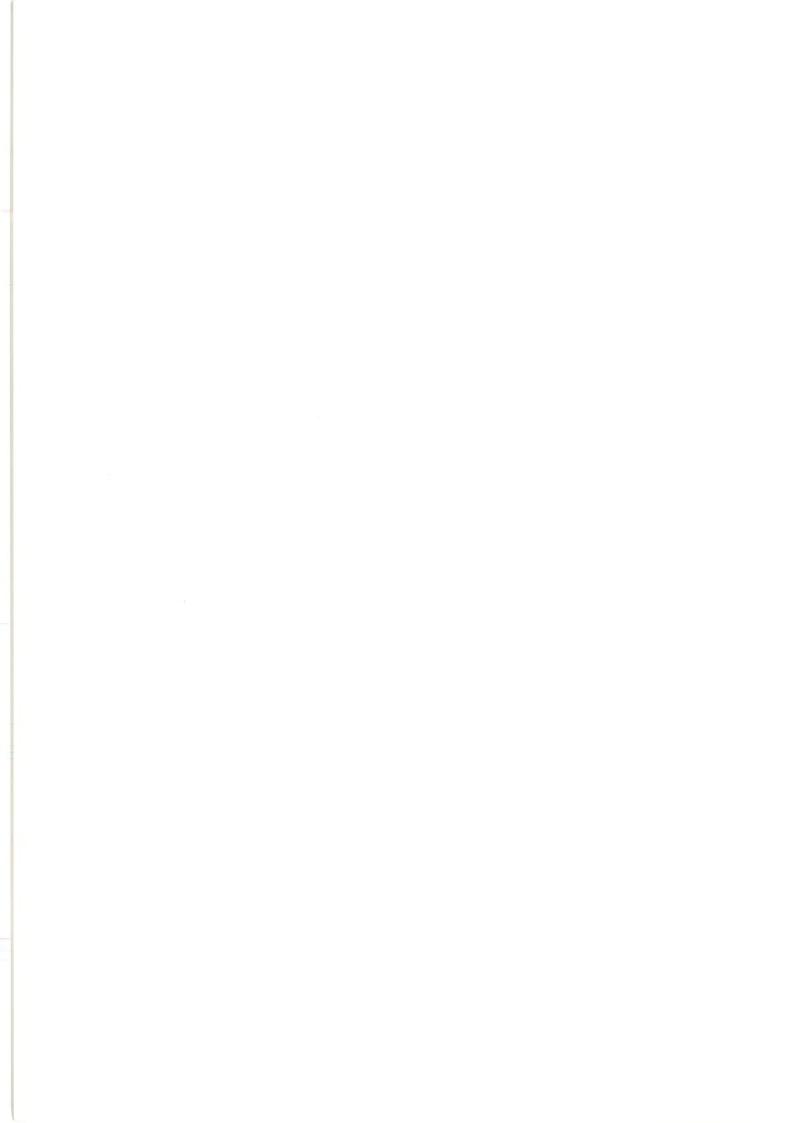


Calculations of PM-10 concentrations in Swedish cities - Modelling of inhalable particles.

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# **SMHI** REPORTS METEOROLOGY and CLIMATOLOGY

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Report Summary / Rapportsammanfattning Issuing Agency/Utgivare Report number/Publikation Swedish Meteorological and Hydrological Institute RMK No. 76 S-601 76 NORRKÖPING Report date/Utgivningsdatum Sweden April 1997 Author (s)/Författare Björn Bringfelt, Hans Backström, Sven Kindell, Gunnar Omstedt, Christer Persson and Anders Ullerstig Title (and Subtitle/Titel Calculations of PM-10 concentrations in Swedish cities - Modelling of inhalable particles Abstract/Sammandrag The project was initiated by the Swedish Environmental Protection Agency in order to improve the basis for making standards for PM-10 concentrations in urban air. Model development has been made for Norrköping and Gothenburg. Modelling has been necessary of both long-range and local contributions. The long-range dispersion has been simulated by the MATCH-Europe and MATCH-Sweden models, the former being a conventional dispersion model based on meteorology and emission data and the latter including also results from background air pollution measurements in Sweden and neighbouring countries. For the local scale, both car exhaust particles and resuspended particles from a traffic source inventory are dispersed in the city using meteorological data and a Gaussian dispersion model and, if needed, a street canyon dispersion submodel. For particle resuspension, a new model has been developed. The parameters of the resuspension model have been adjusted to two streets/measuring periods in Norrköping. At street level in Norrköping, the local model showed to contribute to a large part of the total concentration, especially in late winter and early spring due to resuspension. The MATCH model gives some underestimate due to the absence of organic compounds and simulation of PM-2.5 instead of PM-10. However, at some episodes in Norrköping and, in general, for a station at roof level in central Gothenburg and at the rural station Aspyreten, the model estimates of regional PM-2.5 concentrations constitute a larger part of the observed PM-10 concentrations. A good covariation in time with measured data occurs for both the MATCH model and the local model. Key words/sök-, nyckelord PM-10 particles, PM-2.5 particles, inhalable particles, road dust, street sanding, street particle depot, particle resuspension, air pollution dispersion model, long-range transport, MATCH model Language/Språk Supplementary notes/Tillägg Number of pages/Antal sidor English 77 ISSN and title/ISSN och titel 0347-2116 SMHI Reports Meteorology Climatology Report available from/Rapporten kan köpas från: SMHI

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#### SUMMARY

The project was initiated by the Swedish Environmental Protection Agency in order to improve the basis for making standards for PM-10 concentrations in urban air. About 1½ years of PM-10 measuring data from five Swedish cities have been studied. The PM-10 concentrations have been measured hourly by the TEOM method, with addition of an ACCU equipment to get 85 days of some 20 tracer elements in a street in Norrköping analyzed by the PIXE method. Factor analysis of the PIXE data showed dominance in the PM-10 mass of burning, car exhaust and tyre material (Pb, S, Zn) to the PM-10 mass in November - December 1993. In January to March 1994 sand material (Si, Fe, Ti) dominated. This shows that local resuspension may be an important part of the PM-10 concentrations in Swedish cities, especially in the later part of the cold season, when the streets have been sanded repeatedly.

This work comprises model development and simulation of PM-10-concentrations in city air. Model development has been made only for Norrköping and Gothenburg. Modelling has been made for different scales:

- · Transport on national and European scale.
- Contributions from surrounding city and local street.

The former will be treated by the so-called MATCH model. For the latter part a local model has been developed. The PM-10 concentrations are for both scales calculated hourly and separately by submodels. For the local model, this report describes the primary model development for Norrköping and a test for Gothenburg.

#### The MATCH model

Some simplified studies of formation and transport of PM-2.5 particulate matter over Sweden, on the regional scale, have been performed using the SMHI MATCH (Mesoscale Atmospheric Transport and CHemistry) model. MATCH is a three dimensional Eulerian atmospheric dispersion model including sulphur and nitrogen chemistry, and for this study some additional, rather simplified, calculation routines have been added in order to estimate the main part of the PM-2.5 particulate mass. Two different versions of the MATCH model, MATCH-Europe and MATCH-Sweden, have been tested and model results have been compared to measurements of PM-10. The two model versions are operating on different scales and are also run in somewhat different ways. MATCH-Europe is a conventional dispersion model being based on only meteorology and emission data, while MATCH-Sweden includes also an optimum interpolation of results from back-ground air pollution measurements in Sweden and the neighbouring countries.

First a simplified model test has been performed, on the European scale, for some commonly studied sulphur and nitrogen compounds. The comparisons between obtained MATCH-Europe results and daily measurements at Scandinavian EMEP-stations for the November/December 1993 and March/April 1994 periods indicate a fairly good agreement. The MATCH model versions were then applied to calculations of PM-2.5 particulate mass in the air over Sweden. The results indicate for MATCH-Europe as well as for MATCH-Sweden that, in general, the model estimated regional scale PM-2.5 concentrations are lower than observed PM-10 concentrations. Compared to measurements on a street in Norrköping the model results are, during most of the time, much lower than observed PM-10. However, at some episodes in Norrköping and, in general, for a station at roof level in central Gothenburg and at the rural

station Aspvreten, the model estimates of regional PM-2.5 concentrations constitute a larger part of the observed PM-10 concentrations. For Aspvreten the correlation between model results and observations is fairly good, with a correlation coefficient of about 0.8 for both model versions, and the model calculated PM-2.5 mass is about half of the observed PM-10 values. This seems to be quite a realistic difference, since measurements indicate that about 30% of the PM-2.5 particulate mass consists of organic compounds (not included in the present model version) and an increase in particle size from PM-2.5 to PM-10 (used in the measurements) should also add some extra particulate mass.

#### The local model

In the source inventory, the PM-10 emission from both car exhaust and resuspension in a street, is assumed proportional to the traffic intensity. The traffic loads in both cities were invented using data issued by the Traffic Offices. Major streets and thoroughfare roads were treated as line sources and smaller streets were grouped into area sources.

Both car exhaust particles and resuspended particles are dispersed in the city by a Gaussian model and, for most street level applications, by a street canyon submodel, using the model Dispersion developed previously at SMHI.

Two kinds of resuspended particles are treated:

- particles from street sanding and salting.
- particles released from the pavement and tyres, primarily by the action of studded tyres.

The first source is largest in winter and spring, the second source is largest in winter but occurs also in summer. A model for resuspended particles has been constructed calculating the PM-10 concentrations from these two sources. The source strength for resuspension is regulated by the two corresponding particle depots on the street. It is also reduced by functions depending on the moisture amount in the depot. A function, using current weather observations, for wetting or drying the depot, and for determination of sanding/salting occasions is used for modifying resuspension.

The decay rate used for the decrease of the particle depots will act most strongly in dry conditions. The decay rate for the particle depots has been coupled to the resuspension loss by budget equations. A study of PM-10 data in Norrköping and results in Claiborn et al. (1995) show the emission factors for resuspension to be higher than given by Larssen (1991). Therefore, twice as high emission factors as given by Larssen, have been used here. The results of the resuspension model are sensitive to the decay rate of the particle depot. This decay rate regulates the size of the depots especially at later stages in the cold season and the resulting concentrations. The value of the decay rate has here been adjusted to the PM-10 data at two periods and streets in Norrköping.

Too high emission factors for resuspension, may be a reason for the high values of PM-10 at G:la Rådstugugatan, Hörngatan and Kungsgatan in Norrköping calculated by the local model. At Kungsgatan a reason is also that we have used the street canyon dispersion submodel whereas the real site was located more openly. For roof level at a site in central Gothenburg, the local PM-10 model, using the resuspension parameter values adjusted for Norrköping, gave some high values agreeing well with the measuring data.

In general, the resuspension model describes well the time course of the concentration during a cold season, if a reasonable value for the parameter decay is used. More tests are needed

studying primarily the values of the resuspension emission factor and the parameter decay, using existing and new PM-10 measuring data. A framework for such and other tests has been created here.

#### Sum of both models

The added concentrations from both models agree rather well for Norrköping and the time covariation with the measured data is remarkably good. However, the agreement in the size of the values can be partly ascribed to some overestimate by the local model and underestimate by the MATCH model. For roof level in Gothenburg, the MATCH model contributes significantly most of the cold season 1993-94, and the local model is important in January to March 1994 due to the action of local resuspension. Different episodes of high concentrations arising from different kinds of sources can be identified in the data. The contributions from the MATCH model and the local model are of different proportion depending on the type of episode.

#### 1. INTRODUCTION

The basis for making standards of inhalable particles is considered to be weak depending on scarcity of interpretation and generalisation of measured data. There are proposals of a standard for PM-10 by the Swedish Environmental Protection Agency (1994):  $20~\mu gm^{-3}$  as maximum average for winter half year and  $100~\mu gm^{-3}$  as maximum daily average. The Agency has initiated measurements of PM-10 in some Swedish cities. In addition it has been considered necessary to use a meteorological model to make maximal use of the measuring data and to generalise the results. A status report for this project was given by Bringfelt et al. (1995).

Results in this study show, that local resuspension may be an important part of the PM-10 concentrations in Swedish cities. Therefore, a simple model for particle resuspension has been developed.

This work comprises model development and simulation of PM-10-concentrations in city air. About 1½ years of PM-10 measuring data from five Swedish cities have been studied. Model development has been made only for Norrköping and Gothenburg. Modelling has been made for different scales:

- Transport on national and European scale.
- Contributions from surrounding city and local street.

The former will be treated by the so-called MATCH model. For the latter part a local model has been developed where the PM-10 concentrations are calculated hourly and separately by each submodel. Then their contributions are added and 24-hourly values are formed. All models except the street canyon model give concentrations in a grid net. For model calibration against measuring values, calculation are made primarily for the positions of the measuring stations. The following tasks are included in the project:

- providing emission data
- providing meteorological data
- providing air pollution data
- model development
- model calculations
- calibration of model against air pollution data
- production of a model system
- generalisation

Section 2 will describe the sites and methods for the PM-10 measurements. In Section 3 the PM-10 data are presented and discussed. Section 4 treats the MATCH model and Section 5 the local model. The source inventories, meteorological data and results will be presented within these sections respectively. In Section 6 the combined model results will be compared to measured data. Section 7 will discuss the total model system.

#### 2. THE PM-10 MEASUREMENTS - SITES AND METHODS

Particle data have been collected in five Swedish cities. The data are of somewhat different character as regards instrumentation, measuring height, street type, time periods etc. In all cases are given hourly values of PM-10 (TEOM), while the data for the PIXE analysis (ACCU) are on a 24 hr basis. In Table 2.1 below, the measuring data are summarised. For Norrköping, the measuring sites to be considered here, have been marked on the map in Figure 5.5.

The mass concentration of PM-10 in the air has been measured by eight TEOM instruments. TEOM (Tapered Element Oscillating Method) is a system for continuously recording the particle mass. A small tube is attached to a plate and is supplied with a changeable filter in its upper part. The tube is vibrated with a resonance frequency determined by the properties of the tube and the mass of the filter. The air is sucked through the filter catching the particles. The filtered air penetrates the tube and the airflow is watched to be held at a constant level.

While the particles are collected by the filter, the vibration frequency of the tube will be changed successively. This frequency is recorded continuously. The growing collection of particle mass on the filter is then calculated using a known relation between mass and frequency. Using the air flux, the mass concentration in the outdoor air is calculated continuously.

ACCU (Automatic Cartridge Collection Unit) is a method to collect large amounts of particles on several successive filters which are changed automatically. The filters are then analysed for tracer elements by e g the PIXE method. In Norrköping, one of eight successive filters has been changed every 24 hrs. Then, manual change is needed only weekly. Using ACCU combined with TEOM can give analysable particle quantities during a limited time such as 24 hours. The TEOM data (without ACCU) concerns PM-10 particles with cut-off corresponding to 50 % particle mass at 10 μm. The ACCU data are considered to have approximately the same cut-off. The data from Norrköping in 1992-1994 have been described by Nyquist (1995).

Table 2.1. Summary of the PM-10 measuring data. Period: 1992 10 01 - 1994 03 31.

G: Street level (generally about 3 metres above the street floor)

T: Roof level of surrounding houses.

(The Norrköping site called Hörngatan is really at G:la Rådstugugatan at the parking house Spiran facing the opening of Skomakargatan).

