ITM-report 110

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# Concentrations and sources of PM10 and PM2.5 in Sweden

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## Abstract

A network of Swedish sites, ranging from regional rural background, urban background to kerbside sites, measuring PM2.5 and PM10 on one hour basis has been operated over 2 years in Sweden. The results show two important features determining the concentrations, long distance transport of particles and re-suspension of road dust. Strong low-level inversions especially in the inland of Northern Sweden during winter lock out the long distant transport and lead to less dilution of the local emissions and induce at times very high concentrations.

Typical annual mean values for PM2.5 at rural background, urban background and kerb side sites are 5 – 10, 6 – 11 and 10 – 15 µg/m³, respectively, while for PM10 it is 7 – 13, 12 – 18 and 20 – 30. The ratio PM2.5/ PM10 is about 0.8, 0.6 – 0.7 and 0.4 – 0.6 at the rural background, urban background and kerb side sites respectively. The strongly decreasing ratio implies the strength of road dust emissions and other mechanical wearing processes giving coarse particle emissions.

Using NOx as a tracer for traffic emission and assuming a fixed relation to NOx for the particle exhaust emissions the non-exhaust emission factors for PM2.5 and PM10 is estimated to 25 and 200 mg/vehkm compared to the estimated 23 mg/vehkm for exhaust related particle emissions. The ranges observed for the non-exhaust emission factors for PM2.5 and PM10 are 0 – 76 and 25 – 554 mg/vehkm, respectively.

Totally the non-exhaust emission factor for PM10 is 9 times larger than the PM10 exhaust emission factor but for PM2.5 the non-exhaust emission factor is equal to the exhaust emission factor.

The high correlation between PM10 and PM2.5 found in this study are explained in this study and possibly others too by the emission of road dust and other coarse particle sources are closely linked to traffic intensity and thus primary exhaust, together totally dominating PM10. In background situations PM2.5 is as such dominating PM10 and thus explaining the correlation.

Road dust emerges as the major local source responsible for exceedance of limit values in Sweden. However it is superimposed on strongly enhanced background concentrations due to long distance transport.

It is obvious from this study that PM2.5 has a strong component of non-exhaust coarse particle sources. PM2.5 thus do not reflect the correct concentrations due to combustion sources and long range transported emissions. Using PM2.5 will complicate the assessment of sources and the choice of abatement strategy. Use of PM1 would avoid these problems.

## Introduction

Ambient particles are today of great concern as a strong link with mortality rate, cardiovascular and respiratory diseases have been demonstrated. The particles have become the major focus as particle mass has been used as indicator of the presence and a quantitative measure of the concentration of air pollution in the epidemiological studies showing health effects connected to elevated particle mass concentrations. Künzli et al., 2000, concluded that about 40 000 persons in Europe annually die due to air pollution and more suffer other serious health problems.

Some epidemiological studies suggest that the mass of fine particles less than 2.5 mm in diameter (PM2.5) has a stronger relation to observed health effects than the particle fraction (PM10) including coarse particle has (Dockery et al, 1994). This caused a strong concern to establish a new measure for air pollution control focusing on limiting the fine particle emissions. PM2.5 is already established in the USA as a standard air pollution indicator mostly due to technical and traditional reasons. PM2.5 has been taken over for European use and the process of developing a standard is well under way. However there has not been any critical review concerning its suitability from health or control strategy point of view.

It is important to emphasize that in critical assessment reports it is clearly shown that there are several potentially harmful components in an air pollution event and so far no specific characteristic or type of particles has been shown to be more or less harmful (Pershagen et al., 2000, US EPA, 1996).

In Switzerland, Gehrig et at al. (2003) report PM10 and PM2.5 concentrations from seven sites ( two rural, four that can be classified as urban background and one kerb side) that range from 19 to 33 and 15 to 24 µg/m³ respectively, excluding one high altitude rural station where the PM10 and PM2.5 concentrations were 11 and 8 µg/m³. The results are based on measurements from 1998 to 2001.

In another Swiss study, Hügelin et al. (2000) studied concentrations and sources of particles in different urban environments. Using 24 hours filter samples, extensive chemical analysis and source receptor modeling they found PM2.5 and PM10 concentrations ranging 8 to 22 and 10 to 44 mg/m3 respectively and attributed 33 - 63% of PM10 and 26 - 37% of PM2.5 to traffic. An other major sources were industry, bio-mass burning and secondary aerosols that might be interpreted as long-distance transport comprise 32 – 52 % of PM10, with the lower fractions at kerb side sites. The traffic sources were dominated by the tail pipe emissions, while the re-suspension of dust including road salt and tire wear comprised usually a third to half the traffic contribution to PM10.

In United Kingdom, AEA reports PM10 concentrations in the range 25 – 45 µg/m³ at kerb/road side sites, 17 – 30 µg/m3 at urban background sites and 12 – 20 µg/m³ at remote and rural sites during 1999 and 2000 (AEA, 2002).

In an investigation on mainly background sites in the UK Turnbull and Harrison , 2000, show that secondary particles contribute 28-35% of site-mean PM10, primary combustion particles from 20 to 57%, sodium chloride 11-34% and “other”, mainly crustal particles, 3-21%. The urban site has the higher contribution of primary combustion and crustal particles. However the authors feel the analysis underestimates the crustal contribution. In another study focused on urban sites Harrison et al., 2001, find the resuspension to be vehicle induced with approximately equal source strength as the exhaust emissions, which is clearly indicated in lower PM2.5/PM10 ratios at urban sites.

EMEP reports rural PM10 measurements mainly from Switzerland, Netherlands, Germany, and Spain (Lazaridis et al., 2000 a). The Swiss sites range 14 – 26 µg/m³, average 21 µg/m³ excluding the high alpine site Jungfraujoch. The German sites range from 13 to 26 µg/m³, average 24 µg/m³. In Spain the levels were between 20 and 44 µg/m³, average 30 µg/m³ during the last years. As PM10 measurement just recently has been appointed a standard, measurements are not always performed in a comparable way.

A first systematic collection of annual mean concentrations levels of PM2.5 and PM10 in Central and Eastern Europe was performed within the CESAR study (Houthuijs et al., 2001). 25 sites of urban and urban background character in Bulgaria, Czech Republic, Hungary, Poland, Romania and Slovak Republic showed to have fairly similar PM10 values, 41 to 98 mg/m3, with PM2.5 ranging 29 to 68 mg/m3 and PM10-PM2.5 ranging 12 to 40 mg/m3. The seasonal variation in PM10 concentration was mainly driven by PM2.5 increasing during heating season leading the authors to conclude the variation to depend on local heating.

EMEP has summarized PM10 data from the AIRBASE air quality database managed by the European Topic Centre on Air and Climate Change (Lazaridis et al., 2000 b). Air quality data from a large number of monitoring stations in various networks is stored in AIRBASE. Rural annual national averages range from 13 to 39 µg/m³, while at urban and street sites the national averages range from 18 to 40 µg/m3 and 18 to 55 µg/m³, respectively.

In major European cities road traffic is the dominating source of particles. Most vehicle exhaust particles are less than 100 nm but vehicles also contribute to emission of coarse particles due to the re-suspension of particulate matter deposited on the road surface.

In residential areas of North European cities, biomass burning for domestic heating may be an important source of fine particles during winter. For instance in Sweden, wood is most frequently burned in boilers constructed for multiple energy sources (oil, wood and electricity). Recently, low emission boilers have been introduced into the market, but stoves have come into use as an additional heating device, commonly also in urban areas. Emissions from wood combustion may be important for particulate matter, but also for many particle-bound hydrocarbons, such as polycyclic aromatic hydrocarbons. The aerosol in biomass combustion may substantially differ from that originating from vehicle exhausts, both regarding chemical composition and particle size distribution. For instance, PM from biomass burning may contain a larger fraction of hygroscopic particles (Hedberg et al., 2002).

Below the particle mass concentrations PM10 and PM2.5 are presented for various environments from the south to the north of Sweden, covering rural background, urban background and kerb site measurements performed during 2 years. Using high time resolution and simultaneous PM10 and PM2.5 data the influence of long-range transport and different local sources are quantified. Influencing geographical and meteorological conditions are discussed. It should be noted that when re-suspension is mentioned it includes also other particle sources producing particles due to wear of road, tires and brakes.

