2 together with the highway contribution to the particle number size distribution at Station 1. It is clearly shown, that the highway contribution in the range from 50-100 nm (the Aitken mode range) is about twice as high as at the background station. For the even smaller particles (below 50 nm) the difference is even larger. At about 10 nm the number concentrations of the particles is about 60000 particles/cm³ in comparison to only about 2000 particles/cm³ at the background site.

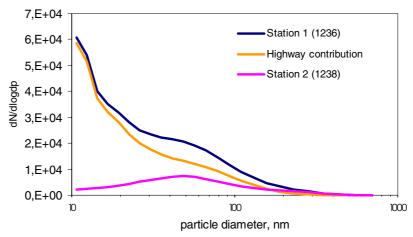


Figure 7.6 Mean particle number size distribution (10-700nm) at southerly wind directions $(105^{\circ}$ and $225^{\circ})$ for Station 1 and 2 and the highway contribution at Station 1.

8 Emission factors for $PM_{2.5}$ and PM_{10}

The total emission factors for $PM_{2.5}$ and PM_{10} due to the highway traffic can be determind from the ratio between the highway contribution to PM and the highway contribution to the concentration of NO_x multiplied by the emission factor for NO_x at the same conditions (see also Chapter 2). The average ratio between PM and NO_x concentration for $PM_{2.5}$ and PM_{10} can be determined from plots of $PM_{2.5}$ and PM_{10} as function of NO_x concentration (Figure 8.1 and 8.2).

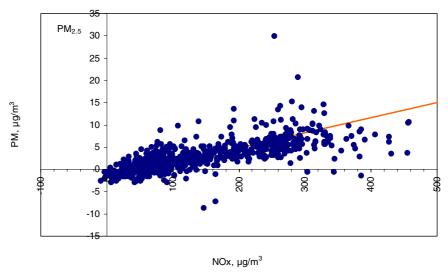


Figure 8.1 Highway contribution for TEOM $PM_{2.5}$ as function of highway contribution to NO_x concentrations at wind directions between 105° and 225°. The regression line is calculated by orthogonal regression analysis.

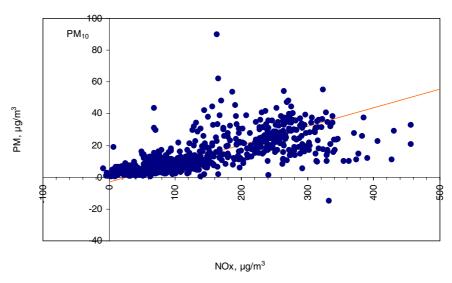


Figure 8.2 Highway contribution for TEOM PM_{10} as function of highway contribution to NO_x concentrations at wind directions between 105° and 225° . The regression line is calculated by orthogonal regression analysis.

From the diurnal variation of especially $PM_{2.5}$ (Figure 6.5) it can be seen that the contribution from the highway traffic to $PM_{2.5}$ is very small during night time. The analysis was therefore repeated without the night time results (nights = 22:00 to 3:30) (Figure 8.3 and 8.4).

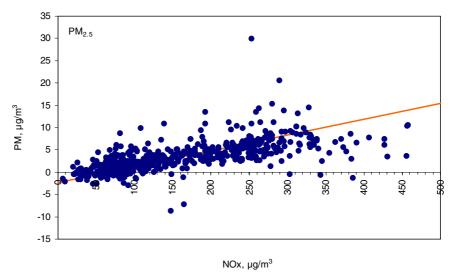


Figure 8.3 Highway contribution for TEOM $PM_{2.5}$ as function of highway contribution of NO_x concentrations at wind directions between 105° and 225° and during day time (4:00 to 21:30). The regression line is calculated by orthogonal regression analysis.

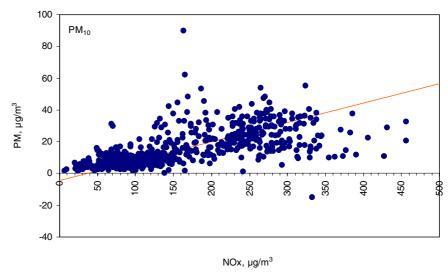


Figure 8.4 Highway contribution for TEOM PM_{10} as function of highway contribution of NO_x concentrations at wind directions between 105° and 225° and during day time (4:00 to 21:30). The regression line is calculated by orthogonal regression analysis.