Measuring location	Level		tarting, closing time (year-month
Norrköping			
Kungsgatan	G	TEOM	921027-930123
Hörngatan	G	TEOM	930210-0503
Vattenverket		TEOM	930517-0910
G:la Rådstugugatan	G	TEOM, ACCU	930922-940331
Lindö		TEOM	921001-930930
Gothenburg			
Femmanhuset (central cit	ty) T	TEOM	The whole period
Mobile measuring station	1:	TEOM, ACCU	
- Majorna			921001-930401
- Utby			930403-1005
- Haga			931113-940221
- St Jörgens Park			940221-0331
Stockholm			
Ulvsundavägen	G	TEOM	930120-930610
Torkel Knutssonsgatan	Т	TEOM	930722-0809;930929-1107
Sundsvall			
Mobile measuring station	1	TEOM	
- Sjögatan			921001-1026;931001-1030
- Köpmangatan			921028-1220;931101-1130
- Skolhusallen			930101-0331;940105-0131
- Storgatan			931202-940103
Umeå			
City		TEOM	930201-0330
Ön		TEOM	940111-0331
Background station			
Aspvreten		TEOM	Whole period

#### DATA ON TOTAL PM-10 AND TRACER ELEMENTS

#### 3.1 General presentation

In Tables 3.1 and 3.2 are given the periods and sites for the TEOM and ACCU measurements respectively. Figure 3.1 a, b and c show time series of PM-10 in the five cities and PM-10 and SO<sub>2</sub> concentrations at the background stations Aspvreten and Vavihill respectively. The figure shows values surpassing the proposed standard for PM-10 of 100 μgm<sup>-3</sup> of the Environmental Protection Agency, only in a few cases: in Norrköping on 12th April 1994 and at the mobile station in Gothenburg (Haga) on 25-26 November 1993. Table 3.1, giving how many measuring days there were each week, shows the most complete series to be in Gothenburg and Norrköping. The measuring equipment has been moved in some cases.

Table 3.3 shows monthly PM-10 averages. As an example of measured values, Figure 3.2a shows 24-hourly concentrations at G:la Rådstugugatan in Norrköping, recorded during the same period as the ACCU data. There is a diurnal variation, shown in Figure 3.2b. The corresponding picture for the background station Aspvreten (Figure 3.3b) shows no such diurnal course. This is due to diurnal variation of the traffic intensity in Norrköping. From the figures can be seen that the median values in background air is about 8  $\mu$ gm<sup>-3</sup>, whereas the median values at G:la Rådstugugatan vary between 12  $\mu$ gm<sup>-3</sup> (at night) and 20  $\mu$ gm<sup>-3</sup> (daytime). The background concentrations there seem to form a significant part in the city air.

In February to May 1993 there are rather high measured PM-10 values in all the cities studied, see Figure 3.1. In the second week of March and in May this seems to depend largely on long-range contribution, since there are also high values in Aspvreten. In February, the rest of March and in April, however, an important source also seems to be local resuspension.

In the cold season 1993-1994 the long-range contribution was important for the PM-10 concentrations in Norrköping. In March and April 1994 the local resuspension became quite dominant.

#### 3.2 Episodes

In Figures 3.1 a, b and c, it may be seen, that the PM-10 concentrations are high at several locations during some episodes. During other episodes there are high concentrations only at a few locations. This indicates that the episodes are of different kinds: in the former case there is a long-range transport contribution, or there is similar local weather conditions at all locations causing high concentrations from near-by sources. In the latter case with high values only at a few locations, we have local sources and locally bad dispersion conditions. Aspyreten is a background station, and high concentrations there, often indicate contribution from long-range transport. Then, if there are also simultaneous high values in other locations/cities, these are probably due to long-range transport. About ten episodes with high concentrations have been listed in Appendix 1, referring to measured PM-10 values in various cities and background stations.

### 3.3 PIXE analysis of tracer elements in PM-10

Via the Institute of Applied Environmental Research (ITM, Stockholm) air concentration data have been obtained for tracer elements in PM-10. About 85 daily ACCU filters have been analysed by the Technical University in Lund, Division of Nuclear Physics using the PIXE method (Particle Induced X-Ray Emission). The measurement was made at G:la Rådstugugatan in Norrköping during November 1993 to March 1994. The periods used have been marked in Figure 3.1c. The size of the particles collected correspond roughly to PM-10. Table 3.2 shows for every week the number of days with ACCU filters used for PIXE analysis.

Figure 3.4 shows curves of daily concentrations of the tracer elements. It can be seen that the most important tracer elements (Fe, Mn and Ca for resuspension, S and V for remote sources and Pb and Br for exhaust gases) are rather frequent.

The same data are presented monthly in Figures 3.5 a, b and c for total PM-10 and the tracer element groups belonging to three source types:

earth surface (a) burning, exhaust, tyre material (b) and burning (c).

The occasions of precipitation are also given. In some cases it may be seen, that precipitation events are associated with dips in the concentration of PM-10 and tracer elements, see e.g. November 1993 and March 1994.

Gaps in the data have been introduced when the concentration is below the detection limit. The given detection limit of the filters (ng cm<sup>-2</sup>) varies from day to day (within a factor of about two), because spectral maxima for a tracer element can be disturbed by neighbouring maxima from other elements. In addition, the detection limit for an air concentration (ng m<sup>-3</sup>) may vary from day to day due to different air flow in the instrument. This second effect is stated to be small, due to rather constant air flow. For each tracer element, the detection limit in ng m<sup>-3</sup> has been calculated as constant for all days, using the given mean detection limit (ng cm<sup>-2</sup>) divided by two, a value obtained from the current technical measurements data. If an element has a value much larger than the detection limit, which is often the case, the abovementioned two disturbing effects are small.

## 3.4 Comparison between data from Norrköping with old data from Nyköping

Concentrations of total PM-10 and tracer elements from two periods in Norrköping are compared below with old data from Nyköping. It may be seen that a few elements like Cr and Ni, have a larger share in Norrköping than in Nyköping. Most other elements have a smaller percentage, in particular Fe and Pb, for Pb due to lower concentrations in car exhaust gases.

	Nyköping 1	1984-85 Norrköpin 4 Nov - 2			Norrköping 21 Jan - 22	
	ng m <sup>-3</sup>	%	ng m <sup>-3</sup>	%	ng m <sup>-3</sup>	%
S	3480	7.9	1547	8.3	828	4.2
Cl	230	0.5	44	0.23	217	1.1
K	437	1.0	105	0.56	90	0.46
Ca	154	0.35	35	0.19	46	0.23
V	10.4	0.023	2.8	0.015	3.0	0.015
Cr	2.9	0.0066	2.9	0.015	2.8	0.014
Mn	13.3	0.03	3.4	0.018	2.9	0.015
Fe	550	1.25	70	0.37	83	0.42
Ni	1.8	0.004	2.4	0.012	2.3	0.012
Zn	45.4	0.10	19	0.10	11	0.056
Pb	536	1.22	11	0.06	6.1	0.031
PM-10 total	43857	100	18727	100	19699	100

## 3.5 Comparison between the Norrköping data and the Aspvreten background data

The following table shows monthly values of PM-10 (µg m<sup>3</sup>) in Norrköping and the background station Aspyreten and quotients in per cent.

Year	1992	1993	1994
Month	10 11 12	01 02 03 04 05 06 07 08 09 10 11 12	01 02 03
Norrköping	15 14 17	16 14 26 30 20 8 11 11 14 17 22 15	15 19 26
Aspvreten	6	- 13 13 13 18 7 11 10 8 9 27 11	9 17 9
Aspvreten/ Norrköping			
in per cent	41	97 48 43 91 85 94 86 59 57 124 73	59 88 36

Aspvreten shows a rather large part of the concentration in Norrköping indicating that long-range transport is important for the PM-10 concentrations in Norrköping.

In November 1993 Aspyreten showed apparently larger concentration, due to smaller number of measuring days: only 6 against 29 days in Norrköping. In the end of November 1993 there was also an episode of high S and Pb values at both locations (see time curves for tracer elements and PM-10 values). During this episode the PM-10 values were rather similar in Norrköping and Aspyreten, indicating important long-range contribution at both locations.

In January to March 1994, Aspvreten in average had some 60 per cent of the concentrations in Norrköping. In February 1994 there were occasionally high PM-10 concentrations in Aspvreten.

## 3.6 Factor analyses

In a computer program for factor analysis, the time series of the tracer elements having similar variation are first grouped into a smaller number of time series of so-called factors, which often turn out to be related to the source types. If two or more tracer elements vary similarly, they will be grouped into the same factor or source type. After that, the factor series are transformed into absolute series comparable to measured total PM-10 values. Regression of measured total PM-10 concentrations against the absolute factor series for the different source types gives linear regression equations. Substitution of the time mean values of the factors into the regression equations gives the mean mass shares of total PM-10 corresponding to the source types. It is important to prescribe a correct number of factors to use. One has to try different numbers. The number to use depends on the number of different types of covariations in the tracer elements data. Using too small number will give too much weight to the constant term in the regression equation. All these steps have been coded into the computer program and the method was described in detail by Thurston and Spengler 1985.

Two "absolute" factor analyses of tracer element data from the ACCU equipment and total PM-10 concentrations in Norrköping gave the following result. Tracer elements with a correlation coefficient to the current factor series greater than 0.7 have been indicated.

Period in the end of 199	3 (4 Nov - 27	Dec), 44 days:			
Typical tracer elements	Pb Zn Se S K As Mn	Si Ca	V Ni		
Probable source type	Burning +Exhaust (+Tyre mate	Earth Surface	Burning	Indefinite (Constant term)	Total
Mass shares in PM-10 (μg m <sup>-3</sup> )	12.7	4.6	0.06	1.4	18.7
Mass share in per cent	68	25	0.3	7.4	100

## Period in the beginning of 1994 (21 Jan - 22 March), 41 days:

0.6

3.3

PM-10 (µg m<sup>-3</sup>)

Mass share in per cent

Typical tracer elements	S Ni Pb P V	Si Fe Ti	Cu		
Probable source type	Burning +Exhaust	Earth Surface	Burning	Indefinite (Constant term)	Total
Mass shares in					

15

77

0.7

3.6

3.2

16

19.7

100

In the period in the end of 1993 (first analysis above), there was a great mass contribution from "Burning+Exhaust". This is partly due to the above-mentioned episode in November dominated by long-range transport. According to Johansson (1996), Zn occurs in tyre material, which could also contribute to the largest source type in the first analysis above. Data from the beginning of 1994 (second analysis) show a great contribution from the earth surface to be ascribed to local sources in Norrköping such as resuspension of sand particles. This is due to larger depot of sand particles later in the winter due to sanding of the streets. In Norrköping there is so strong co-variation between the Pb and S values, that the program has brought them to the same factor (source type). Some of the features above, may also be seen by direct inspection of the curves for measured values in Figure 3.4. It should also be pointed out that important source types for PM-10 mass may have no tracer elements in the data, such as NO<sub>x</sub> and NH<sub>4</sub>.

**Table 3.1.** Numbers of measuring days at the measuring sites for PM10 (TEOM) in each week during the model test period 1992-10-01 to 1994-03-31. 240 means week No 40 in 1992.

	2222222222 444444444 123456789	5555	000000000	1111111111	222222222	3333333333 3333333333 0123456789	444444444	555	000000000	1111
1. 2. 3.	Norrköping Kungsgatan,373477 Norrköping Hörngat : Norrköping Vattenverket: Norrköping G:la Rådstugug.:	7777 	576 5777	77777771-	7726777777	7777775				
4. 5. 6. 7.	Norrköping G:lä Rädstugug.:  Norrköping Lindö4777777777  Umeå  Ume älv  Sundsvall Storgatan	1111	111111111	115-116111	1111111531	1//////////				
8. 9. 10.	Sundsvall Sjögatan:47771 Sundsvall Köpmangatan:577777 Sundsvall Skolhusallen:	77	777777757	7773		3	7776		57771	
13.	Göteborg Femmanhuset:4777777777 Göteborg Majorna5 Göteborg Utby Göteborg Haga	7777	77777777	7774	767777777	777777777	3			4444
17.	Göteborg Haga		5777777	777777777	77744	7715	77777	150		222

**Table 3.2.** Numbers of measuring days with ACCU method, used for PIXE analyses of tracer elements in PM10, at Gamla Rådstugugatan in Norrköping in each week during the model test period. 344 means week No 44 in 1993.

Table 3.3. Monthly PM10 average concentrations ( $\mu g \ m^3$ ) at eight measuring sites. N is the monthly number of days used which have at least 18 hours of measuring data.

	Umeå	Sunds- vall	Stock- holm	Norr- köping	Lindö	Göteborg	Göteborg Mobil	Asp- vreten
Okt '92 N	0	9.86 30	. 0	14.60 1	8.58 28	111.59 31	. 0	5.99 6
Nov '92 N	0	14.53 30	0	14.06 18	8.64 30	11.38 30	0	0
Dec '92 N	0	18.27 20 '	0	17.13 31	12.99 31	14.33 31	23.48 29	0
Jan '93 N	0	15.12 31	10.59 12	16.05 18	10.96 31	19.39 31	30.95 29	0
Feb '93 N	7.61 23	20.60 26	15.83 28	14.25 17	12.18 28	23.07 28	32.29 28	13.76 6
Mar '93 N	17.25 26	40.18 29	24.23 31	26.00 29	16.62 25	26.11 31	43.04 31	12.59 27
Apr '93 N	. 0	. 0	26.86 30	30.44 29	15.80 24	25.19 30	24.59 28	13.17 30
Maj '93 N	. 0	. 0	27.65 30	20.17 16	20.28 31	20.25 31	23.71 27	18.41 28
Jun '93 N	. 0	0	11.17	8.42 20	8.98 30	11.86 27	21.02 29	7.17 27
Jul '93 N	. 0	0	11.35 10	11.47 30	11.64 24	9.40 21	16.88 29	10.74 31
Aug '93 N	. 0	. 0	11.45 9	11.27 31	10.79 31	11.45 16	18.52 31	9.73 31
Sep '93 N	. 0	. 0	13.56	13.76 17	9.56 30	9.84 22	15.09 30	8.06 30
Okt '93 N	. 0	12.06 30	22.43 27	16.65 29	. 0	11.96 31	16.68 7	9.43 28
Nov '93 N	. 0	20.89 30	15.99 7	21.98 29	. 0	19.56 30	45.40 18	27.41 6
Dec '93 N	. 0	12.85 25	. 0	15.36 31	. 0	11.67 31	19.83 30	11.25 20
Jan '94 N	5.22 18	11.57 28	. 0	15.20 31	. 0	14.91 31	17.43 31	8.95 31
Feb '94 N	8.58 17	. 0	0	18.92 28	. 0	17.66 28	19.05 24	16.63 21
Mar '94 N	9.90 25	0	. 0	26.06 31	. 0	18.43 31	16.57 30	9.40 31