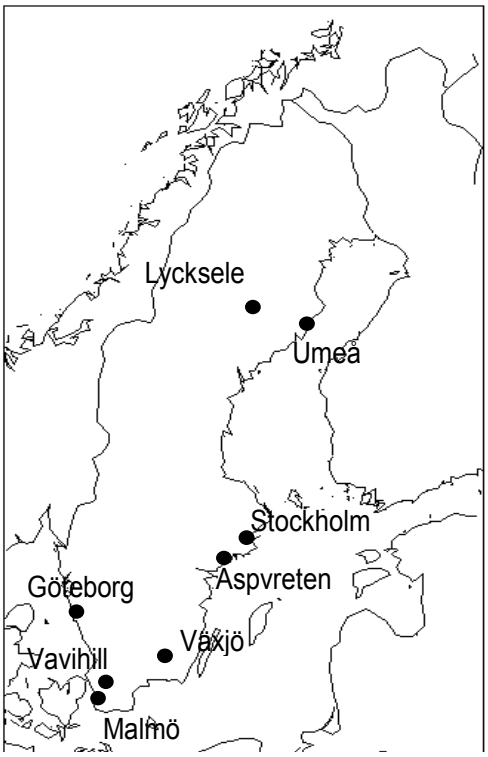
## Methods

### Measurement sites

PM10 and PM2.5 were measured at 14 measurement sites; see Table 1, during the period September 1, 1999 to August 31, 2001.

The measurement sites were selected to be representative for the major cities in Sweden, for areas in cities where bio mass combustion is one of the major domestic heating methods and for background, rural, areas with no local sources. The sites were also selected to give a good geographical coverage of Sweden, see Figure 1.

Vavihill is a rural background site situated in Southern Sweden about 20 km east of Helsingborg and about 60 km north Malmö. Aspvreten a rural background site situated 2 km west the Baltic coast and about 80 km southwest of Stockholm. In the major cities the measurements were made at one site representative for the urban background and one or two sites at the kerb side of major roads. The urban background sites were in general placed at roof level in the city centers.




*Figur 1. Location of measurement sites*

### Measurement methods

Continuous measurements of PM10 and PM2.5 were made by the Rupprecht and Patashnick (R&P) Tapered Element Oscillating Microbalance (TEOM®), instruments (Patashnick et al., 1991). The data was stored as one-hour averages.

The TEOM monitors used in this study were all equipped with a specially constructed inlet, which alternatively sample the PM10 or the PM2.5 fraction. Every 15 minutes the inlet changes from sampling of one fraction to the other. The change between fractions introduces a disturbance in the mass readings during two to three minutes. These readings have to be removed before calculation of the hourly averages. Each hourly PM10 and PM2.5 average is thus based on about 25 minutes of data. Based on the manufacturers data on the precision of the original TEOM monitor, the precision of the hourly PM10 and PM2.5 averages obtained with the special dual inlet is about 5 µg/m³.

Sampling of the PM10 fraction was made with the R&P US version PM10 inlet. The inlet has been shown to meet the US EPA criteria for PM10 inlets (VanOsdell, 1991). PM2.5 was sampled with the classical R&P PM2.5 cyclone (of a design also denoted as URG cyclone). The cut-off characteristic of this cyclone is not very sharp, which gives an overlap between the fine and coarse fraction of the airborne particles. This means that PM2.5 might be influenced by particles larger than 2.5 µm, especially when the coarse particle mass fraction is large compared with the PM2.5.

Many studies have shown that the TEOM technique may give lower concentrations than the gravimetric methods that are (or are proposed as) reference methods for PM10 and PM2.5 in USA and the European Union (Allen et al, 1997, King et al., 2000). The reason is that the sample air is heated in the TEOM method. This heating leads to evaporation of particle bound water, which is desired, but might also lead to undesired losses of volatile compounds, i.e. ammonium nitrate and some organic compounds.

In order to harmonize PM10 data measured in Europe an EU Member States Working Group has given recommendations on how to establish a consistent relationship between automated instruments

and the reference method (EC, 2001). As an interim solution for Member States who not have established the relationship, a default factor of 1,3 for the correction of results from TEOM and other automated instruments was recommended.

A comparison of the TEOM method and a gravimetric filter method performed at one rural and one urban station in Sweden indicates that the factor 1,3 might be to high, especially at PM concentrations below about 15 – 20 µg/m³ (Ferm and Hansson, 2003). However, as no 'Swedish' correction factor has been established yet, the EU default factor has been applied for PM10 as well as for PM2.5 in this report.

| *Measurement site* | *Site type* | *Remark* |
|---|---|---|
| Lycksele | Urban background | Influenced by traffic and bio-mass burning |
| Umeå | | |
| Storgatan-E4 | Kerb side | |
| Town Library | Urban background | At roof level in city center |
| Stockholm | | |
| Hornsgatan | Kerb side | Busy street canyon |
| Rosenlund | Urban background | At roof level in city center |
| Aspvreten | Rural background | 80 km southwest Stockholm |
| Vavihill | Rural background | 60 km north Malmö |
| Göteborg | | |
| Gårda-E6 | Kerb side | |
| Järntorget | Kerb side | |
| Femman | Urban background | At roof level in city center |
| Växjö | Urban background | In residential area with bio-mass burning |
| Malmö | | |
| Lernacken | Urban background | Near the Öresound bridge on the outskirts of Malmö |
| Town Hall | Urban background | At roof level in city center |
| Mobile station | Kerb side | Four placements at streets with various traffic density |

*Table 1. Overview of measurement sites*

## Results

### Levels of PM10 and PM2.5

The 24-hour average PM10 levels are about 16 µg/m³ at the rural sites in southern Sweden. The urban background concentrations of PM10 range from 17 to 23 µg/m³, in southern Sweden and 15 to 17 in northern Sweden, see Table 2. The 98th percentile of the daily mean values are in the range 35 to 53 µg/m³ at all rural and urban background sites, but Lycksele, the North Sweden inland site, where the PM10 concentrations occasionally can be higher. At Lycksele, the 98th percentile is 67 µg/m³.

The PM10 concentrations are substantially higher at the kerb side sites. The mean concentrations range from 23 to 50 µg/m³ and the 98th percentile from 85 to 185 µg/m³. However, the 98th percentile at the kerb side station in Malmö, in the far south is substantially lower, 52 µg/m³.

The PM2.5 concentrations are similar at all sites. The average concentrations are 8 – 15 µg/m³ at the rural and urban background sites and only slightly higher, 13 –18 µg/m³ at the kerb side sites. The 98th percentiles are also in the same range, 27 – 40 µg/m³, irrespective of site category. However, at the urban background site in Umeå in the north of Sweden the 98th percentile is lower, 18 µg/m³.

| | N | Median | Mean | Percentile 90 | Percentile 98 | Maximum |
|---|---|---|---|---|---|---|
| **PM10, µg/m³** | | | | | | |
| Rural | | | | | | |
| Aspvreten | 615 | 15 | 16 | 23 | 35 | 55 |
| Vavihill | 455 | 15 | 16 | 26 | 45 | 56 |
| Urban background | | | | | | |
| Lycksele | 523 | 13 | 17 | 30 | 67 | 130 |
| Umeå, Town Library | 655 | 12 | 15 | 24 | 40 | 74 |
| Stockholm, Rosenlund | 703 | 16 | 19 | 31 | 43 | 62 |
| Göteborg, Femman | 472 | 18 | 20 | 34 | 49 | 71 |
| Växjö | 566 | 15 | 17 | 29 | 47 | 84 |
| Malmö, Lernacken | 630 | 18 | 20 | 30 | 47 | 59 |
| Malmö, Town Hall | 647 | 21 | 23 | 35 | 53 | 65 |
| Kerb side | | | | | | |
| Umeå, Storgatan | 303 | 21 | 25 | 40 | 85 | 148 |
| Stockholm, Hornsgatan | 722 | 35 | 51 | 116 | 183 | 239 |
| Göteborg, Gårda | 497 | 27 | 34 | 58 | 104 | 195 |
| Göteborg, Järntorget | 177 | 32 | 36 | 56 | 86 | 108 |
| Malmö, Mobile stn | 658 | 21 | 23 | 34 | 52 | 62 |
| **PM2.5, µg/m³** | | | | | | |
| Rural | | | | | | |
| Aspvreten | 628 | 11 | 12 | 18 | 27 | 42 |
| Vavihill | 450 | 11 | 12 | 20 | 38 | 46 |
| Urban background | | | | | | |
| Lycksele | 535 | 10 | 12 | 19 | 33 | 67 |
| Umeå, Town Library | 656 | 8 | 8 | 13 | 18 | 26 |
| Stockholm, Rosenlund | 663 | 11 | 12 | 19 | 29 | 47 |
| Göteborg, Femman | 406 | 10 | 12 | 21 | 34 | 43 |
| Växjö | 572 | 10 | 12 | 21 | 34 | 59 |
| Malmö, Lernacken | 625 | 11 | 13 | 23 | 37 | 52 |
| Malmö, Town Hall | 567 | 13 | 15 | 25 | 40 | 53 |
| Kerb side | | | | | | |
| Umeå, Storgatan | 304 | 13 | 13 | 22 | 27 | 30 |
| Stockholm, Hornsgatan | 722 | 17 | 18 | 28 | 39 | 61 |
| Göteborg, Gårda | 497 | 15 | 17 | 25 | 38 | 46 |
| Göteborg, Järntorget | 173 | 15 | 16 | 23 | 33 | 39 |
| Malmö, Mobile stn | 657 | 12 | 14 | 23 | 39 | 49 |