Table 8.1 presents the slopes and intercepts calculated for the regression lines in Figure 8.1 to 8.4. The slopes and intercepts do not differ significantly with and without night time results. However, at least for $PM_{2.5}$ there has been a decrease in the scatter.

There is a small negative intercept in all four cases. However, the regression lines are calculated on the basis of measurements on four instruments (NO_x at station 1 and 2 and TEOM at station 1 and 2). Small calibration errors may therefore explain the intercept. Moreover, other local sources at the measurement sites may also interfere with the determina-

tion of the highway contribution and thereby explain the negative intercept.

The total emission factor for NO_x in 2003 has for Danish urban driving conditions been given to about 1.2 g/(vehicle km) (Ketzel et al., 2007). However based on the manual traffic counts at the Holbæk Highway 47.1 km east of Roskilde (see Figure 2.7) it has been possible to calculate a more specific emission factor for the conditions during the campaign measurements at the highway. The results from the manual traffic counts are shown in table 8.2 together with the resulting emission factors for NO_x . The emission factors were calculated with the emission module of WinOSPM that implements the European COPERT 4 (EEA, 2007). The used traffic composition is based on the manual counting of traffic, the Average Daily Traffic (ADT) was set to 57,000 vehicles per day and a standard diurnal variation of the traffic was assumed.

Based on the NO_x emission factors the total emission factor for $PM_{2.5}$ and PM_{10} can be calculated by multiplication of the slopes in table 8.1 with the emission factor of NO_x on 1.33 g/(vehicle km) (Table 8.2). This gives emission factors for $PM_{2.5}$ and PM_{10} to 45 mg/(vehicles km) and 155 mg/(vehicle km), respectively (table 8.1). This is comparable to emission factors previously determined for H.C. Andersens Boulevard ($PM_{2.5}$: 54 mg/(vehicles km); PM_{10} : 158 mg/(vehicles km)) and for Jagtvej (PM_{10} : 91 mg/(vehicles km) (Ketzel et al., 2007). The average daily traffic at H.C. Andersens Boulevard and Jagtvej is 65,000 and 30,000 vehicles per day, respectively.

Table 8.1 Slope and intercepts calculated for the linear orthogonal regression analysis in figure 8.1 to 8.4. Total emission factors for $PM_{2.5}$ and PM_{10} are based on a total emission factor for NO_x of 1.33 g/(vehicles km).

	Slope	Intercept µg/m³	Emission factor mg/(veh.km)
Diunal data			
PM _{2.5}	0,034	-1,93	45
PM ₁₀	0,117	-4,43	155
Only day time			
PM _{2.5}	0,036	-2,31	47
PM ₁₀	0,122	-4,43	162

Table 8.2 Distribution of vehicle types and the resulting emissions factors for NO_x . The distribution of vehicles type is based on manual count of traffic at the Holbæk Highway 47.1 km east of Roskilde carried out on the 3rd. April 2008. Traffic data were delivered by the Danish Road Directorate.

	All vehicles	Passenger car	Van 2-3.5 t	Truck <32 t	Truck >32 t	Bus	Motor cycle
Percentage of ADT, %	100	78.3	13.6	2.6	5.0	0.2	0.2
NO _x emission g/km	1.33	0.73	1.23	5.8	7.5	6.3	Not included

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MEASUREMENTS OF AIR POLLUTION FROM A DANISH HIGHWAY

This report presents the results from a measurement campaign carried out at the Holbæk High-way during 2008. The objective of the campaign was to determine the emission factors for $\rm PM_{2.5}$ and $\rm PM_{10}$ due to highway traffic. The campaign included measurements of $\rm NO_x$, NO, NO₂, TEOM $\rm PM_{2.5}$, TEOM $\rm PM_{10}$, O₃, particle size distribution and local meteorology. The emission factors for $\rm PM_{2.5}$ and $\rm PM_{10}$ were determined to 45 and 155 mg/(vehicle km), respectively. This is comparable to the emission factors previously determined for H. C. Andersens Boulevard in Copenhagen and somewhat higher than found at Jagtvej, Copenhagen.

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