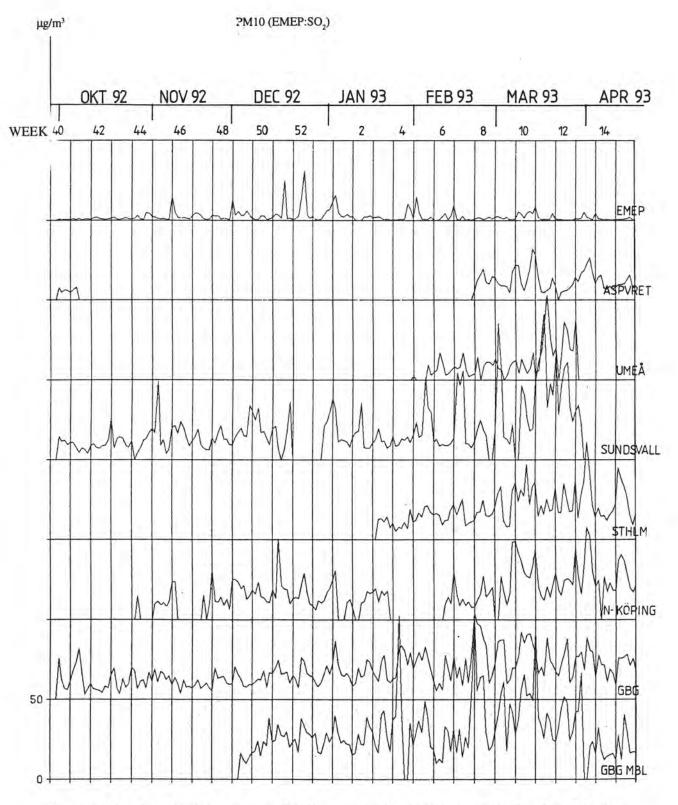
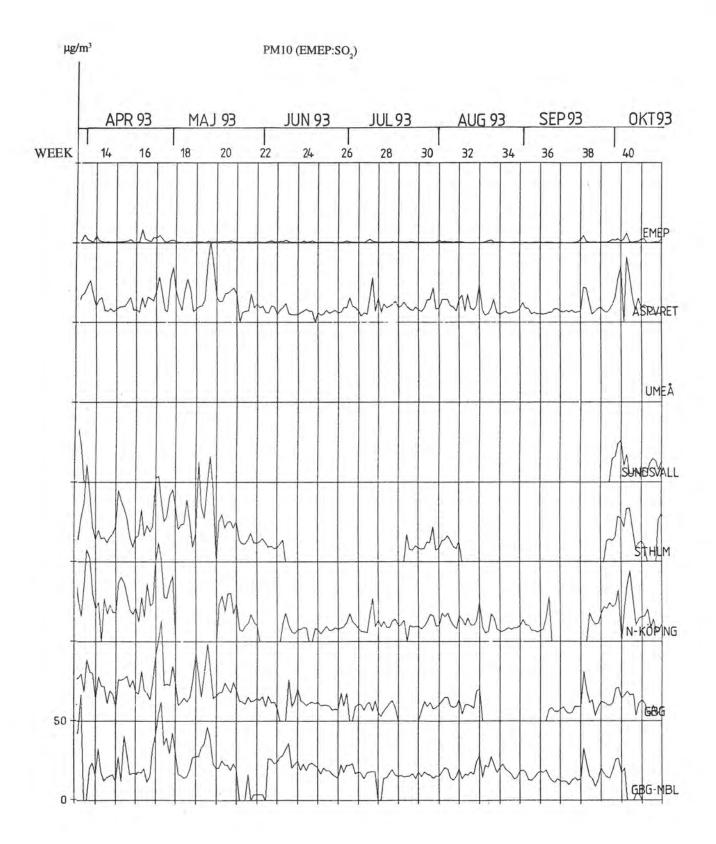
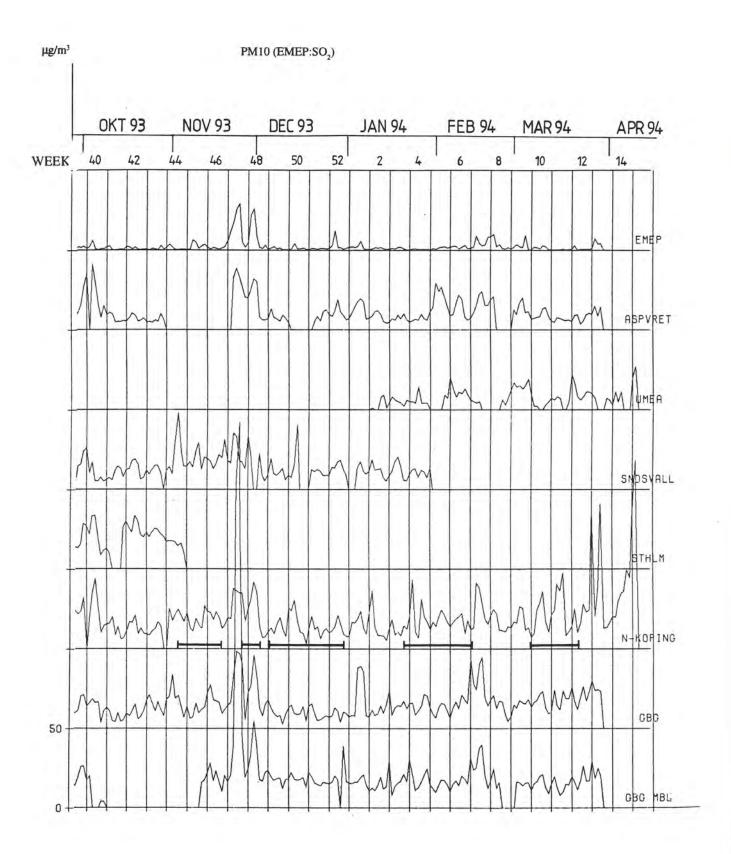


Figure 3.1 a, b and c. Time series of daily measured PM10 in  $\mu g$  m<sup>3</sup> in five Swedish cities and the background station Aspvreten (between Stockholm and Norrköping). The top curve concerns  $SO_2$  concentration in  $\mu g$  m<sup>3</sup> in the EMEP station Vavihill in southernmost Sweden. For Norrköping have been marked 85 days with PIXE-analyses of tracer elements in PM10. GBG MBL refers to the mobile station in Gothenburg. The vertical distance between the zero lines for the stations corresponds to 50  $\mu g$  m<sup>3</sup>.





c

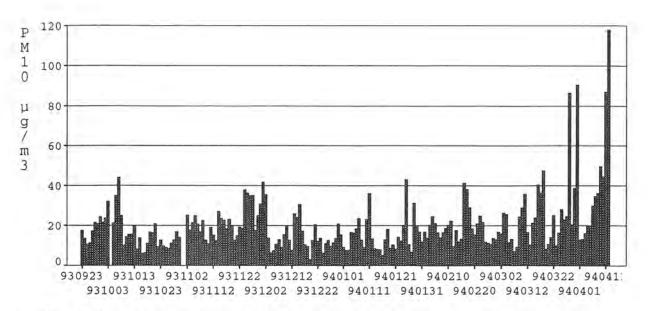


Figure 3.2 a. 24 hour averages of PM-10 at Gamla Rådstugugatan in Norrköping from 1993-09-23 to 1994-04-13 (all existing data at this site during the model test period).

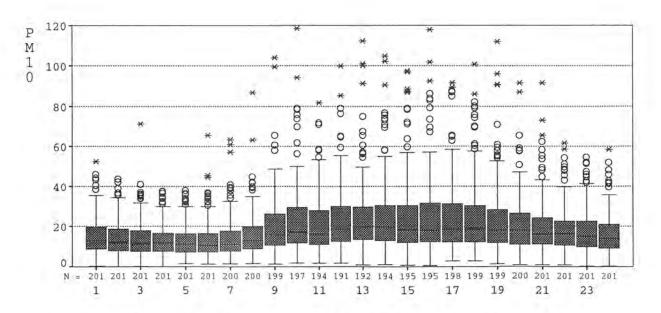


Figure 3.2 b. Statistics for each hour of the day of the PM-10 data of Figure 3.2 a.

For each hour of the day is shown the median (line within box). The
boxes extend to upper and lower 25 percentile. The T-marked lines extend
to the value of 1.5s where s is the difference between the 50 and the 25
percentile. Still higher (lower) values are marked with solitary symbols.

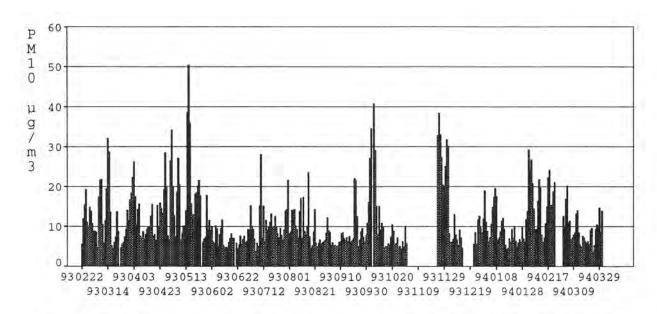


Figure 3.3 a. 24 hour averages of PM-10 at Aspvreten from 1993-02-22 to 1994-03-31 (all existing data at this site during the model test period).

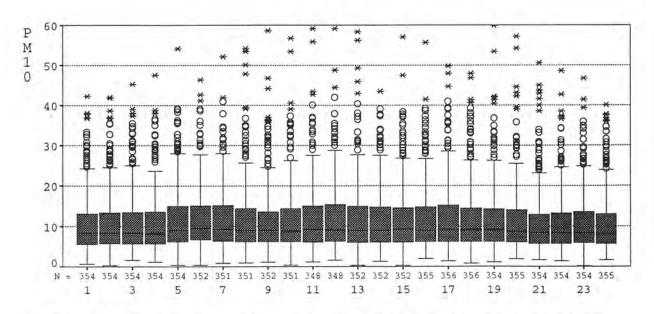


Figure 3.3 b. Statistics for each hour of the day of the PM-10 data of Figure 3.3 a. For explanation, see Figure 3.2 b..



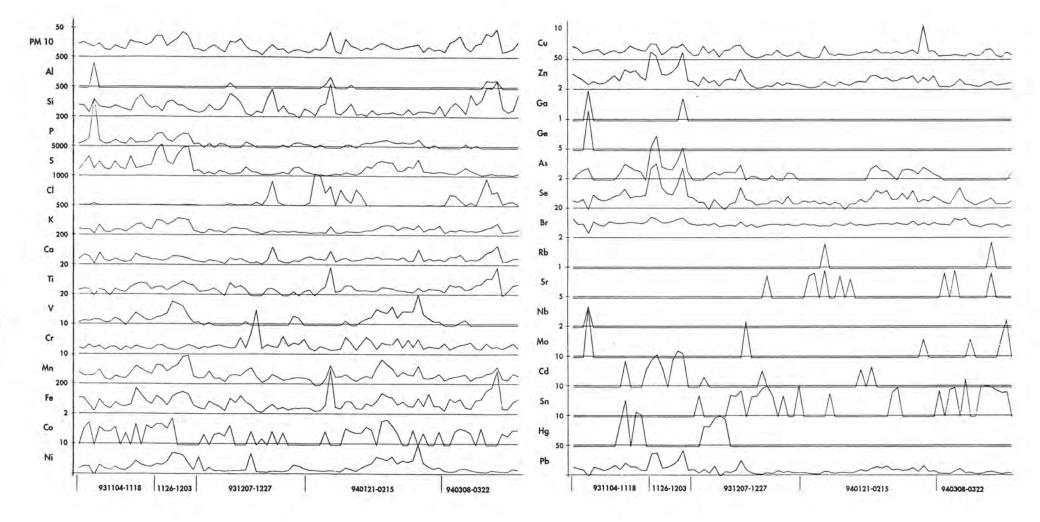
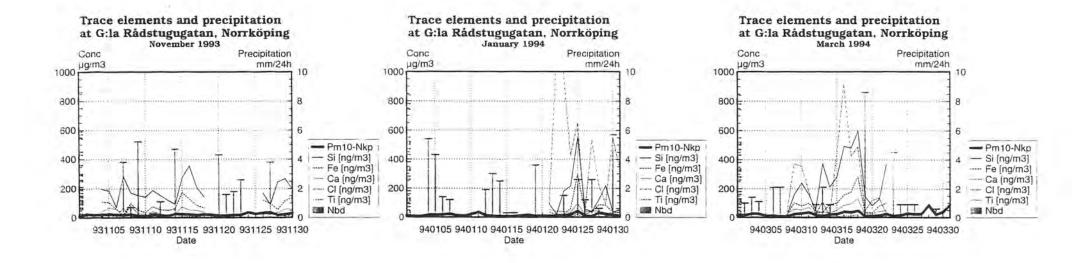


Figure 3.4. Curves of 85 daily concentrations of PM-10 and tracer elements during November 1993 to March 1994. PM-10: scale in µg m<sup>-3</sup>. Tracer elements: scales in ng m<sup>-3</sup>.



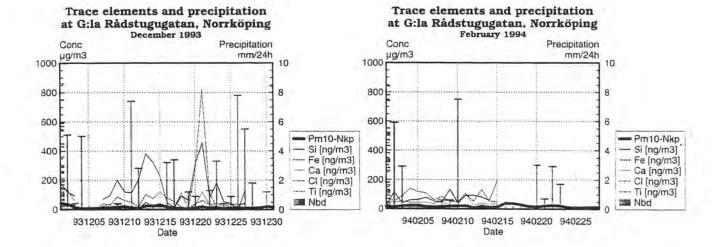
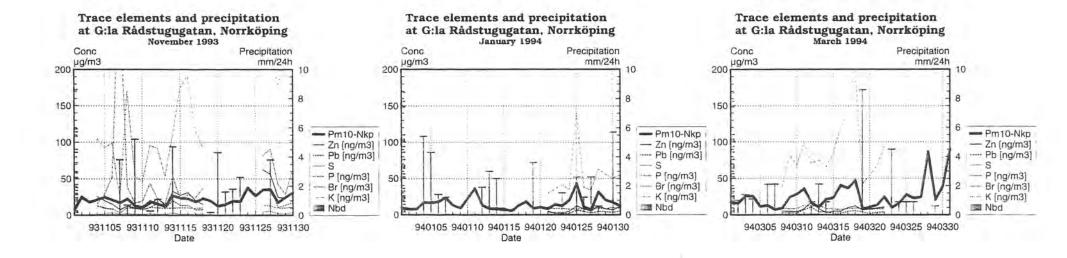


Figure 3.5 a. Monthly curves for measured PM10 and tracer elements characteristic to the earth surface. Precipitation events are given as vertical bars.



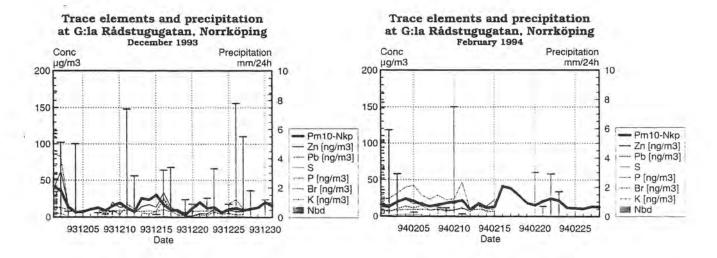
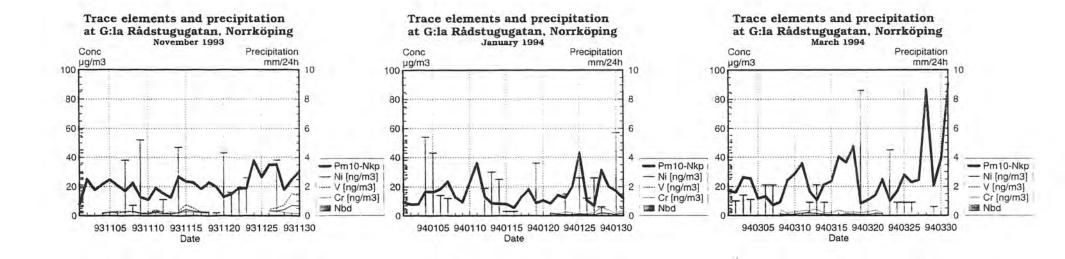


Figure 3.5 b. Monthly curves for measured PM10 and tracer elements characteristic to burning, car exhaust and tyre material. Precipitation events are given as vertical bars.