*Table 2. . Statistical summary of 24-hour average concentrations of PM10 and PM2.5 at fourteen sites in Sweden September 1, 1999 - August 31, 2001. N = Number of days with data (i.e. 18 or more validated one hour averages).*

The fine particle mass, PM2.5, constitutes a large share, about 80%, of PM10 at the rural sites (see Figure 2). There is a general trend that the ratios PM2.5/PM10 are lower at the urban background sites (0.6 – 0.7) than at the rural sites and even lower (0.6 – 0.4) at the kerb side sites. Local sources of coarse particles (PM10-PM2.5) are thus more important than local sources of fine particles (PM2.5).

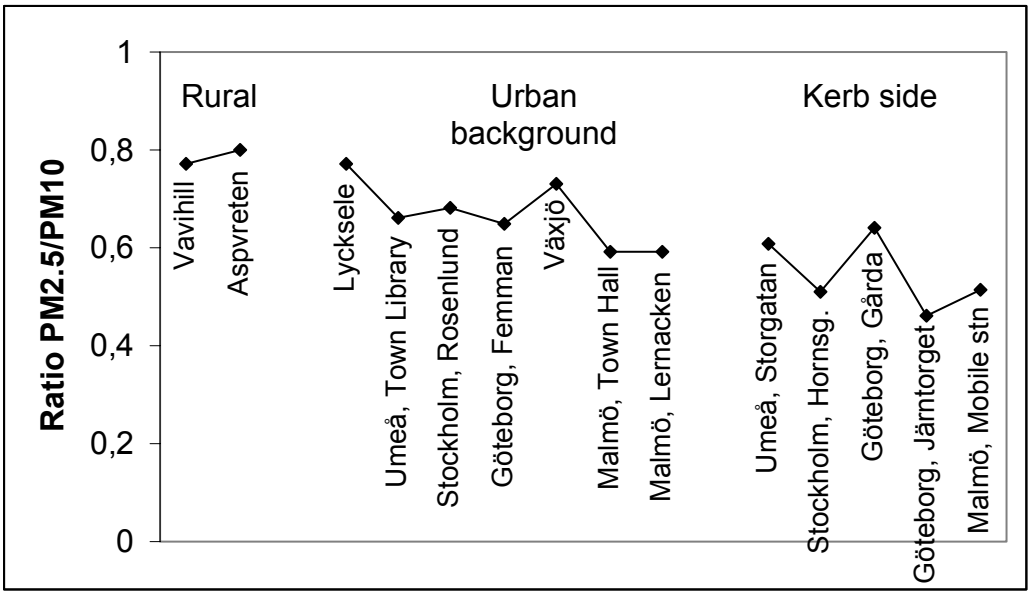



*Figure 2. Average ratio PM2.5/PM10 at 14 Swedish sites*

### Diurnal variation of PM10 and PM2.5

PM10

The mean diurnal variation of PM10 is strong at the kerb side sites, especially during weekdays, see Figure 3. The concentrations are about 2.5 times higher at noon (40µg/m³) than in the early morning (15 µg/m³). During holidays, the daily maximum is about two times the minimum. The concentrations of PM10 raises rapidly during the weekday mornings at the kerb side sites, but considerably slower during holidays. This reflects the difference in traffic rhythm between weekdays and holidays. It should be noted that this is the mean diurnal variation for the whole measurement period.

The diurnal variation at the urban background sites shows a similar pattern as at the kerb side, but the daily maximum hourly mean concentration (22 µg/m³) is only about 1.5 times the minimum (15 µg/m³) during weekdays and about 1.4 times during holidays.

A slight diurnal variation can also be seen at the rural sites. The daytime maximum (17 µg/m³) is about 10% higher compared to the minimum (15 µg/m³). This could be due to stronger winds during daytime, especially in spring and summer, caused by suspension of soil dust. Long-range transport even for larger particle is stronger especially during spring due to strong winds and low precipitation.

As the minimums at the urban background and the rural sites are very similar, one can assume that the minimum at the urban background sites represent air not influenced by any local sources. Based on this, the contribution from local sources during the rest of the day can be calculated. The local sources increases PM10 at the kerb side sites with about 22 µg/m³ and 14 µg/m³ during weekdays and holidays respectively. In the urban background, the local sources increases PM10 with 5 and 3 µg/m³ during weekdays and holidays respectively.

PM2.5

The diurnal variation of PM2.5 is much less pronounced than for PM10. A significant variation can only be seen at the kerb side sites during weekdays. The maximum (about 20 µg/m³), recorded at 8:00 – 9:00, is about 70% higher than the daily minimum. Smaller diurnal variation; the maximums being about 25% higher than the minimum, can also be seen during weekdays and holidays in the urban background and the kerb side sites.

If one assumes, as above, that the early morning PM2.5 concentrations in the urban background not is influenced by any local source, the local sources during the rest of the day increases the average concentrations of PM2.5 with 6 and 4 µg/m³ at the kerb side during weekdays and holidays respectively. The increase at the urban background sites is about 2 and 1.5 µg/m³ during weekdays and

holidays respectively. The increase at the urban background sites is about 2 and 1.5 µg/m³ during weekdays and holidays respectively.

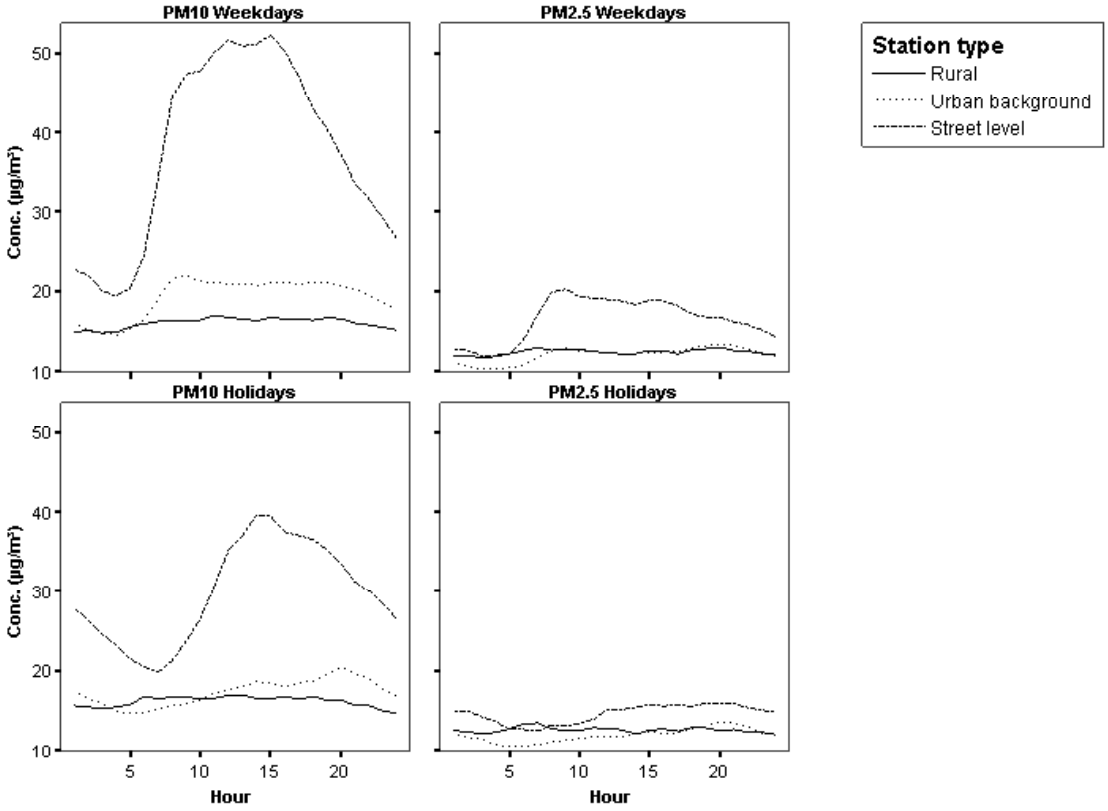



*Figure 3. Diurnal variation of PM10 and PM2.5 during weekdays and holidays at rural, urban background and kerb side sites.*

## Seasonal variation

The data on the diurnal variation presented above represents an average for the whole measurement period. However, the differences between seasons is substantial, see Figure 4.