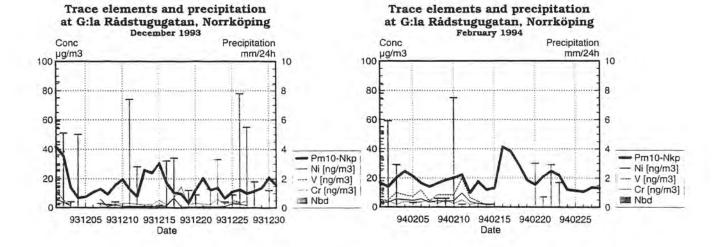


Figure 3.5 c. Monthly curves for measured PM10 and tracer elements characteristic to burning. Precipitation events are given as vertical bars.

#### 4. MATCH REGIONAL MODELLING OF FINE PARTICULATE MASS

The MATCH model is a 3-dimensional Eulerian atmospheric dispersion model, which is designed with options for operational applications on:

- meso-α scale (Europe or larger areas, grid 55 x 55 km)
- meso-β scale (Sweden, grid 20 x 20 km)
- meso-γ scale (different subregions within Sweden, grid 5 x 5 km).

The handling of the vertical mixing, the vertical and horizontal grid size, parts of the chemistry and a special technique for estimating the long-range transport contribution in the meso- $\beta$  and meso- $\gamma$  scale models essentially constitutes the difference between the various scales. In this section we will make a first test to use the MATCH-system for estimates of the regional scale particle-mass concentrations. A brief description is given of the features of the general dispersion model, including estimates of particulate mass concentrations, and the special model options for Europe and Sweden.

#### 4.1 General model description

The MATCH (Mesoscale Atmospheric Transport and CHemistry) model (Persson and Robertson, 1991; Persson et al., 1994, 1995 and Langner et al., 1996) is a three dimensional Eulerian atmospheric dispersion model. The model is a so called "off-line" model, meaning that it requires meteorological data from an external archive at regular time intervals (three hours in the applications presented below) in order to calculate transport, chemistry and deposition. These meteorological data are then interpolated to 1h values. The model version used in the above mentioned references is covering Sweden and parts of the surrounding seas and Nordic countries with a grid resolution of 20 x 20 km. That model version, which is applied to the MATCH-Sweden study below, has three layers in the vertical consisting of: a surface layer, the rest of the mixing layer and a reservoir layer above the mixing height. Recently the MATCH code has been further developed and improved in many ways (Robertson et. al., 1996) and e.g. the number of vertical layers can be chosen freely in the new model version. This new model code is used for the MATCH-Europe model version in the study below.

Horizontal advection, in all MATCH model versions, is calculated using a fourth order flux correction scheme (Bott 1989a, 1989b). The scheme utilises polynomial fitting between neighbouring grid points of the concentration field in order to calculate the advective fluxes through the boundaries of adjacent grid boxes. It is a positive definite mass conserving scheme with low numerical diffusion.

Vertical advection is calculated using a zero-order upstream scheme. Vertical diffusion between layers 1 and 2 in the MATCH-Sweden model version is for the convective case described from a determination of the turn-over time for the boundary layer based on similarity theory, and for the neutral and stable case from a parametrization based on a 40 layers 1-D eddy diffusivity model. Vertical transport is also induced by the spatial and temporal variations of the mixing height. In the MATCH-Europe model version, for which 7 vertical layers have been chosen in the present study, K-theory and a semi-implicit expression is used for the vertical diffusion calculation.

## 4.1.1 Chemistry

The chemistry in the model deals with sulphur oxides and oxidized and reduced nitrogen and is almost identical to that used in the EMEP model (Iversen et al., 1989). The following compounds are included: sulphur dioxide (SO<sub>2</sub>) ammonium sulphate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>HSO<sub>4</sub>), other sulphate particles (SO<sub>4</sub><sup>2</sup>), nitric oxide (NO), nitrogen dioxide (NO<sub>2</sub>), ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>), other nitrate particles (NO<sub>3</sub>), nitric acid (HNO<sub>3</sub>) and ammonia (NH<sub>3</sub>). The main difference compared with the EMEP model is in the specification of ozone (O<sub>3</sub>) concentrations, which is performed in the smaller scale models covering Sweden. In those model versions analysed O<sub>3</sub> distributions with one hourly time resolution are generated from observations. A local adjustment of the O<sub>3</sub> concentration with regard to local NO- and NO<sub>2</sub>-concentration and solar radiation is also done.

#### 4.1.2 Deposition processes

Pollutants are removed from the atmosphere by wet and dry deposition processes. Wet scavenging of the different species is taken as proportional to the precipitation rate and a species specific scavenging coefficient. Dry deposition is proportional to the concentration and a species specific dry deposition velocity at 1 m height. Since the lowest model layer has a thickness of about 75 m, the dry deposition flux calculation is transformed to the middle of that layer using standard similarity theory for the atmospheric surface layer. Dry deposition velocities are specified as a function of the surface characteristics (fraction forest, field etc.). Scavenging coefficients and deposition velocities have in most cases values close to those used in the EMEP calculations. For NO<sub>2</sub> and particles, however, somewhat larger dry deposition values have been used.

#### 4.2 MATCH-Europe

The MATCH-Europe model version, used in this study, has a horizontal grid-size resolution of 55 x 55 km. Our purpose, in this study, is to make a first tentative investigation of the possibilities to use MATCH-Europe simulation results for estimates of the regional scale particulate mass concentrations. We are, however, aware of deficiencies in the present model formulation regarding e.g. a complete description of all types of particulate compounds and their chemical and physical formation and degradation. This is further discussed below in Section 4.2.4.

The model version MATCH-Europe has up till now only been applied to non-chemistry or very simple chemistry problems, where the combined atmospheric chemistry calculations for sulphur and nitrogen compounds are not included. Thus, in this study we make the first attempt to run the MATCH-Europe model version for sulphur and nitrogen compounds. It is therefore important to examine the obtained results for the separate sulphur and nitrogen compounds first, before any summarized particle concentrations from the MATCH-Europe model are presented and discussed. A limited model test is presented in Section 4.2.3 below.

## 4.2.1 Meteorological data

The dispersion model requires meteorological data to calculate transport, chemistry and deposition processes. For studies on the European scale all necessary meteorological data (wind fields, temperature, precipitation, friction velocity, sensible heat flux, the Monin-Obukov length and mixing height) were obtained directly or indirectly from the operational numerical weather prediction model HIRLAM at SMHI. Meteorological information from the earth's surface and the 7 lowest layers of the atmosphere, up to ca. 6.5 km height, was included.

#### 4.2.2 Emissions

Information on European emissions for the year 1993 of sulphur dioxide and sulphate (SO<sub>x</sub>), nitrogen oxides (NO<sub>x</sub>) and ammonia (NH<sub>3</sub>) has been obtained in a 50 x 50 km grid from the EMEP-programme (Meteorological Synthesizing Centre - West, the Norwegian Meteorological Institute). A general description of the EMEP emission data is given by Tuovinen et. al. (1994). These emission data have been interpolated to the different projection and 55 x 55 km grid of MATCH-Europe.

Emission data for black carbon caused by fossil fuel combustion, valid for a typical mid 1980s year, has been obtained from the GEIA global database, "A Black Carbon Emission Data Base for Atmospheric Chemistry and Climate Studies" by Dignon et. al. (1994), see also Penner et. al. (1993). The geographical resolution is 1 degree latitude by 1 degree longitude. These emission data have been interpolated to the different projection and 55 x 55 km grid of MATCH-Europe.

## 4.2.3 Test results - sulphur and nitrogen compounds

The MATCH-Europe model version has not included a combined sulphur/nitrogen chemistry in earlier applications. Therefore, before the results for particle concentrations are discussed, we make a minor separate presentation of the obtained results for sulphur and nitrogen concentrations in air. We also include some comparisons between obtained MATCH-Europe results and measurements from Scandinavian EMEP-stations. Also PIXE-measurements from Norrköping for a short period are included in the comparisons.

In order to get an idea of the relations of magnitude between the most important compounds, which are included in the calculations, we present in Figure 4.1 calculated time series for the location of Aspvreten for the November/December 1993 period. It is obvious that the concentrations of SO<sub>2</sub>-S, SO<sub>4</sub>-S, NO<sub>2</sub>-N and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>-S are substantially larger than concentrations of the other compounds.

A simplified model test has been performed through comparisons between obtained MATCH-Europe results and measured daily concentrations of SO<sub>2</sub>-S, SumSO<sub>4</sub>-S (= SO<sub>4</sub><sup>2-</sup> + (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>/NH<sub>4</sub>HSO<sub>4</sub>), NO<sub>2</sub>-N, SumNO<sub>3</sub>-N (= NO<sub>3</sub><sup>-</sup> + NH<sub>4</sub>NO<sub>3</sub> + HNO<sub>3</sub>) and SumNH<sub>x</sub>-N (= NH<sub>3</sub> + NH<sub>4</sub>NO<sub>3</sub> + (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>HSO<sub>4</sub>) at Scandinavian EMEP-stations for the November/December 1993 and March/April 1994 periods. The Sum-compounds have been used since the measurements are available in that form. In Figures 4.2-4.3 examples are given for comparisons at the EMEP-station Rörvik, on the Swedish westcoast, and in Figure 4.4 comparisons at the EMEP-station Aspvreten on the Swedish eastcoast. The general impression from the Figures 4.2-4.4 and from comparisons for all other Scandinavian EMEP-stations is

that the correlation and agreement between model results and observations are fairly good. However, since this is the first application made for this model version, it is plausible to assume that improvements can be made in the future. The agreements between model results and observations for almost all Scandinavian EMEP-stations are especially good for SumNH<sub>x</sub>-N. This is somewhat surprising, since the emission data were assumed to be rather uncertain for NH<sub>3</sub>. At some stations close to large urban areas the SO<sub>2</sub>-concentrations seem to be overestimated and the SO<sub>4</sub>-concentrations underestimated in MATCH-Europe. This might to some extent be caused by the coarse resolution with 55 x 55 km gridsquares and interpolated emission data. For SumNO<sub>3</sub> there is a systematic and rather substantial underrating and also NO<sub>2</sub>-concentrations seem to be somewhat too low. This is obviously something that should be carefully investigated in the future. Here we only establish that the model at present gives too low nitrate values and thus underrates the particle-mass being associated with nitrate.

We have also made a comparison between MATCH-Europe calculations of SumSO<sub>4</sub>-S and particle measurements based on the PIXE-technique for the city-centre of Norrköping. The results are presented in Figure 4.5, where three curves are given:

- PIXE-S showing measured sulphur
- PIXE-rest showing the total mass from the following compounds Al, Si, P, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Y, Nb, Mo, Cd, Sn, Hg, Pb
- MATCH-S showing the MATCH-Europe calculated SumSO<sub>4</sub>-S concentrations.

It is quite interesting to notice that the PIXE-results and MATCH-calculated sulphate-S concentrations in the centre of Norrköping have a fairly good correlation and also agree quite well in magnitude. The PIXE-analysed sulphate concentrations in the city centre are only slightly larger than simulated on a regional scale. For all the other compounds included in the PIXE-data, no model simulation have been made. However, it is obvious from the measurements that in most cases the air concentration of particulate sulphur alone is larger than the sum of the rest of all components analysed by PIXE. Due to practical arrangements for the sampling on filters in this case, it is plausible to assume that the cut-off for the PIXE samples was about 5 µm. This indicate that the additional particulate sulphur from local sources within the city of Norrköping in this case was small.

Model results for the particulate compounds, which will be used in this study as a basis for estimates of total regional particle concentrations, can be summarized as follows: Results for ammonium agree very well indeed, for sulphate the concentrations seem to be too low during some periods (e.g. Nov/Dec 1993 at Rörvik) but with a good agreement in other situations (e.g. Mar/Apr 1994 at Rörvik and in Norrköping), while nitrate seems to be substantially underrated by the model.

#### 4.2.4 Model assumptions for particulate mass

A quite extensive Regional Particulate Model (RPM) for North America, based on the RADM Eulerian atmospheric chemistry model, has recently been developed and is briefly described by Ching et. al. (1995). PM-10, particulate matter less than 10 µm, is one of several criteria air pollutants for which National Ambient Air Quality Standards (NAAQS) are established by the

United States Environmental Protection Agency to protect human health. In the RPM a rather complete description of the physical and chemical processes for particulate matter is included.

In the present study just a first tentative test of the possibilities to use the MATCH-Europe model for studies of regional formation and long-range transport of particulate matter is performed. Only particles with a size less than about 2.5 µm in aerodynamic diameter are in general included in long-range transport. Such particles are to a large extent products of atmospheric chemistry reactions of sulphur, oxidized nitrogen, reduced nitrogen and organic pollutants or primarily emitted "black carbon" particles. Direct emissions of black carbon particles mainly arise from burning of oil, coal and biomass. For the production of atmospheric organic particulate pollutants photochemical processes certainly play an important role. The fine particulate matter is mainly caused by anthropogenic emissions but also, especially for the organic pollutants, natural emissions can play an important role. The atmospheric lifetime of aerosol particles within the PM-2.5 size range is normally several days and can be transported thousands of km.

In the MATCH system, as described in Section 4.1.1 above, the atmospheric chemistry module can only treat production of particulate matter based on sulphur, oxidized and reduced nitrogen plus a separate model calculation for black carbon particles. For the sulphur and nitrogen compounds the relevant molecular weights are used for the estimates of the total particulate mass. The following compounds are included: ammonium sulphate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>HSO<sub>4</sub>), other sulphate particles (SO<sub>4</sub><sup>2-</sup>), ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>), other nitrate particles (NO<sub>3</sub><sup>-</sup>) and the two gases nitric acid (HNO<sub>3</sub>) and ammonia (NH<sub>3</sub>). These two gases are assumed to be absorbed in the particulate samples and for NH<sub>3</sub> on acid particles, although this might not always be the case. However, these gases are of minor importance for the total calculated mass concentration (see Figure 4.1) and this assumption is of rather little importance for the total calculated particulate mass. "Other sulphate" and "other nitrate" particles are assumed to be associated with compounds with a molecular weight similar to calcium. For black carbon only direct emitted mass is treated in the model calculations.

Thus, the MATCH-Europe model results for particle concentrations in air are based on simulations of only four basic compounds, namely: sulphate, nitrate, ammonium and "black-carbon". These are assumed, at least for most situations during the winter half-year, to be the main contributions to PM-2.5 in Sweden. The limitation of the particle size to PM-2.5 in the MATCH modelling is important to be aware of when comparing with measurements.

For the black carbon calculations the particulate matter has been separated into two fractions: a) newly emitted black carbon consisting of non-hygroscopic particles with dry deposition velocities similar to what is used for sulphate but with no wet deposition, and b) aged black carbon particles which are assumed to be hygroscopic and with the same wet and dry sink parameters as for sulphate. The degradation constant for black carbon from the first fraction is assumed to be  $4 \times 10^{-6}$  s<sup>-1</sup> and caused by atmospheric chemistry processes. This gives a residence time for the first non-hygroscopic fraction of about 3 days. This seems reasonable with regard to that Swedish measurements (Hansson, Westerholm and Kyrklund, 1997) indicate that weak-hygroscopic particles compose about 70 % of all particles in an urban area and about 30 % in rural air.

A complete photochemical module, where the organic compounds are included, is still not fully operational in the MATCH system and has not been applied in the present study. This means that organic particulate pollutants, frequently produced in e.g. ozone episodes during summer periods, are not included.

Also in the treatment of the aerosol dynamics a number of simplifications are made in the present version of the model. E.g. no bi-modal aerosol size distribution is included, thus the newly produced particles are treated in the same size mode as the aged particles and particles produced by cloud processes. Neither is the influence of the humidity on the particle behaviour included. We are well aware of the limitations of the present model. However, we still believe that also tests with the present very simplified model version can be of interest as a basis for a discussion of the local particle air pollution problem and also as a basis for future regional scale studies of particulate pollutants.