The difference between the daytime concentrations of PM10 (average concentration at 13:00 – 14:00) and the minimum concentrations in the early morning (average concentration at 04:00 – 05:00) is 50 – 60 µg/m³ in February – May, but only about 10 in July to September at the kerb side sites.

The reason for the high PM10 levels found at the kerb side sites during spring is re-suspension of dust that has collected on the road surfaces during the strew of grit and the wear of studded tires in winter. The re-suspension is most effective when the roads have dried during spring and before the sand has been removed. It should be noted that the seasonal variation not could be observed at the kerb side station in Malmö, in the far south of Sweden. The reason might be a less frequent use of studded tires as well less sanding and salting.

The seasonal variation is much less pronounced or absent at all other site types and for both PM10 and PM2.5. A similar variation, but much less, as for PM10 at the kerb side, can be seen for PM10 at the urban background sites and for PM2.5 at the kerb side sites. It is likely that the somewhat higher PM levels during daytime at these sites in spring also are due to the re-suspension, as at least 1) some re-suspended dust reaches the urban background and 2) some of the re-suspended particles are small enough to be sampled as PM2.5.

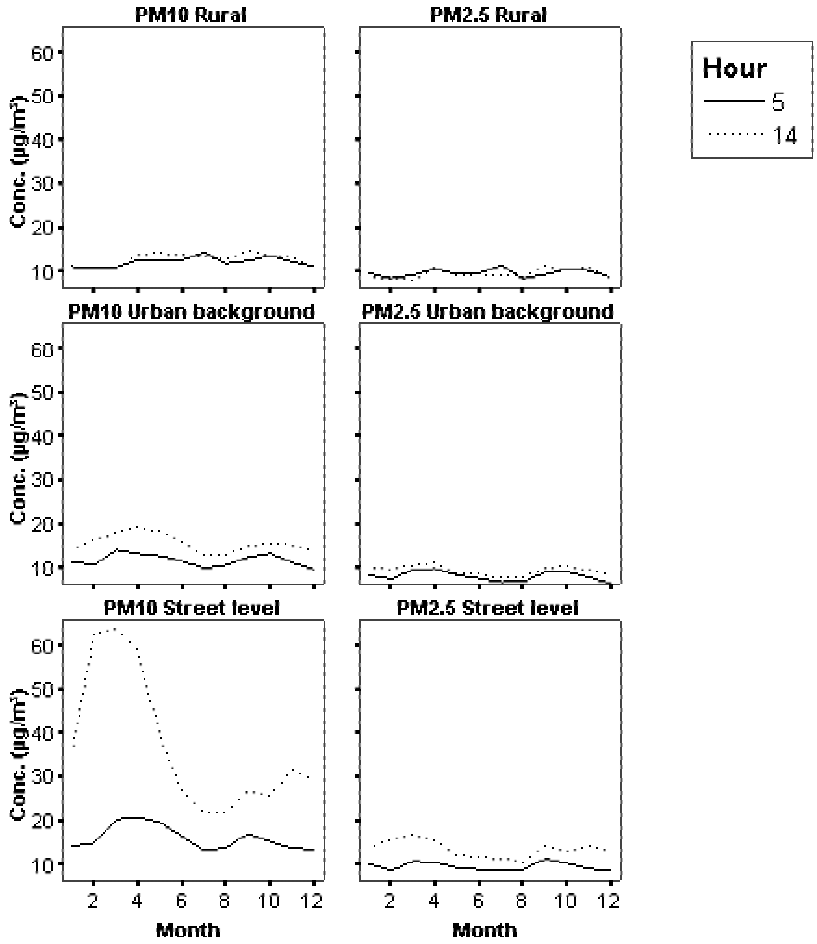



*Figure 4. Average concentrations of PM10 and PM2.5 at kerbside, urban background and rural sites at hour 5 (04:00 - 05:00) and hour 14 (13:00 - 14:00) of the day each month of the year*

## Long range transport

Sweden being situated down-stream of the large emission areas on the continent, with a fairly strong advection most of the year especially for southern Sweden dilutes the emissions from the local sources but on the other hand brings high concentrations from far away sources. The life-time of particles is estimated to 2 to 8 days, dependent on size and chemistry. Considering general wind speeds the sources on the continent is clearly within reach, which is clearly shown concerning sulfates, one of the major inorganic components in the particles.

As shown above the mean levels of PM10 and PM2.5 at the rural sites and in the urban background are about the same. This indicates that the levels are strongly influenced by long range transport. The kerb side sites are also strongly influenced during the periods of the year when re-suspension is relatively low. The strong dominance of the long range transport can be illustrated by the close temporal variation of the PM levels at different sites far apart in southern Sweden. An example showing the 24-hour averages of PM2.5 concentrations at three urban background sites (Malmö, Växjö and Göteborg) and one rural site (Vavihill) is seen in Figure 5.

The episode June 20 is evident at all sites. The correlation between PM2.5 at the different sites is also good during the rest of the month. The episodes around the 10th and 20th were both caused by polluted air originating in Central Europe.

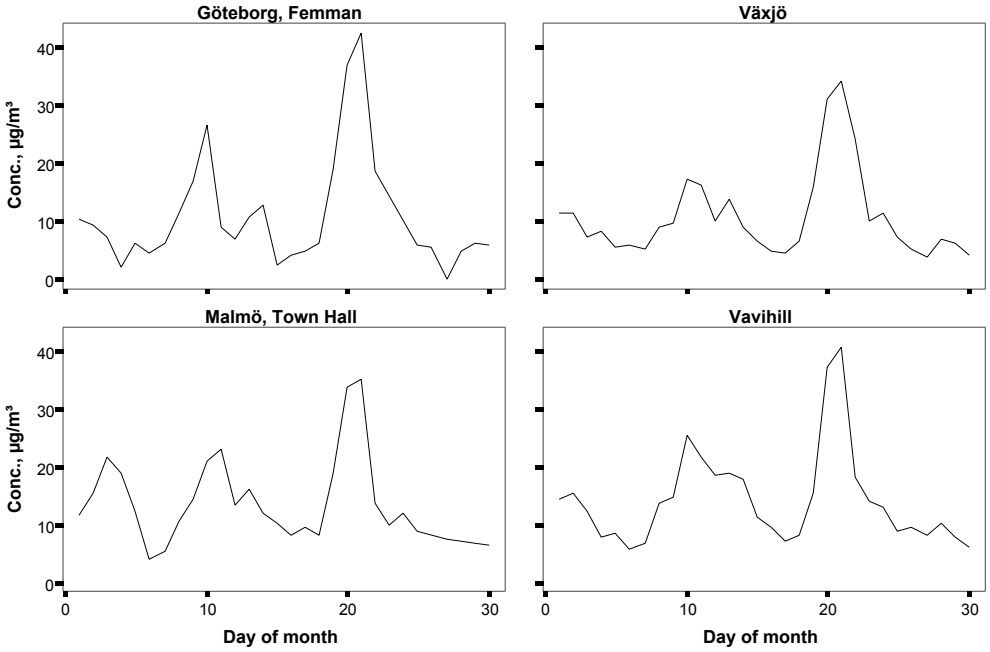



*Figure 5. 24-hour averages of PM2.5 at some sites in south Sweden June 2000*

As data are stored as hourly averages, it is possible to study episodes in detail. The PM2.5 concentrations obtained at three sites; one rural, one urban background and one kerb side site; in the Stockholm region in the end of October 1999 are depicted in Figure 6. The general resemblance of the time-sequence plots of PM2.5 at the three sites is very good. One can e.g. note that the rapid rise at the rural site at noon the 24th is repeated at the Stockholm sites about 12 hours later. This is consistent with a polluted air mass coming from the south first reaching the rural site and then the urban site 12 hours later. The episode the 31st is also conspicuous at all sites. However, at a few occasions (e.g. the 26th and 29th) local episodes are added to the general pattern. The local influence is most evident at the kerb side site, but can also be traced in the urban background. However, it is evident that the PM2.5 variation mainly is governed by changes in the long range transported air masses at all sites during this period.

The correlation between PM2.5 in the rural and urban background is in general very good. This is clear from Figure 7, where the simultaneously recorded PM2.5 24-hour average concentrations at the Stockholm urban background site, Rosenlund, and the rural site, Aspvreten, about 80 km south of Stockholm is plotted against each other.

The cloud of points is gathered around the 1:1 line, except for one point. That point represents PM2.5 concentrations recorded January 1, 2000. The reason for the high PM2.5 levels in the urban background is particles formed during the fireworks at the very early morning on the first day of the new millennium. It is interesting to note that this very special particle source is the only local source inside Stockholm during the study period that increased PM2.5 in the Stockholm background to levels substantially above the levels in the rural areas around Stockholm.

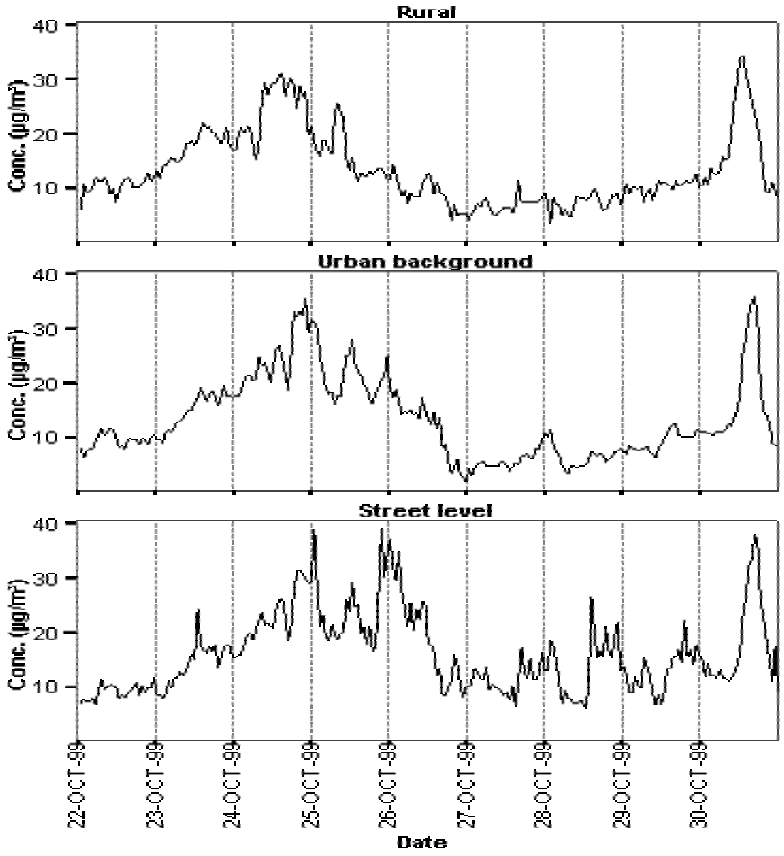



*Figure 6. Hourly averages of PM2.5 at one rural site (Aspvreten), one urban background site (Rosenlund) and one kerb side site (Hornsgatan) in the Stockholm region October 22 -31, 1999*

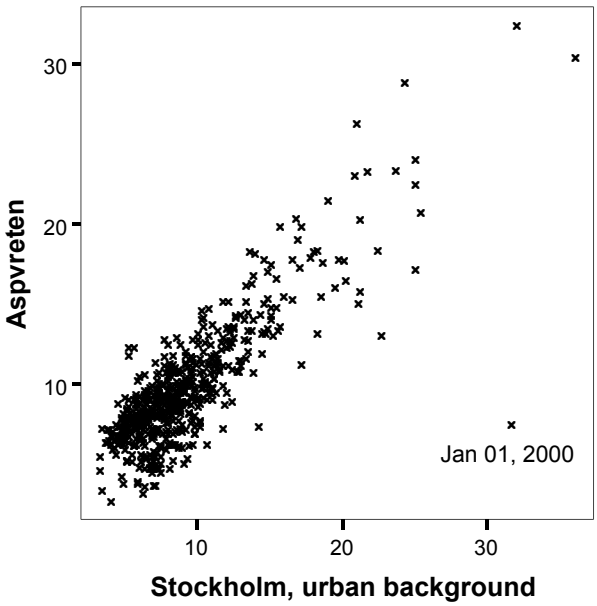



*Figure 7. Scatter plot of 24-hour PM2.5 averages at the Stockholm urban background site, Rosenlund, and the rural site Aspvreten*

## Local episodes

Two types of local episodes were observed during the study:

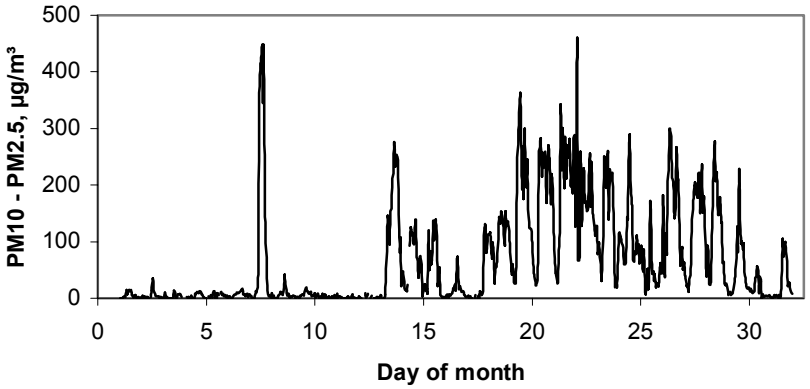
- "Resuspension episodes" where high levels of coarse particles occur at the kerb side sites due to re-suspension of road dust
- "Inversion episodes" where high levels of PM10 and PM2.5 occur mainly in the northern Sweden inland due to a prevailing strong local inversion in combination with relatively high emissions of PM.

### Re-suspension episodes

The re-suspension of dust can increase both PM2.5 and PM10 to very high levels. PM10 concentrations reaching 400 – 500 µg/m³ were recorded during this study at the kerbside sites in Stockholm, Göteborg and Umeå. In Stockholm it is mainly during these episode that the EU directive of 50 µg/m3 as daily average is exceeded. Most exceedances occur during March to April.

The PM concentrations at kerb side are governed by many parameters; e.g. traffic density and the ratio between cars and trucks, speed, meteorological parameters as humidity, wind speed and direction, temperature, precipitation amount and duration between precipitation events, type of precipitation (snow – rain). Other important parameters are the amount of dust available on the road surface and the type of street (with or without surrounding buildings, height of buildings).

On a given street, the meteorological conditions are of the greatest importance. If the street surface is wet, the amount of re-suspended particles can be reduced to practically zero, whilst dry conditions and dust on the road surface increases PM in the air to high concentrations. An example of the large variation governed by the meteorological conditions is given in Figure 8. The concentration of coarse particles, PM10 – PM2.5, at the kerb side site Hornsgatan in Stockholm was very low during the most of first half of the month, but during the second half the daily maximum hourly average was over 100 µg/m³ practically every day.




*Figuer 8. Hourly averages of coarse particles, PM10 – PM2.5, at Hornsgatan, Stockholm, March 2001*

### Inversion episodes

Local episodes due to inversions that trap the emissions from the local source have mainly been observed at Lycksele, the site in the northern Sweden inland. These episodes occurred during cold periods in winter when inversions are frequent. A typical episode occurred in the end of March 2000 (see Figure 9).

During the 25th to 29th PM10 and PM2.5 increase from about 10 - 30 µg/m³ at 4 - 5 in the afternoon to 200 - 400 (PM10) and 100 - 200 (PM2.5) at 10 - 11 in the evening. The rapid rise occurs while the temperature decreases from about zero to minus 10 - 20 degrees. This rapid decrease is typical for inversions.

In Lycksele it has been estimated that annual emissions of PM due to local wood burning is 50 times higher than emissions due to vehicle exhausts. Since wood burning emissions occur mainly during the cold periods which often coincide with stable meteorological conditions it is likely that a large fraction of the high PM10 and PM2.5 levels during these episodes are due to incomplete combustion in wood boilers and wood stoves. However due to the high meteorological stability the topography strongly influence the wind flow and thus the pollution situation vary strongly depending on geographical position and upwind sources.

It is likely that the conditions are similar in other small cities in the inland of not only northern Sweden but also in the south, where wood burning in old boilers is the main source of heat in residential areas or in areas where wood stoves are used frequently partly for heating and partly for pleasure.