## 4.2.5 Results for fine particulate mass concentration

Presentation of November/December 1993 time-series for calculated regional black-carbon concentrations for Norrköping, Gothenburg and Aspvreten are given in Figures 4.6 - 4.8. No observations to compare with have been available, but the values are of the same order of magnitude as observed levels of "soot" at EMEP-stations in southern Scandinavia during 1994. On a regional scale over Sweden, both the present model results and measurements of PM-2.5 indicate that the total mass of sulphate, nitrate and ammonium particles dominates over the mass of black carbon particles with a rather large factor. Measurements indicate that this factor should be about ten. The general picture for rural air (Heintzenberg, 1989) indicate a composition of the particulate mass roughly as:

- inorganic salts ca. 50%

- black carbon (soot) ca. 5%

- organic compounds ea. 43%

- metals ca. 2%.

The model and measurement results, Figure 4.1, also indicate that the particle mass contribution from sulphate dominates over ammonium and nitrate.

In Figures 4.9 - 4.12 the MATCH-Europe estimated concentrations of PM-2.5, during the November/December 1993 period, are compared to the observed concentrations of PM-10 at Aspyreten, Norrköping, Gothenburg fixed site and Gothenburg mobile site. In general the model estimated PM-2.5 concentrations are lower than observed PM-10 concentrations. In a street in central Norrköping the model results are during most of the time much lower than observed PM-10. However, at some episodes in Norrköping and in general for the station at roof level in central Gothenburg and at the rural station Aspyreten, the model estimates of regional PM-2.5 concentrations constitute a larger part of the observed PM-10 concentrations. For Aspyreten the correlation between model results and observations is fairly good, with a correlation coefficient of almost 0.8, and the model calculated PM-2.5 mass is nearly half of the observed PM-10 values. This seems to be quite a realistic difference since data above (Heintzenberg, 1989) indicate that about 30% of PM-2.5 particulate mass in e.g. Swedish rural areas consists of organic compounds (not included in present model version) and an increase in particle size from PM-2.5 to PM-10 should probably increase the particulate mass with some 10 per cents. Measurements for Swedish urban areas, where such an increase should be expected to be larger, indicate that the particulate mass at roof level is roughly doubled (Hansson, Westerholm and Kyrklund, 1997).

#### 4.3 MATCH-Sweden

The MATCH-Sweden modelling system, with a horizontal grid-size of 20 x 20 km, covering Sweden, gives another possibility to estimate regional fine particulate (PM-2.5) concentrations over Sweden.

The operational MATCH-Sweden studies, which are performed yearly within the frame of the national environmental supervision, includes daily information regarding the particulate compounds sulphate, nitrate and ammonium. Thus, there is a possibility to use this archived information for attempts to estimate the most important parts of the regional scale particulate mass concentration over Sweden, although the information is not available in real-time. We give a brief presentation of MATCH-Sweden below and then show results from some estimates of fine particle concentrations over Sweden during a period of 1994.

MATCH-Sweden consists of three parts: 1) An objective analysis system for meteorological data, 2) the regional atmospheric dispersion model, briefly described above, including modules for emission, chemistry and deposition of sulphur and nitrogen compounds and 3) an objective analysis system for air- and precipitation chemistry data.

#### 4.3.1 Meteorological data

An objective meteorological analysis system is applied. The system makes use of routine meteorological observations to derive a number of parameters including wind fields, temperature, precipitation, friction velocity, sensible heat flux, the Monin-Obukov length and mixing height required by the dispersion model. The analyses are performed at three hour intervals. The precipitation analysis is given special attention: About 800 stations measuring daily precipitation is combined with precipitation and weather information from synoptic stations to give precipitation fields with three hourly time resolution and high horizontal resolution. Corrections for sampling losses and topographic effects are applied.

A high resolution data base for topography and land use has been mapped to the 20 x 20 km grid of MATCH Sweden. The data base provides topography, surface roughness and land use (fraction of forest, field, water, urban).

#### 4.3.2 Emissions

Information on Swedish emissions of sulphur dioxide (SO<sub>2</sub>), sulphate (SO<sub>4</sub>), nitrogen oxides (NO<sub>x</sub>) and ammonia (NH<sub>3</sub>) has been mapped to the 20 x 20 km grid of MATCH-Sweden reflecting the emissions for 1993 which have been used for the calculations regarding 1994. In general a one year delay in emission data has to be used in the operational air pollution survey studies performed by MATCH-Sweden.

#### 4.3.3 Long-range transport contribution

The dispersion model described above, combined with Swedish emission estimates and meteorological data provides daily estimates of concentrations in air and precipitation as well as dry and wet deposition of the simulated sulphur and nitrogen compounds. These results refer to contributions from sources within Sweden. To derive contributions to these quantities from sources outside Sweden the following method is employed: Model calculated daily contributions from Swedish sources are deducted from observed daily values of concentration in air and precipitation at background locations (about 15 air measuring stations in Sweden and the surrounding countries) on a point by point basis. The residual is termed long-range transport contribution. These residuals are analysed using an optimum interpolation method, where differences in observation quality can be accounted for, to give distributions of longrange transport contributions of concentrations in air and precipitation over the modelling domain. The basic idea behind this method is that the long-range transport contributions to the concentrations can be expected to vary more smoothly in space than the total concentrations which are affected to some extent by local sources, and should therefore be more suitable for interpolation. Considerable efforts have been spent on quality control of both input chemical observation data and resulting analysed concentration distributions.

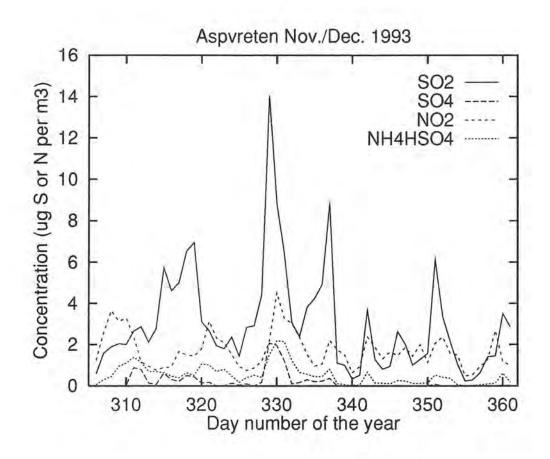
#### 4.3.4 Model assumptions for particulate mass

The particulate mass concentrations have, in this case, been based on information on sulphur and nitrogen particulate compounds already available from earlier MATCH-Sweden studies. The recalculations to obtain particulate mass concentrations were performed in the same way as described for MATCH-Europe, except that the black carbon contribution in this case was estimated through a simple assumption of proportionality to sulphate concentration. A factor 0.5 times the SumSO<sub>4</sub>-S concentration was assumed to give the black carbon contribution. This factor was determined in a rough way from a comparison to the results obtained with MATCH-Europe for black carbon.

#### 4.3.5 Results for fine particulate mass concentrations

In Figure 4.13-4.15 we present estimates of PM-2.5, based on the MATCH-Sweden results for January-March 1994, which are compared to available measurements of PM-10 at Aspvreten, Norrköping and Gothenburg. The MATCH-Sweden estimated PM-2.5 values are lower than measured PM-10 values, especially in the cities of Norrköping and Gothenburg. This difference is what can be expected, since the estimates based on MATCH-Sweden only include PM-2.5 while measured data include also larger particles up to PM-10. Especially in cities, with important local sources, these differences can be expected to be large. However, the correlation between MATCH-Sweden estimates for PM-2.5 and measurements of PM-10 for the rural site Aspvreten is fairly good.

MATCH-Sweden has been run operationally for the years 1991, 1994 and 1995. The delay in data from real time is about one and a half year. For the studied years, daily values of concentrations in the air over Sweden of sulphate, nitrate and ammonium are available at SMHI and can be used as a basis for estimates of daily regional scale PM-2.5 mass concentrations over Sweden. This method is rather simple to perform, but has the drawback that only 1 - 2 years old archived data can be used.



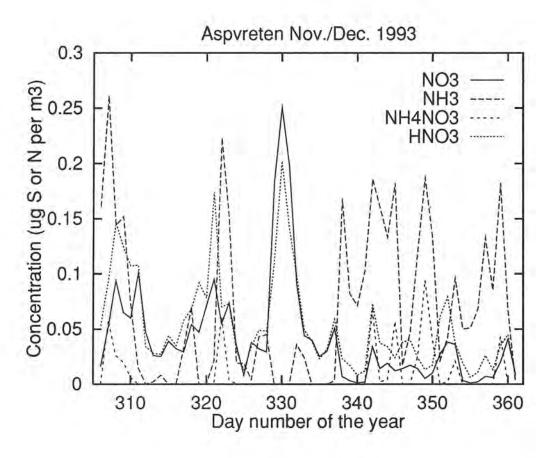


Figure 4.1 Time series showing daily mean concentrations of a)  $SO_2$ -S,  $SO_4$ -S,  $NO_2$ -N,  $NH_4HSO_4$ -N and b)  $NO_3$ -N,  $NH_3$ -N,  $NH_4NO_3$ -N,  $HNO_3$ -N (µg/m³) obtained from MATCH-Europe calculation for a period in November/December 1993.

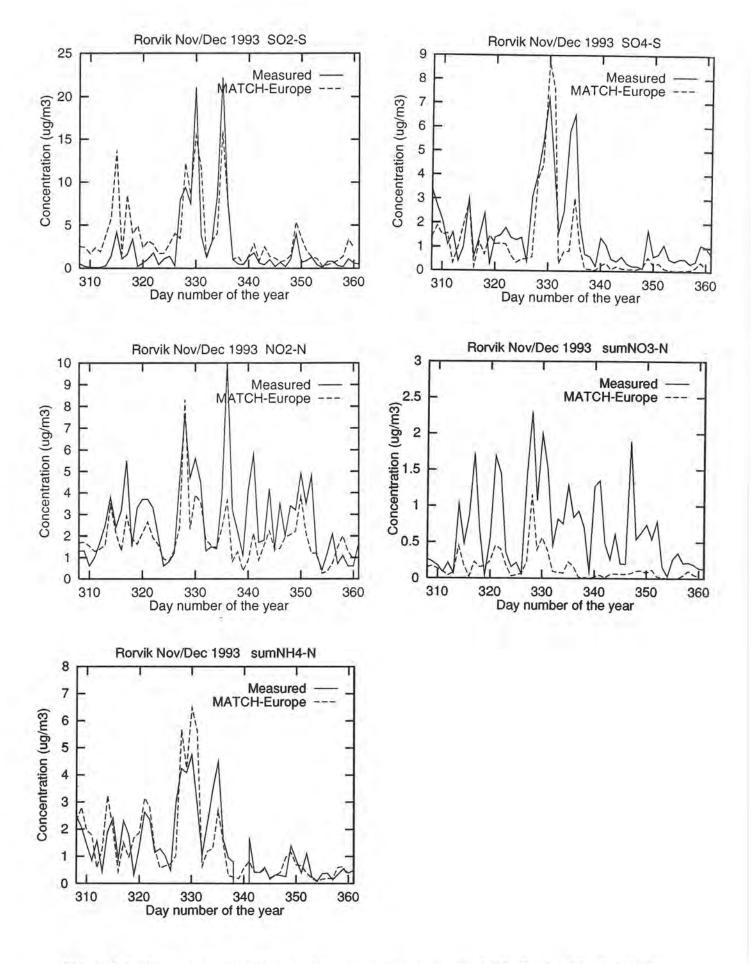


Figure 4.2 Time series showing daily mean concentrations of SO<sub>2</sub>-S, SumSO<sub>4</sub>-S, NO<sub>2</sub>-N, SumNO<sub>3</sub>-N and SumNH<sub>3</sub>-N (μg/m³) obtained from MATCH-Europe calculation for a period in November/December 1993 compared with measurements from the EMEP-station at Rörvik.

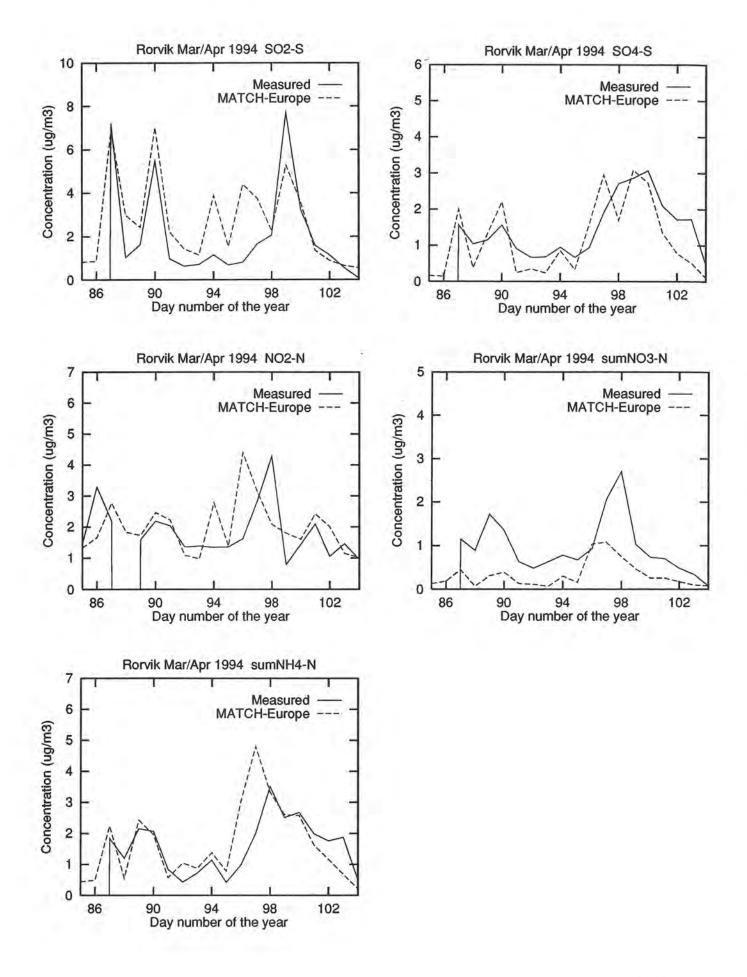


Figure 4.3 Time series showing daily mean concentrations of SO<sub>2</sub>-S, SumSO<sub>4</sub>-S, NO<sub>2</sub>-N, SumNO<sub>3</sub>-N and SumNH<sub>3</sub>-N (µg/m<sup>3</sup>) obtained from MATCH-Europe calculation for a period in March/April 1994 compared with measurements from the EMEP-station at Rörvik.