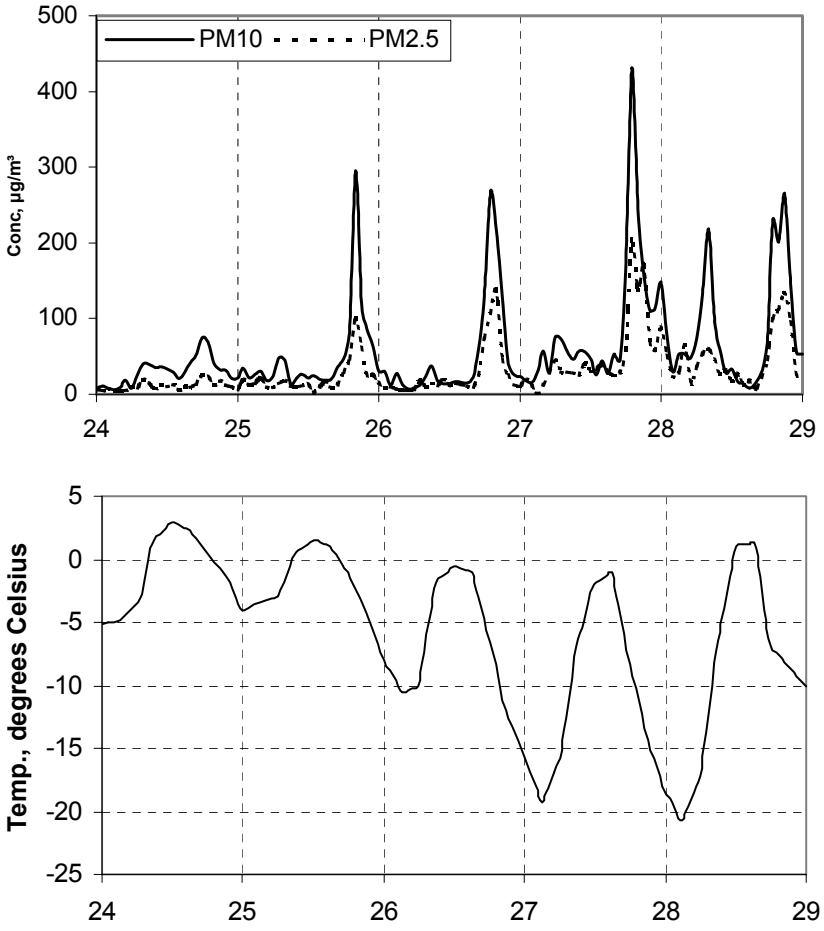



*Figure 9. One hour averages of PM10, PM2.5 and temperature in Lycksele March 24 –29, 2000*

## Discussion

### Concentrations in general

Comparing the Swiss studies, Hügelin et al., 2000 and Gehrig et al., 2003, and the data presented here show comparable; perhaps somewhat lower PM10 concentrations in Sweden. However the PM2.5 concentrations are lower in Sweden, besides the rural site in Switzerland that shows comparable concentrations with the Swedish rural sites.

The tail pipe contributions at Swedish kerb sites comprise only a small fraction of both PM2.5 and PM10 compared with similar Swiss sites. It seems like the Swiss sites have considerable less influence of long range transport and of re-suspended road dust. The probable causes are major differences in meteorology and less dusty roads. The meteorological situation, as noted by Hügelin et al. (2000) is strongly determining the absolute PM concentrations, indicating a similar situation as in northern Sweden with frequent inversions, thus causing bad ventilation and limited dilution. The local sources become dominating which also is the case in the Swiss study.

In comparison with the PM10 concentrations in United Kingdom, reported by AEA, the UK kerb/road side has similar concentrations, while the UK urban background sites, remote and rural sites show somewhat higher concentrations (AEA, 2002). The EMEP report on rural PM10 measurements mainly from Switzerland, Germany, Spain, Italy and the Netherlands shows also that the Swedish rural sites have lower concentrations compared to Western Europe with about 5-12 mg/m3 except for some Swiss stations that has comparable concentrations. The Spanish stations reach considerable higher concentrations (Lazaridis et al., 2000 a).

Comparing with Central and Eastern Europe the concentrations at urban and urban background stations in Bulgaria, Czech Republic, Hungary, Poland, Romania and Slovak Republic the Swedish concentrations for PM10 and PM2.5 values are about 20 – 40 µg/m3 lower (Houthuijs et al., 2001).

### Importance of local and distant sources

The two rural sites in this study represent the conditions in South Sweden. The source regions in Europe less influence the PM levels in north Sweden, i.e. PM is lower. As the PM levels at the urban background sites during the early morning hours not are influenced much by any local sources, the data from the two northernmost urban background sites in Lycksele and Umeå can be used to estimate the rural background concentrations in Northern Sweden. In this way the rural PM2.5 and PM10 concentrations in northern Sweden can be estimated to 7 and 10 µg/m³ respectively, compared to 12 and 16 µg/m³ in the south.

The long-range transport component of the concentration of PM10 and PM2.5 in the urban background and at kerb sites may be obtained from the measurements at the rural sites. The temporal variation of the long-range transported PM is very different from the variation found at urban background and kerb sites. As shown above there is also a substantial gradient of the rural concentrations going from southern Sweden to the north due to the distance to the anthropogenic emissions in central Europe. Subsequently it is feasible to describe the main sources for the different types of sites.

In Figure 2 the ratio of PM2.5/PM10 is shown for different types of sampling sites. Going from the rural to the kerb side sites the mean ratio decrease from about 0.8 to 0.5 indicating the influence of a coarse particle source, e.g. re-suspension of road dust, wear of tires or brake linings. This is also clearly shown in the daily variation of PM10 and PM2.5 (see Figure 3). Further exploring the ratio PM2.5 / PM10 looking at the relation to PM10 at a kerb side site in Stockholm, Hornsgatan, shows how dominant the coarse particle emissions are (see Figure 10).

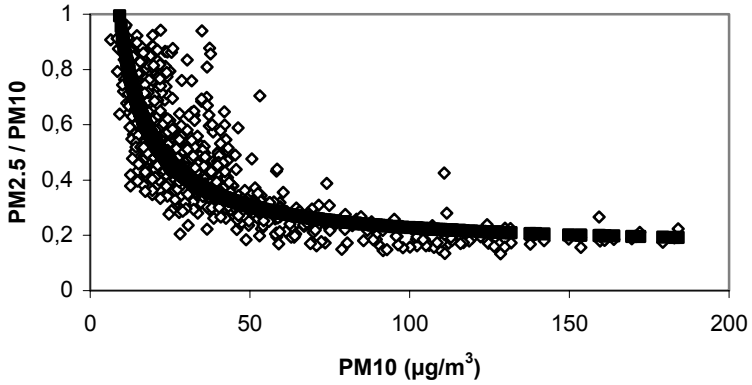



*Figure 10. The ratio PM10/PM2.5 versus PM10. Based on measurements from a kerb side site, Hornsgatan, in Stockholm during two years. The coarse line shows a model calculation*

The PM2.5 and PM10 concentrations at kerb side can be expressed as follows

$$PM2.5 = \frac{N}{D} q^{f}_{exhaust} + \frac{N}{D} q^{f}_{wear} + PM2.5^{wind}_{wear} + PM2.5_{Background}$$

$$PM10 = PM2.5 + \frac{N}{D} q^{C}_{wear} + PMC^{wind}_{wear} + PMC_{Background}$$

where $N$= number of vehicles passing, $q$ = source strength of fine ($q^f$) or coarse particles ($q^c$) from one vehicle (subscripts refer to sources), and $D$ = Dilution volume.

The background refers to roof top concentrations at Rosenlund as given in Table 2. Rosenlund is close to the street Hornsgatan.

Harrison et al., 2001, show the importance of the wind driven re-suspension of road dust but at Hornsgatan, which is a street canyon site, the coarse PM concentration does not correlate with wind speed. This indicate that the wind driven resuspension is normally much less important than that induced by the vehicles. Consequently this component is probably not significant at this specific site and thus excluded in the further analysis.

When N is large the background component becomes negligible and the ratio PM2.5 / PM10 simply becomes PM2.5/PM10 = $q^f/(q^f + q^c)$. At the highest measured values of PM10 the ratio is about 0.15 giving $q^c/q^f$ = 5.7, i.e. the coarse particle mass emissions are about 6 times the fine particle emissions from one vehicle.

Fitting this simple model to the data in Figure 10 using the setting discussed above gives the model fit shown in Figure 11. It is also reflected in Figure 11 fitting the surprisingly linear relation between PM10 and PM2.5, which is also found when studying the variation in emission factors at different PM10 concentrations ranges.

Performing the same analysis of PM2.5 and PM10 data collected at a roof site close by the street discussed above are not as successful. Occurrence where background air dominate and mixing with air mixed up from the street gives ratios of PM2.5 / PM10 between 0,2 and 0,8. The characteristics of the two types of aerosols confine the area in which the data points are found.

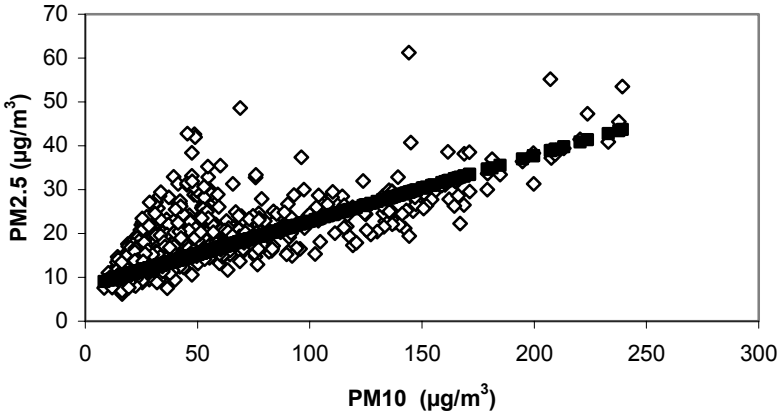



*Figure 11. PM2.5 versus PM10. Based on measurements from a kerb side site, Hornsgatan, in Stockholm during two years. Model calculation shown by coarse lines*

## Emission factors

A more accurate way of getting an estimate of the emission factor for resuspended particulates at the street canyon site in central Stockholm is to use NOx as a tracer. At this site NOx, PM10 and PM2.5 are measured both at street level (3 meters above the street and at roof level (urban background). The contribution of the emissions of the local traffic at the street may then be estimated as the difference between the concentrations at the street and the urban background. Then the emission factor for PM10 and PM2.5 may be estimated as

$$e_f^{PMx} = e_f^{NOx} \cdot \frac{C_{PMx}^{local}}{C_{NOx}^{local}}$$

where $e_f^{NOx}$ is the NOx emission factor for the vehicle fleet at Hornsgatan and $C_{PMx}^{local}$ and $C_{NOx}^{local}$ are the concentrations due to the emissions of PMx (PM10 or PM2.5) and NOx at the street. The emission factor for NOx has been calculated based on the road traffic model of the National Road and Traffic Administration of Sweden (EVA model). The vehicle composition at Hornsgatan is rather well known due to both manual and automatic traffic registrations during several years. For 1999 to 2001 a value of 1.4 g NOx per vehicle kilometer was applied and the resulting monthly mean emission factors for PM10 and PM2.5 are shown in Figure 12. For PM10 there is a very large variation with maximum values during February to April. This is due to the contribution from coarse particles since the emission factor for PM2.5 does not show the same increase during this period. The average value for the whole period is 223 mg/vehkm for PM10 and 48 mg/vehkm for PM2.5. These emission factors include vehicle exhaust emissions and emissions due to resuspension of road dust and wear of tyres and brake linings.

These estimates may be compared with estimates made based on measurements in German cities. Lohmeyer (2001) found emission factors between 60 and 140 mg/vehkm for PM10, i.e. somewhat lower than in Stockholm. The differences found may be due to different road maintenance — in Scandinavia sand and salt is put on the roads to decrease friction and studded tyres is commonly used in these countries. Differences may also be large between cities in Sweden due to large differences in climate and thereby the frequency of sanding/salting and also the use of studded tyres may vary in the country.

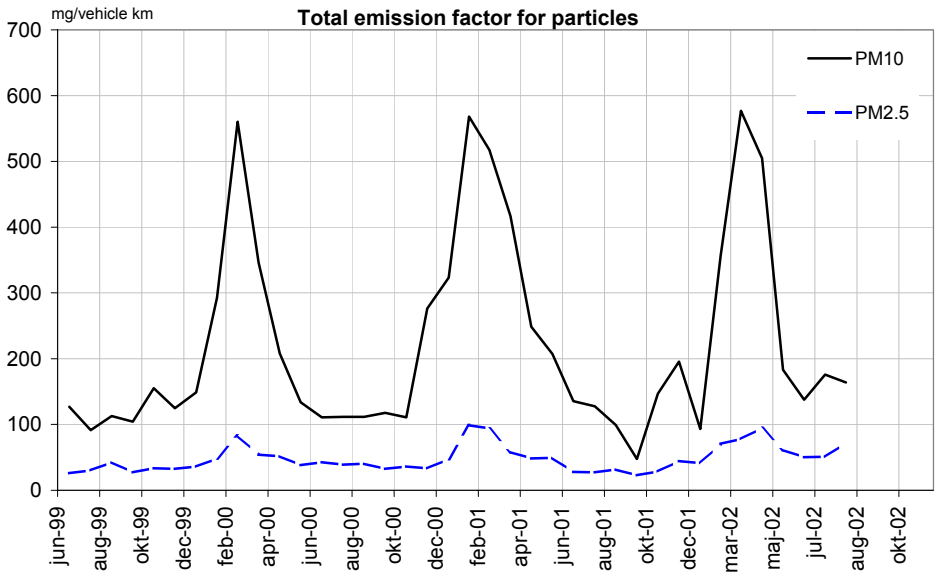



*Figure 12. Emission factors for PM10 and PM2.5 calculated using NOx as tracer for traffic intensity.*

Assuming that the average vehicle exhaust emission of particles is approximately equal to the minimum value of the calculated $PM_{2.5}$ emission (23 mg/vehkm) the non-exhaust emissions of $PM_{10}$ and $PM_{2.5}$ may be estimated. For $PM_{10}$ the non-exhaust emissions would thus contribute with 200 mg/vehkm. Then 85% of the locally generated PM10 concentrations are due to non-exhaust particle emission. For PM2.5 non-exhaust emissions of PM contributes with 25 mg/vehkm, which means that 45% of the locally generated $PM_{2.5}$ concentration is due to non-exhaust emissions.

Using these emission factors the mean of the ratio giving $q^c/q^f = 3.4$, which is lower than found with the simple model shown in previous chapter. The discrepancy depends on the former model assumes the emission factors to be constant over the whole PM10 concentration range and set the relation using the highest concentration when actually the non-exhaust emissions factors are highest.

In a UK study Harrison et al., 2001, came to the conclusion that find the resuspension to be vehicle induced with approximately equal source strength as the exhaust emissions. Both our modeling efforts show considerable higher source strength for resuspension and wear connected to traffic at Swedish road conditions.

Found emission factors for non-exhaust emissions for PM2.5 and PM10 are equal and 9 times larger than for exhaust emission factors. These findings explain the found linear relation between PM2.5 and PM10. It also points out that any abatement strategy excluding these sources will be unsuccessful.

### High resolved size distribution measurements

Direct measurements of the particle size distribution with a set of instrumentation, as Condensation Particle Counters, Differential Mobility Analyzers and Optical Particle Counters, covering sizes from 3 nm to 10 mm used in the same environment as discussed above, i.e. Hornsgatan in central Stockholm shows a strong support for assuming the coarse particles being emitted with a considerably higher emission factor (see Figure 13) (Isaksson, 2000). However when going to a suburban area a different relation ship is found probably depending on the large influence of long distant transported background air.