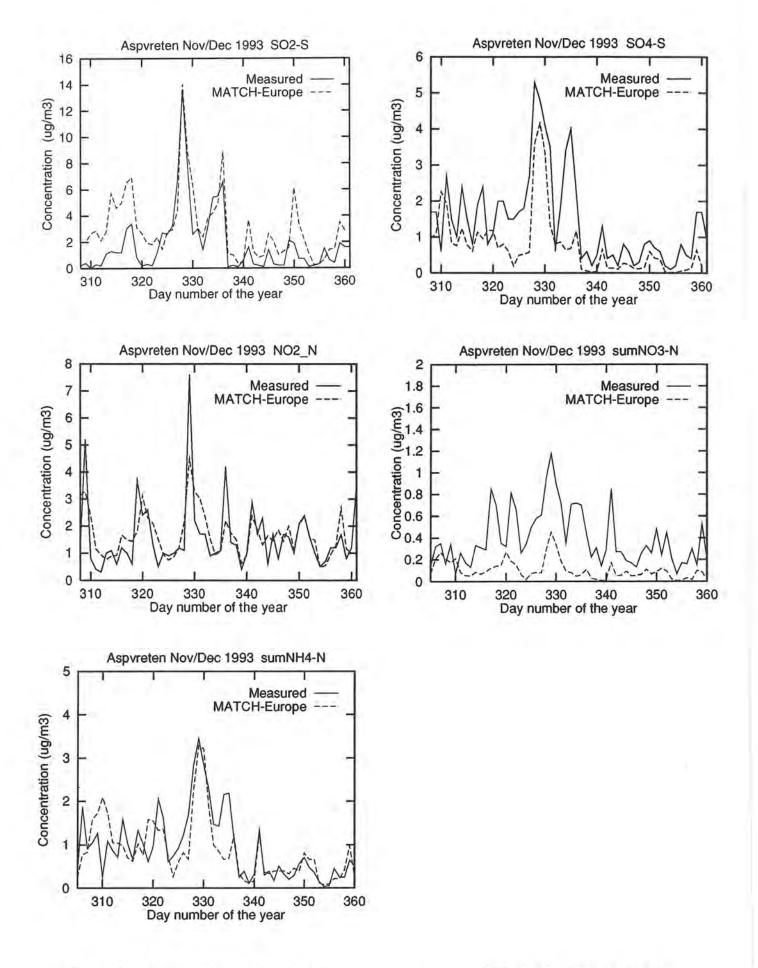


Figure 4.4 Time series showing daily mean concentrations of SO<sub>2</sub>-S, SumSO<sub>4</sub>-S, NO<sub>2</sub>-N, SumNO<sub>3</sub>-N and SumNH<sub>3</sub>-N (µg/m<sup>3</sup>) obtained from MATCH-Europe calculation for a period in November/December 1993 compared with measurements from the EMEP-station at Aspyreten.

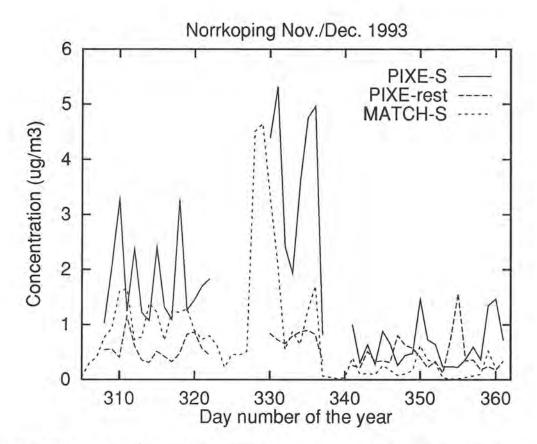


Figure 4.5 Time series showing daily mean concentrations of SumSO<sub>4</sub>-S (µg/m³) obtained from MATCH-Europe calculation for a period in November/December 1993 compared with PIXE-sulphur and PIXE-other\_compounds measurements from central Norrköping (see text for explanation of other compounds).

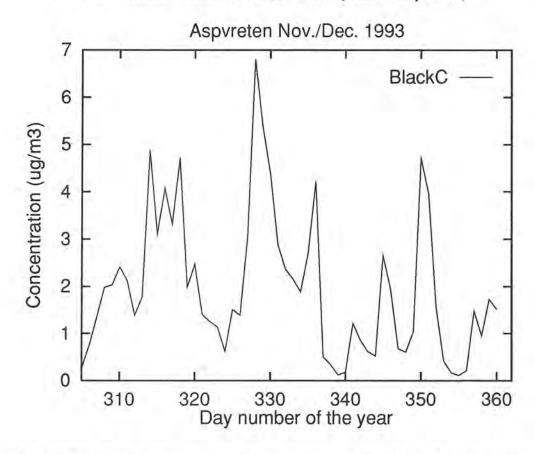


Figure 4.6 Time series showing daily mean concentrations of black-carbon (µg/m³) at Aspvreten obtained from MATCH-Europe calculation for a period in November/December 1993.

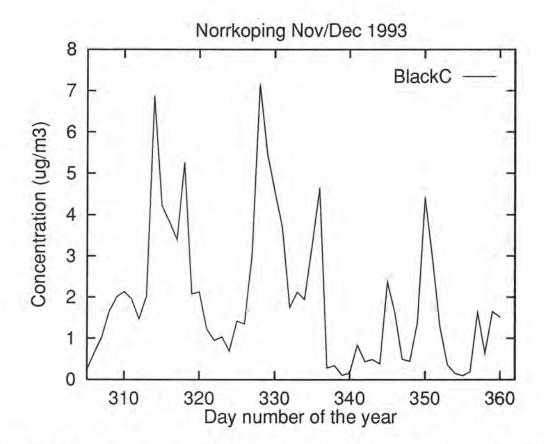


Figure 4.7 Time series showing daily mean concentrations of black-carbon (µg/m³) in Norrköping obtained from MATCH-Europe calculation for a period in November/December 1993.

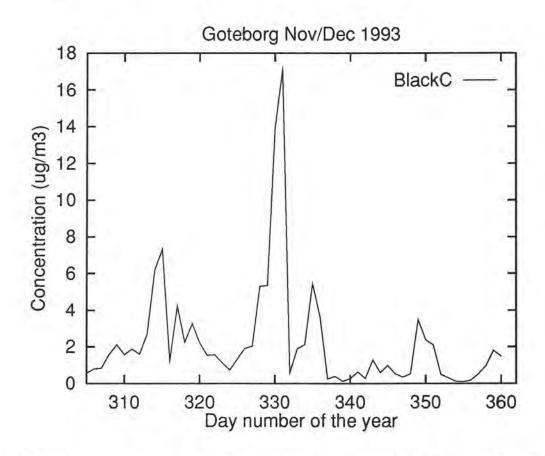


Figure 4.8 Time series showing daily mean concentrations of black-carbon (µg/m³) in Gothenburg obtained from MATCH-Europe calculation for a period in November/December 1993.

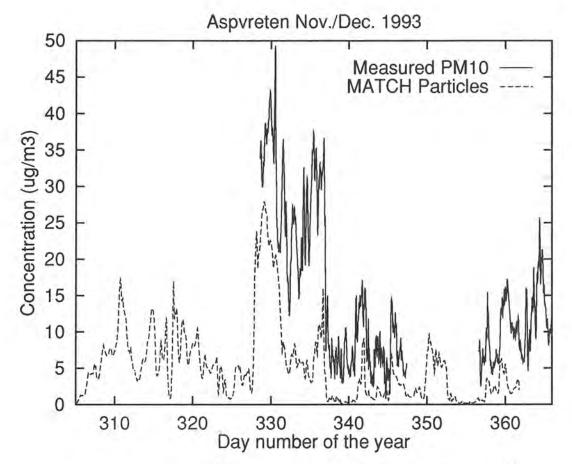


Figure 4.9 Time series showing daily mean concentrations of PM-2.5 (µg/m³) obtained from MATCH-Europe calculation for a period in November/December 1993 compared with measurements of PM-10 at Aspvreten.

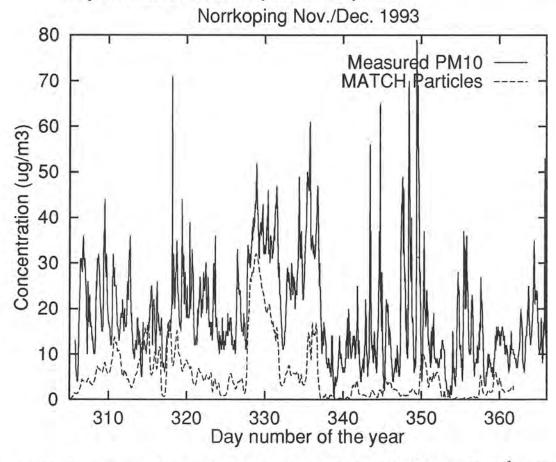


Figure 4.10 Time series showing daily mean concentrations of PM-2.5 (µg/m³) obtained from MATCH-Europe calculation for a period in November/December 1993 compared with measurements of PM-10 for central Norrköping.

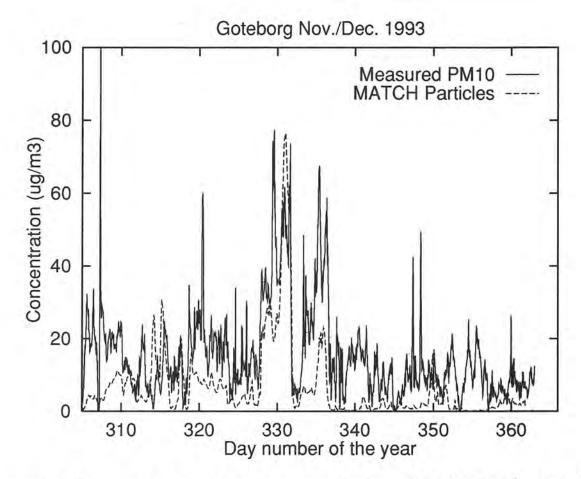


Figure 4.11 Time series showing daily mean concentrations of PM-2.5 (µg/m³) obtained from MATCH-Europe calculation for a period in November/December 1993 compared with measurements of PM-10 for stationary station in Gothenburg.

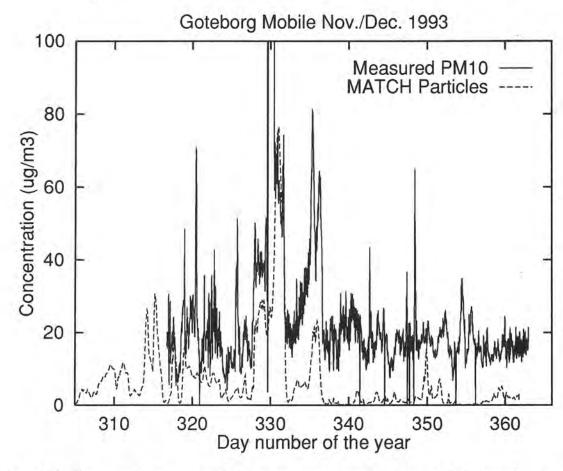


Figure 4.12 Time series showing daily mean concentrations of PM-2.5 (µg/m³) obtained from MATCH-Europe calculation for a period in November/December 1993 compared with measurements of PM-10 for mobile station in Gothenburg.

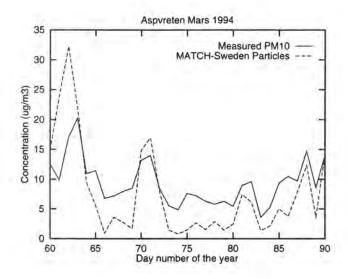


Figure 4.13 Time series showing daily mean concentrations of PM-2.5 (µg/m³) estimated from MATCH-Sweden for March 1994 compared with measurements of PM-10 at Aspyreten.

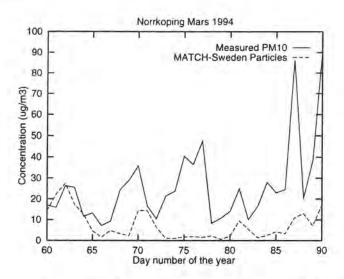


Figure 4.14 Time series showing daily mean concentrations of PM-2.5 (μg/m³) estimated from MATCH-Sweden for March 1994 compared with measurements of PM-10 for central Norrköping.

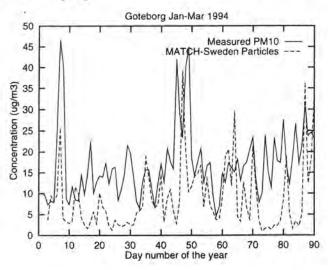


Figure 4.15 Time series showing daily mean concentrations of PM-2.5 (µg/m³) estimated from MATCH-Sweden for January-March 1994 compared with measurements of PM-10 for central Gothenburg.

#### THE LOCAL SCALE PM-10 MODEL

The local scale PM-10 model has many modules in common with SMHI's standard model for local air pollution calculations (Dispersion). The model has been complemented with preprocessor modules for new sources of meteorological data and modules for treatment of PM-10 particles.

#### Field of application

The local scale PM-10 model is a mathematical model intended for calculating dispersion of inhalable particles on a local scale up to 10-20 km away from the source. The model is intended for regulatory purposes but it may also be used for forecasting mass concentrations of inhalable particles. The model can be used in urban areas or near major industrial sources.

The basis for the model is: good access to surface observations and/or upper air observations; access to a method to describe turbulence parameters using those observations; access to an analytical method to solve the diffusion equation; access to detailed source inventories using known emission factors and access to a system for analyzing the results in a geographic information system.

The hourly mass concentration of particles is calculated in a large number of receptor points. The contributions to the mass concentration in a receptor point may emanate from one or several sources, described in the form of traffic statistics, heating plans or other inventories. The resulting mass concentrations during a period are treated statistically to yield air quality characteristics that can be compared to e.g. guideline values.

#### Working method

In principle, the model can be divided into three blocks: A) meteorological pre-processing, B) dispersion calculations and C) postprocessing. Each block is a separate program, which cooperates with the other blocks using a well defined interface.

The work flow starts in a meteorological data set with hourly observations of wind, temperature, precipitation, humidity, cloudiness, radiation, ground state, weather phenomena and other parameters.

The meteorological data are input to a meteorological preprocessor, which calculates turbulence parameters, i. e. friction velocity, buoyancy and Monin-Obukhov length. It also calculates different terms in the energy balance equation near the ground: net radiation, sensible heat flux, latent heat flux and ground heat flux. Surface resistance and aerodynamic resistance for deposition of particles may also be calculated.

Another set of input data to the calculation is the emission data. Typical emission sources may be large point sources with industrial emissions of inhalable particles, area sources like residential areas with wood or fossil fuelled heating. The sources are defined with position and source characteristics. Traffic is an important source with directly emitted particles and resuspended particles. Traffic can be described as line sources, area sources or street canyons. Traffic sources differ from other source types in the way that emission factors are included in the dispersion model. The traffic sources are described as predefined street types with characteristic diurnal variation and characteristic mix of vehicle categories.

These data, together with a description of the site, are input to the local scale dispersion model. The model calculates hourly mass concentrations of particles. A period of one month, one year or thirty years with different meteorological observations is looped through. The resulting mass concentrations during a period are treated statistically.

The data sets and methods that are included in a dispersion calculation will be described in some detail in the following sections. The results of the calculations will also be validated.

#### 5.1 Presentation of meteorological data

The SMHI network for synoptic meteorological observations includes 45 manual stations that report every 3 hours or more often. Another 115 automatic stations report every hour or more often. There are some other observation networks at SMHI for precipitation, temperature and sun radiation observations, which include some 250 stations. These stations report data less frequently and not always in real time.

During the PM-10 project investigation period October 1, 1992 to March 31, 1994, the observation networks have gathered 13128 hourly observations, which corresponds to 547 days. Some stations have gathered data during the entire period, while others have closed down for short periods.

The most important factors of transmission for pollution processes on local scale are: 1) wind direction, 2) effective height of sources, 3) wind speed, 4) dispersion, 5) mixing layer height, 6) dry deposition (aerodynamic and surface resistance), 7) wet deposition (precipitation) and 8) transformation. Some of the factors will be described for Norrköping and Gothenburg in the following sections.

#### 5.1.1 Norrköping

In Norrköping, the manual synoptic station at Bråvalla Airport was permanently closed from spring 1993. For the model, standard 1-hourly observations have been aimed at, primarily because the model normally uses such data. When Bråvalla was closed, then standard data from Malmslätt Airport near Linköping were therefore used.

There is also a solar station which reports sunshine duration, temperature, relative humidity, wind, global radiation, direct radiation, diffuse radiation and long wave radiation. The Norrköping City Environment Office also owns a meteorological mast at Kungsängen Airport. Sensors in the mast measure temperature, vertical temperature difference 2-8 m, precipitation, wind and global radiation. Data from the these stations have also been used to prepare meteorological data for the particle dispersion model.

An overview of meteorological data for the entire period is given in Figures 5.1 - 5.4. From wind roses (not shown here) it can be seen that the wind speed and direction observations from Bråvalla (including Malmslätt when data are missing in Bråvalla) and the solar station have quite similar frequencies of wind in the different main wind directions, while Kungsängen airport has less southerly and south-westerly winds and more easterly winds. Kungsängen airport also has a higher frequency of calm winds.

The seasonal variation in wind speed is shown in Figure 5.1. The wind speed is divided into seven wind speed classes, including calm winds. The overall pattern shows that high wind

speeds are more frequent in winter (December to March), but February 1994 had a high frequency of low wind speeds. The wind speeds were usually lower at Kungsängen airport.

Figure 5.3 shows daily values of temperature, precipitation and wind speed for both winter seasons during the model test period. The data are taken from Malmslätt near Linköping since the most complete data are found there.

#### 5.1.2 Gothenburg

The regular synoptic weather station at Säve airport, near Gothenburg, was reporting the whole period, October 1992 - March 1994. These measurements were judged to be useful for representing the meteorological conditions of Gothenburg in the model.

#### 5.2 Turbulence pre-processor

One of the most important factors that influence pollution processes is turbulent diffusion. Turbulent diffusion is caused by internal friction forces in air near the ground or close to obstacles in the air flow. It is also caused by radiation heating the ground, which leads to turbulent mixing between rising thermals and ambient air. The turbulence intensity and static stability in the air can be calculated from meteorological data in a rural environment, instead of directly measuring it. The calculations are made with a program that is called a pre-processor for turbulence in dispersion calculations. The pre-processing is made with three different methods: 1) The profile method for mast measurements, 2) The resistance method for surface observations from a standard synoptic station, 3) The resistance method for surface observations from a solar station.

The methods have been evaluated by the European harmonization project COST-710. A final report from this evaluation will be published in 1998. The three methods are used to calculate the buoyant production/consumption of turbulent energy and the mechanical production of turbulent energy. The ratio between mechanical and buoyant turbulent energy determines which kind of turbulent flow that arises. The methods have been applied to the three different data sets in Norrköping, to compare different formulations of the profile/resistance method.

Figure 5.2 shows how the weather situations have been classified as stable, neutral or unstable each month with the three different methods. The atmosphere was most unstable in May and most stable in December 1992. It can also be seen that the surface observations from Bråvalla/Malmslätt were reduced to three-hourly terms during January and February 1992 at Bråvalla and during January - March at Malmslätt. The solar station also lacks some data during December 1992 and other months, while Kungsängen has practically every observation. Figure 5.4 shows the diurnal variation in net radiation at the different stations during the winter season 1992/93. The net radiation is defined as the balance of incoming short-wave radiation, outgoing reflected short-wave radiation, outgoing heat radiation and incoming heat radiation. The solar station measures all necessary components to calculate the net radiation. The synoptic station uses cloudiness to calculate global radiation and net radiation, whereas the mast station measures global radiation. However, the heat radiation is not measured with the mast, and the cloudiness is unknown from this station. Therefore, the mast station, in its current state, works only during daytime hours. We have chosen to use the surface observations from Bråvalla/Malmslätt for the calculations, because there are algorithms in the model that correct for lower observation frequency, and this gives a complete data set for the entire period. If the solar station in Norrköping had been chosen, we would have missed important data in December 1992.

The urban environment may have effects upon the stability and mixed layer height due to anthropogeneous heat emissions and forced mixing of the air near large buildings, which will increase the turbulence. These effects have been treated in Appendix 3 and could be used in the PM-10 model. An alternative approach has been chosen in the model, which is described in Section 5.4.

Precipitation is also treated in the turbulence pre-processor. It is in this algorithm that 12-hour accumulated precipitation is distributed over each hour between the precipitation reports. At synoptic weather stations, precipitation measurements are performed twice a day (06 and 18 GMT). This is the case for the meteorological data used, both for Norrköping and Gothenburg. However, hourly precipitation values are required to enable the PM-10 model to predict street surface moisture for every time step of concentration calculation. This problem is solved through introducing a scheme utilizing manually observed present weather (reported with a two-digit "ww"-code). An example could make clearer the method of this scheme: E. g. if a 18 GMT observation reports 2 mm of rainfall, the ww-code for 13 and 14 GMT say steady moderate rain, but no other hour reporting precipitation, then the 2 mm are distributed with 1 mm on each of the two hours with rain. In other cases, different hours report precipitation of different intensity. Then the 12 hour amount will by the scheme get distributed with a greater part of the total amount on hour(s) with heavier precipitation intensity, than on hour(s) with light rain.

#### 5.3 Source inventories for PM-10

The existing model of SMHI, Dispersion, which has been introduced as an essential part of the PM-10 model, consists primarily of a grid net model of at most 20x20=400 points where concentrations at roof level are calculated. Also, it is possible to calculate PM-10 within a single street canyon. Then, the hourly results of the grid model act as a "city background" concentrations to which the contributions from the street canyon are added. The model calculates the emission data to be used hour by hour, using input data of two kinds:

- A mapping of the traffic flows in different streets and roads
- A measure of the emission per vehicle.

These aspects will be discussed below.

Local sources to PM-10 are traffic sources like car exhaust gases, resuspended particles and stationary sources like district heating plants, industry and small combustion sources. Stationary sources have not been considered here. Both Norrköping and Gothenburg have a large share of district heating units with particle filters giving a small environmental PM-10 contribution. The contribution from small combustion sources are considered to be small in the street environment studied here. Local resuspension from natural areas as agricultural fields has also been disregarded.

#### Street traffic samples

Traffic data have been obtained from Norrköping and Gothenburg which have been treated here.

It is possible to subdivide traffic data into two groups:

- 1) Extensive data. That is routinely repeated 24 hourly traffic counts on many streets without any consideration of different vehicle types.
- 2) Detailed measurements, divided on vehicle weight classes and sometimes hourly data. This occurs only for a few streets during limited periods.

For streets in Norrköping studied according to 1), there have been traffic counts during periods of three days to one week, repeated after one to twelve months. The streets with monthly counts have been used to construct full-year traffic cycles also for other streets. Then, yearly data have also been utilized to get general absolute traffic levels.

The measuring equipments are either mobile or permanently installed. For detailed counts, which may include vehicle classes and driving speeds, are used dual tubes crossing the street combined with an analysis device. At G:la Rådstugugatan in Norrköping such detailed measurements have been made at the same place and time as the PM-10 measurements.

#### Model input on traffic flow

The traffic flow mapping will result in a model input to be briefly described before discussing the two model cities. The traffic data are introduced as line sources and area sources - see below. Larger streets or roads are suitably treated as individual line sources, while the smaller streets are put together geographically into area sources.

The line sources are divided into straight line segments, each of which is assigned a value on traffic flow (vehicles per day). More variables are needed, such as diurnal variation for different types of vehicles, yearly distribution, typical driving speed etc. If available data only consist of traffic flow, it is possible to use standard set-ups for the missing variables. The input is then denoted by the type of street: thoroughfare, city, residential, bus, industrial etc. The input for a specified street canyon is similar to a line source, complemented with the cross section geometry of the street canyon.

Different area sources can be given different size and shape. The model then transforms to a regular grid net of squares or rectangles. The input needed consists of traffic load within the rectangle (vehicle kilometres per 24 hours), complemented with other information as for the line sources.

#### 5.3.1. Source inventories for Norrköping

An inventory has been made of the traffic sources in Norrköping. To create traffic emission data, two kinds of basic information are needed:

- a traffic inventory used to insert the traffic load information into the model divided into line and area sources.
- 2. emission factors (emission per vehicle depending on category of vehicle).

The emission data for the grid point model are based on published traffic data (Norrköpings Kommun, 1993). A map of the traffic flow in the central city is given in Figure 5.5 and a map for the whole city was given in a status report (Bringfelt et al. 1995). The yearly traffic volumes have been distributed over the hours of the day, the days over the year and into different kinds of vehicles by standard patterns for different kinds of roads/streets.

In some of the PM-10 model runs, the standard patterns of traffic flow for different vehicle types have been replaced by more detailed information for the street canyons, where PM-10 concentration has been monitored. Here, there are often contemporary and detailed traffic data of daily and hourly resolution. Ten categories of vehicles are used regarding their weight, occurrence of catalytic converter, type of fuel etc.

In Norrköping, three streets have been run with the local PM-10 model, including the street canyon submodel, to compare with measured PM-10 data. In G:la Rådstugugatan, traffic counts have been made during three weeks within the periods of PM-10 measurements and partly also with the ACCU monitoring. The periods are 14 - 20 October, 4 - 10 November and 1 - 7 December 1993. The vehicles have been sorted into the categories MC, private car, private car with trailer, heavy vehicles and heavy vehicles with trailer. For each category has been measured the number of vehicles per hour and the mean driving speed (in total: 41 km/hr). Figure 5.6 shows the diurnal traffic counts for an average week. Over the whole measuring period, the private cars were 97 per cent and the total number of vehicles per 24 hours on Monday to Friday was 6058. Hörngatan (a continuation of G:la Rådstugugatan), with 7370 vehicles per 24 hours, is supposed to have the same time and vehicle type distribution as G:la Rådstugugatan. For both streets, the input to the model has been created with regard to these traffic data. For the third street, Kungsgatan, with 20600 vehicles per 24 hours, standard time and vehicle type distributions for a thoroughfare street have been used.

The traffic in the whole of Norrköping has been used. The streets have been used as individual line sources if they have at least 10000 vehicles per 24 hours. Smaller streets have been brought together to area sources of 500 x 500 metres associated to the central calculation area.

More distant suburbs have been grouped into line sources, the only source type accepted outside the central calculation area. On an overview map (Figure 5.7) of the environment of Norrköping is shown line sources and area sources describing the traffic load.

Traffic counts have been made in most major central streets and some minor streets. Thus most streets have not been counted and are responsible to a non-negligible part of the emissions. This contribution has been estimated by using a standard of 10000 vehicle kilometres per average 24 hrs and km² for a densely built up area. This corresponds to 500 vehicles per average 24 hrs in a street net of block size 100 x 100 metres. For more sparsely built up areas the traffic load has been reduced in proportion to the settlement density. For the central part of Norrköping a more detailed method has been used: The total length of all uncounted streets have been summed and multiplied by 500 vehicles per average 24 hrs.

#### 5.3.2 Source inventories for Gothenburg

For making traffic data for the model has been used a report supplied by the Traffic Office of Gothenburg (Göteborgs Stad 1995). This report, containing detailed traffic data, for every street gives the traffic counts collected in different years. Since the present data will refer to the period October 1992 to March 1994, data from 1993 have primarily been used. For streets with this year missing, have been used data from the year nearest in time, whereas, if necessary, a correction to hold for 1993 has been made.

The processing of traffic data is similar to that for Norrköping. The traffic volumes have been distributed over the 24 hours of the day, the days of the year and of the classes of vehicles. This has also been made for the standard street types, i. e. treating thoroughfare streets as line sources and small city streets as central city area sources. Streets and roads with more than 10000 vehicles per 24 hours have been assigned to line sources, while the smaller streets have been assigned to 1 km by 1 km square area sources.

The fine mesh street network is not very well mapped with traffic counts. Therefore, a standard treatment has been carried out, similarly as for Norrköping. Here the value 1000 vehicles per 24 hours has been used, corresponding to 20000 vehicle kilometres in a densely built up 1 km by 1 km square with city blocks of 100 by 100 metres. A reduction for less densely built up areas has been made in proportion to the estimated total street length within the square.

The calculation area comprises 12 km by 12 km. The estimated area sources outside this area, have been transformed to line sources in the same way as for Norrköping. In Figure 5.8 have been indicated the calculation area and the suburbs treated in this way.

#### 5.3.3 Emission factors for resuspension

The emission of PM-10 particles from vehicles is partly described as a direct emission from exhaust pipes, and partly as a vehicle related resuspension of tyre particles, pave wear and other road dust from anti-skid material (sand and salt). The road dust is accumulated during the winter season, in pace with anti-skid treatment and the use of studded tyres. The resuspension of road dust is limited by moisture in the street.

It is possible to describe the combined effect of vehicle related direct emissions and resuspension of road dust as a total emission factor for vehicles:

$$e_f^{tot} = e_f^{direct} + F_{qe}^*(e_f^{dust}),$$

where  $e_f^{direct}$  is the direct emission from vehicle exhaust pipes, described in Section 5.3.4,  $e_f^{dust}$  is an emission factor for road dust particles, which depends on the use of studded tyres and anti-skid treatment.  $F_{qe}$  is a resuspension factor that acts to reduce the resuspension of particles in wet conditions and early in the season, when the road dust depot is small.

The resuspension is related to the vehicle generated turbulence. We have chosen to describe resuspension and direct emissions as a combined emission factor for each vehicle.

For cars, the resuspension is proportional to the square of vehicle speed, but for trucks, it is proportional to the square root of vehicle speed, according to measurements by Sehmel (1984). The resuspension is equal from cars and trucks at the speed 110 km/h.

Larssen (1991) has made measurements of resuspension from one road in Trondheim with 35000 vehicles per day at 70 km/h and 10% heavy traffic. The total resuspension in Trondheim was 3 g/km per vehicle, measured in April when the road dust depot was large.

With this measurement and Sehmel's assumption, we have calculated an emission matrix for resuspension from cars and trucks at different speeds, which is called  $e_f^{dust}$ , see Tables 5.1 a and b.

Table 5.1 a Emission factors for resuspension of road dust from cars at different vehicle speeds.

Speed interval [km/h]	Emission factor [g/km]	Emission [mg/s]
0 - 19	0.05	0.1
19 - 26	0.28	1.8
26 - 32	0.47	3.8
32 - 41	0.75	7.6
41 - 50	1.16	14.6
50 - 78	2.29	40.7
78 - 90	3.95	92.1
90 - 102	5.16	137.5

Table 5.1 b Emission factors for resuspension of road dust from trucks at different vehicle speeds.

Speed interval [km/h]	Emission factor [g/km]	Emission [mg/s]
0 - 3.8	0.89	0.5
3.8 - 11.3	1.77	3.7
11.3 - 18.8	2.50	10.4
18.8 - 26.3	3.06	19.1
26.3 - 33.8	3.54	29.5
33.8 - 41.3	3.95	41.2
41.3 - 48.8	4.33	54.1
48.8 - 56,5	4.68	68.2
56.5 - 63.8	5.00	83.3
63.8 - 71.3	5.30	99.4
71.3 - 78.8	5.59	116.5
78.8 - 86.3	5.86	134.7

The meteorological conditions during Larssen's resuspension measurements are not known to us. The moisture on the street is a factor that influences resuspension very much. Therefore, it is probable, that the highest possible resuspension factors are higher than those listed above. Our comparisons between PM-10 measurements and model results indicate that the highest resuspension rate may be higher. We have used emission factors that are twice as high as the ones listed above. Claiborn et al. (1995) give an emission factor of 6.7 g per vehicle kilometre for two-lane roads with daily traffic less than 10000 vehicles, stated to exceed the value 3.7 "given by current formulae". This also gives some support for using a higher value than given in Table 5.1 a. Claiborn et al. also list resuspension factors for other road types. A major highway has lower emission factors and an unpaved road has much higher.