The size distributions in Figure 13 further also indicate that the PM2.5 contains a fair fraction of coarse particles. These results are corroborated by the findings by Hedberg et al (2002) at the northernmost site, Lycksele, were re-suspended dust were shown by source receptor techniques to

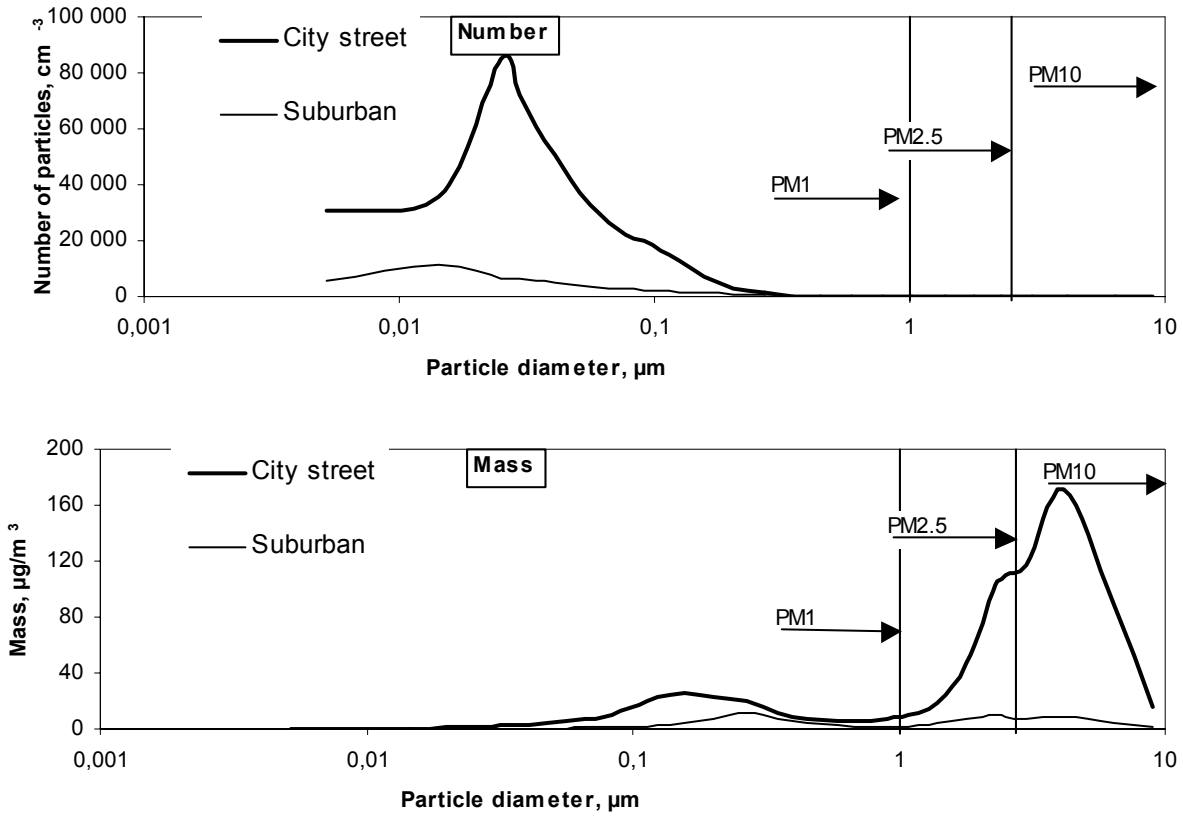



*Figure 13. Number and mass size distribution measure at an inner city and a suburban site in Stockholm PM1, PM2.5 and PM10 are indicated.*

comprise 1/3 to 2/3 of the PM2.5 mass during local air pollution episodes. Swietlicki et al. (2002) finds about 35% of PM2.5 is soil or sea spray related at the background station Vavihill. This supports our findings with the tracer model. The simple model gave a relation of 6 times more mass of coarse particles than fine particles were emitted, based on the PM2.5 / PM10 ratio. If roughly half the PM2.5 fraction is coarse particles the actual relation in source strength between coarse and fine particle is roughly 12, in mass units from traffic, which is still an overestimate but acceptable as a rough estimate.

It is obvious from both the tracer model result and the size distribution measurements that PM2.5 is strongly disturbed by non-exhaust coarse particle sources. PM2.5 do not reflect the correct concentrations due to combustion sources and long range transported emissions. Using PM2.5 will strongly complicate the assessment of sources and the choice of abatement strategy.

### Overall estimate

The collected information on the different sources allows an estimate of mean annual PM2.5 and PM10 concentrations in different environments all over Sweden (see Table 3). Background, i.e. influence of long distant transport decrease roughly with a factor of 2 comparing north and south. Subtracting the background from the found concentrations and using above found relation between the coarse and fine emissions it is possible to assess the contribution from re-suspension in urban environment where traffic is the main local source. It should be noted that Table 3 only give annual mean concentrations so concentrations at occasions from the different sources can vary very much more.

In situations with inversion the local sources will totally dominate the concentrations and very high concentrations can occur with small sources. Basically the meteorology determines the concentrations.

| | | Source type | | | |
|---|---|---|---|---|---|
| Site type | Fraction | Long range | Re-suspension | Other local source | Sum |
| Rural | PM10 | 8 – 16 | - | - | 8 – 16 |
| | PM2.5 | 7 – 12 | - | - | 7 – 12 |
| Urban background | PM10 | 8 – 16 | 5 | 1 | 14 – 23 |
| | PM2.5 | 7 – 12 | 1 | 1 | 9 – 15 |
| Kerbside | PM10 | 8 –16 | 22 | 4 | 34 – 43 |
| | PM2.5 | 7 – 12 | 3 | 4 | 14 – 20 |

*Table 3. Estimates of annual mean source contribution in µg/m³ at different type of sites. The range given for the long range contribution is depending on location in Sweden, lowest values in the north.*

The number of exceedances in Lycksele, a small town situated in the inland of northern Sweden that is the region with most occurrences of inversions, will give an idea of the importance of this phenomena. During the two year period 10 days with higher concentrations than 50 µg/m³ where detected in Lycksele. However it is likely that a major part of the inland is affected by such events. Consequently local inversion and local sources can have considerable effects in spite of small emissions.

## Summary and conclusions

Two years of measurements of PM2.5 and PM10 in different environments in Sweden, ranging from rural background, urban background to kerbside sites, on 1-hour basis show distinct features depending on time and spatial variability of the different sources. Based on this dataset it has then been possible to assess the importance different sources for the annual PM10 and PM2.5 levels.

Long distance transport has been shown to be substantial giving an annual mean contribution of 7 and 8 µg/m³, for PM10 and PM2.5 respectively in the north and 13 and 17 µg/m³, in the very south. The most important local source in urban areas is re-suspension of road dust giving in at kerbside sites annual mean contributions of 17 to 22 µg/m³. Other local sources are of minor on an annual basis, possibly with the exception of the contribution from wood burning mainly in urban areas situated in the inland of Northern Sweden.

Further it is shown that at sites where traffic is the major source, the coarse particles mass is about 6 times the fine. However it is also found that PM2.5 can contain a considerable fraction of re-suspended particles, i.e. the same source as the fine particles.

Typical annual mean values for PM2.5 at rural background, urban background and kerb site are 7 - 12, 8 - 15 and 13 -19 µg/m³ respectively, while for PM10 corresponding values are 9 – 17, 16 – 23 and 25 – 40 µg/m3. The ratio PM2.5/ PM10 is about 0.8, 0.6 – 0.7 and 0.4 – 0.6 at rural background, urban background and kerbside, respectively. This strongly decreasing ratio is due to the importance of emissions of particles generated due to mechanical wear of roads, tires, brake linings and the ejection of particles from the pavement due to resuspension processes.

Low level inversions may lock out the long distance transport contribution and allow accumulation of the locally emitted air pollutants. Measurements at an inland urban site of northern Sweden have shown that hourly PM10 levels may be 50 times higher than the annual average levels during such episodes, which mainly occur during winter time. It is likely that wood burning is the main source of PM10 and PM2.5 in these areas.

In the largest cities road dust emerges as the major local source responsible for exceedance of limit values for PM10. Using NOx as tracer for traffic exhaust it has been shown that the annually averaged emission factor for non-exhaust particles is about 10 times the exhaust particle emission of PM10. For PM2.5 the non-exhaust particle emission is about 2 times higher than the exhaust emission. The non-

exhaust emission show a very pronounced seasonal variation, with maximum values during the spring (March to April).

This means that exhaust emission control measures in Sweden will have little or no effect on future PM10 levels in urban areas of Sweden. The only local measures that would result in lower PM10 levels is connected with the coarse particle generation, i.e. road, tire and brake wear and measures to reduce the ejection of particles residing on the roads (road maintenance). The introduction of new boilers and wood stoves will lead to reduced emissions of PM and this may be important in some residential areas where wood is commonly used for heating during winter.

Since the long-range transport of PM10 due to emissions in central Europe, stronger emission control legislation at international level would also be needed. This involves not only direct emissions of PM but also the emissions of precursors (NOx and SO2) as secondary particles are formed during long-range transport.

It is obvious from this study that PM2.5 is strongly disturbed by non-exhaust coarse particle sources. PM2.5 do not reflect the correct concentrations due to combustion sources and long range transported emissions. Using PM2.5 will strongly complicate the assessment of sources and the choice of abatement strategy. Use of PM1 would avoid these problems.

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