Section 5.4.1 will describe the moisture modification factor and depot factor, which act to reduce the resuspension of particles.

#### 5.3.4 Emission factors for exhaust pipes

Larssen (1991) has made a literature review over emission factors for inhalable particles from petrol and diesel fuelled vehicles. The main bulk of the particle emission from fossil fuel burning is considerably smaller than  $10~\mu m$  in diameter. Particles in e.g. diesel exhaust has the typical diameter 0.2 -  $0.7~\mu m$ . The emission factors are presented for different vehicles in the unit mg/km travelled in Table 5.2.

Table 5.2 Emission factors for inhalable particles from cars and trucks.

Vehicle category	Emission factor [mg/km]
Petrol fuelled catalyst car	5
Petrol fuelled car (leaded petrol)	20-40
Diesel fuelled car (<3.5 ton weight)	450
Diesel fuelled truck (3.5 - 10 ton weight)	750
Diesel fuelled bus/truck (10-20 ton weight)	1500
Diesel fuelled truck (>20 ton weight)	2000

The table shows a relatively large variation in the emission factor values. The car type and technical standard of the individual vehicle has great influence over the real emissions. The speed and acceleration also has influence on the emissions. These factors give a great uncertainty in the use of emission factors for the entire set of vehicle categories in a city.

The emission factors for trucks are based on measurements in Sweden.

#### 5.4 The local dispersion model

SMHI's standard model for local air pollution calculations (Dispersion) has been developed in 1993. The model is validated in a set of European workshops: "Next generation of dispersion models in Europe", where different local dispersion models are compared and recommendations are given for harmonisation of the models. The model can be used for different pollutants, i.e. nitrogen oxides, sulphur dioxide, volatile organic hydrocarbons, carbon monoxide, inhalable particles and others. The model transfers traffic statistics, heating plans and other surveys into hourly air quality characteristics. In an hourly loop, meteorological data and turbulence data are read, actual emission is evaluated and mass concentrations are calculated. Each day, month, half and full year is described in statistical terms. A flow chart of the model is discussed and presented in Appendix 2.

#### Urban surface parameter module

During the project, we have developed a new module for the urban atmosphere, see Appendix 3. The module calculates the energy balance at the urban site with an extra term for anthropogeneous sensible heat flux and net radiation correction for a slightly higher temperature in a city. The stability is recalculated and an urban wind profile based on the new conditions is made. We have so far found that the mixing layer height is too high and that the lowest wind speed is too high.

Therefore, we have chosen another approach to the urban atmosphere, which is recommended by Hanna (1992). Because of uncertainties in the anthropogeneous heat flux and the strong influence it has on the heat balance at ground level, it is recommended that the anthropogeneous heat flux should not be used. Hanna also recommends that a lower limit on the Obukhov length should be set, so that the most stable cases are replaced with more neutral conditions in the urban atmosphere. We have followed this recommendation and set a lower limit of 25\*(the roughness length) as urban stability. This has been used in the calculations.

#### 5.4.1 Resuspension model

Two kinds of resuspended particles are treated:

- particles from street sanding and salting.
- particles released from the pavement and tyres, primarily by the action of studded tyres.

The emission factors for resuspension have been described in Section 5.3.3. The first source occurs mostly in winter and spring, the second source in winter but also weakly in summer. A model for resuspended particles has been constructed calculating the PM-10 concentrations from these two sources. The source strength for resuspension is regulated by the two corresponding particle depots on the street. It is also reduced by functions depending on the moisture amount in the depot. A function, using current weather observations, for wetting or drying the depot, and for determination of sanding/salting occasions is used for modifying resuspension.

#### Description of some variables:

g Moisture amount in the street dust and on the street surface (mm)

rr Hourly precipitation (mm)

gred Hourly reduction factor for g
epot Hourly potential evaporation (mm)

t Temperature (C)

td Dew point temperature (C)

rf Hourly runoff (mm)

frf Hourly decay of street dust depot due to runoff by rainfall (interval: 0.95-1.00)

ww Actual weather (according to meteorological code)

anskid Accumulated number of days with slippery conditions - where sanding/ salting is assumed to be made - during the current winter season.

skiddate Date for the previous sanding/salting occasion

tyre Ratio of accumulated depot of tyre and road particles to the highest depot at the end of the winter season. (0.0 - 1.0)

g is calculated successively each hour:

$$g = min(1.0, g + rr) * gred$$
 (5.1)

Here, occurring precipitation rr is added and the sum is minimized to 1.0 mm. g is reduced hourly by the factor gred, using the potential evaporation epot obtained in kg of water vapour during each hour:

epot = 
$$3600 * \{ delta * rn + rho * cp * \Delta e / ra \} / \{ L (delta + gamma) \}$$
 (5.2)

epot is calculated by the Penman Formula (see e. g. Houghton 1985) giving potential evaporation as dependent on incoming net radiation rn (W m<sup>-2</sup>), saturation vapour pressure deficit  $\Delta e = esat(t) - esat(td)$  (HPa) and the aerodynamical resistance ra (s m<sup>-1</sup>). delta is the derivative of saturation vapour pressure with respect to temperature: d(esat (t))/dt (HPa K<sup>-1</sup>), gamma is the psychrometer constant (HPa K<sup>-1</sup>), L is the latent heat of vaporization (J kg<sup>-1</sup>), rho is the density of the air (kg m<sup>-3</sup>) and cp its heat capacity (J kg<sup>-1</sup> K<sup>-1</sup>).

gred is calculated by

$$gred = exp(-k*epot/24)$$
 (5.3)

obtained by solving the budget equation:

$$-dg/dt = g * em / gm$$
 (5.4)

where gm is maximum moisture amount and em is evaporation rate when g = gm. em, corresponding to potential evaporation epot, gives quicker drying out in spring and summer than in autumn and winter. k is an empirical factor, found to be k = 0.075 by studying particle concentrations during several days without precipitation.

The hourly water runoff rf calculated as

$$rf = max ( (g + rr) * gred , 0 )$$
 (5.5)

is used to get the hourly dust depot decay factor frf due to runoff. If there is no precipitation or if runoff rf is smaller than 2 mm, there is no decay, and frf is put to 1.0. For rf between 2 mm and 10 mm, then frf is interpolated by

$$frf = 1 - 0.05 * (rf - 2) / (rfmax - 2)$$
 (5.6)

and for rf greater than rfmax=10 mm, then frf is put to 0.95. Thus for small or no runoff there is no hourly reduction of the dust depot (frf = 1.0). If runoff is large, there is at most a reduction by a factor frf = 0.95.

Increased moisture in the street dust depot should reduce the source strength for resuspension. This reduction factor, called fq, has been derived from earlier studies in Nyköping as

$$fq = 1 - 0.9273 * g ag{5.7}$$

Both rainfall and snowfall are considered as active in reducing resuspension. The hourly relative decay of the dust depot due to reduction by resuspension is calculated from

$$fsusp = 1 - decay * fq$$
 (5.8)

The two main kinds of resuspended particles mentioned above are now treated:

Sanding and salting.

If a day has rainfall or snowfall (as given by the hourly meteorological observations) and if temperature is between -2 C and +1 C and if the street is considered as moist (fq < 0.1)

anskid is increased by one sanding/salting occasion. This is coded as

$$anskid = anskid + 1 (5.9)$$

corresponding to a 5 per cent increase in sand/salt depot. Every hour anskid is reduced due to rainfall (frf) and weakened dust depot (fsusp):

$$anskid = anskid * frf * fsusp$$
 (5.10)

Action by studded tyres.

In a summer day, where anskid = 0, only a small amount of material is considered to be released by friction between tyres and pavement:

$$tyre = tyre + 0.001$$
 (5.11)

In winter, the increase is considered to be tenfold due to studded tyres:

$$tyre = tyre + 0.01 \tag{5.12}$$

This means that the depot of tyre and road material can be filled in 100 days, which is about the length of a winter season.

Every hour, tyre is reduced in the same way as anskid:

$$tyre = tyre * frf * fsusp (5.13)$$

When the winter season is finished (the 1:st of May used here), anskid is put to zero and tyre is reduced by the factor 0.1, both due to actions of cleaning the streets.

Finally, the factor fqe describing the action of resuspension - to be multiplied by the source strength in vehicle-kilometres - is then calculated as follows:

$$d = resusp\alpha * tyre + resusp\beta * min (anskid, 20)/20$$
 (5.14)

d is reduced to 1 if d becomes greater than 1. The number of sanding occasions is normalized by 20, whereas the relative source strength becomes unity. Observe above that anskid is reduced due to losses by resuspension and runoff.

Reduction of resuspension due to moisture:

$$fqe = d * fq ag{5.15}$$

resusp $\alpha$  and resusp $\beta$  may be termed as form factors for studded tyres and sanding/salting respectively: resusp $\alpha$  is the share of tyre and road particles in the total road dust depot at the end of the winter season, resusp $\beta$  is the corresponding share for sand/salt particles. Their sum usually equals unity, but in a city without studded tyres resusp $\alpha$  can be zero or small without increasing the value of resusp $\beta$ . In a city where sanding/salting is also absent both factors are small.

For the model runs in both Norrköping and Gothenburg, we used  $resusp\alpha = 0.5$  and  $resusp\beta = 0.5$  assuming similar contributions from tyres and sanding/salting. Possibly, the relative values of  $resusp\alpha$  and  $resusp\beta$  in the model, could be determined, using as main source direct measurements of typical tracer elements.

For Gothenburg, Johansson (1996, paper II, p 12) found that "the contribution from the pavement wear and the de-icing salt to the road dust mass are approximately equal". His data also indicated, that the pavement wear contributed about 5 times more than the tyre wear. In the model test for Gothenburg, his results could well be used. For a first comparison of the results, it was preferred here to have the model versions for Norrköping and Gothenburg as similar to each other as possible.

The factor decay above, for the hourly decrease of the particle depot will act primarily in dry conditions (g = 0). Equations 5.8, 5.10, 5.14 and 5.15 together form a budget for the sand/salt depot (anskid), that is, they form the solution of an equation of the same type as Equation 5.4. The same is valid for the depot of tyre/road particles.

It is highly recommendable to start a run with the resuspension model before the cold season on, say, October the 1:st, using small initial values of the depot variables anskid and tyre. This is so because, at the start of the cold season, the depots are known to be small. When running, the model itself modifies them successively each hour. Suitable depot values may also be used to initialize a model run on, say, February the 1:st. However, the results will then be more uncertain, because it is difficult to estimate the depots at this time, when their values are usually rather large.

#### 5.5. Results from the local model

Figures 5.9 a-c and 5.10 a-c show some details concerning resuspension variables for the runs made in the cold seasons 1992/93 and 1993/94 respectively. The value 0.001 has been used below for the important variable decay in Equation 5.8. The tables show that the number of sanding/salting occasions (anskid) was calculated to be much larger in the latter cold season than in the former. This is because the precipitation events occurred to a larger degree in cold weather in 1993/94, see Figures 5.9 a and 5.10 a. This is a reason why the two cold seasons are different from each other. In 1993/94, the resuspension factor grows to higher values than in 1992/93, giving higher values of resuspended PM-10 concentration, see below.

Figures 5.12 and 5.13 show comparisons of PM-10 concentrations calculated by the local model with the difference between measured urban values in Norrköping and measured rural values in Aspvreten. These differences are considered to be comparable to the results from the local model, since they should represent the local contribution only. However, it is then supposed, that there are no local PM-10 sources in Aspvreten. If such sources occur, one could sometimes expect negative urban minus rural differences. Such negative values occur, as can be seen, but they can also result from measuring errors. These problems should be kept in mind in adjusting the local model to the differences.

The value of decay has been adjusted to the daily measured urban - rural concentration differences on Hörngatan and G:la Rådstugugatan in Figures 5.12 and 5.13 respectively.

Other parts of the model have also been adjusted to get good agreement with the measured differences:

- 1. The source strength for resuspension, derived by Larssen (1991) from data in Trondheim, has been multiplied by two, see Section 5.3.3.
- 2. The coefficient k (Section 5.8), for the decay rate of moisture in the street dust, was found to be k = 0.075 in comparing increase of resuspended PM-10 values in a dry period.
- 3. The lowest possible wind speed in urban Norrköping, used by the model when calm conditions are given at the airport, is found to be important. Tests of an urban meteorological version of the local model, using a lowest urban wind speed of 1.3 ms<sup>-1</sup>, gave too low maximum values of PM-10. The standard meteorological version of the model Dispersion, gives weaker urban winds in calm conditions, in fact smaller than 0.5 ms<sup>-1</sup>. This, in combination with a rather high value of surface roughness length (0.8 m), leads to higher and more realistic predictions of the highest PM-10 values. This model version has been used for the data presented here.

The agreement in Figures 5.12 and 5.13 is encouraging with a good covariation. Before arriving at this reasonable agreement, the following test runs were made:

- A run was made for 1993-94 for G:la Rådstugugatan and the agreement was acceptable. However, when the same model version was run for 1992-93 at Hörngatan, the concentrations became only some 10 per cent of the measured differences. It was found that the value of decay was very critical to the result. The value used here was decay = 0.01.
- For this run, decay = 0.003 was used in order to avoid the too rapid decrease of the depots of tyre and sand material in the periods preceding the test period at Hörngatan, which had caused the low concentrations in run No 1. For G:la Rådstugugatan, the moisture in the depot was higher, considered to reduce the effect of changing the value of decay. The number by which anskid is normalized, was 10 as in run No 1. The resulting concentrations at Hörngatan increased markedly to some 50 per cent of the measured differences.
- To raise the concentrations further, decay was reduced to 0.001 and the normalizing number for anskid was raised to 20 in order to postpone filling of the depots until later in the winter. The latter should have some reducing effect on the concentrations counteracting the reduction of decay. However, for both streets/seasons this gave too high concentrations. In G:la Rådstugugatan, the sand depot still rose to its maximum in end of December, which is too early, causing high concentrations already in January and February. The value of decay could be increased to some 0.0015 to reduce the concentrations to better agreement in Figures 5.12 and 5.13.

In Figure 5.11, giving results at Kungsgatan using the local model adjusted above (decay = 0.001), data from Aspvreten are missing, so no urban - rural measured differences can be formed. However, comparison to the total urban measured values, shows that the calculated concentrations are too high in December 1992 - January 1993. Earlier tests with higher values of decay showed lower concentrations. However, for decay = 0.002, the calculated values were still more than twice as high as the total measured values. A reason for the overestimate may be the higher traffic intensity at Kungsgatan (some 20000 vehicles/day), than at Hörngatan and G:la Rådstugugatan (some 7000 vehicles per day). Thus, it is possible, that the model

overestimates the source strength of resuspension assuming direct proportionality to traffic intensity. Also, the street canyon dispersion used by the model, is unrealistic at the Kungsgatan measuring site, since the PM-10 measuring station was located more openly than in a regular street canyon: at a parking place on the west side of Kungsgatan.

Figure 5.14 compares the concentrations calculated by the local PM-10 model at roof level in central Gothenburg with measured concentrations. The resuspension model is the same as used for Norrköping, that is decay = 0.001. The source inventory and local meteorological data for Gothenburg presented above, have been used. Some high values are calculated in January and February 1994 which seem to agree well with measured values. The addition also of concentrations by the MATCH model will be shown in Section 6.

As discussed in Section 5.3.3 there are indications that the emission factors for resuspension should be higher than those given by Larssen (1991). Therefore, we have here used twice as high values as given by him. This may be a reason for the high values of PM-10 at G:la Rådstugugatan, Hörngatan and Kungsgatan in Norrköping calculated by the local model. At Kungsgatan a reason is also that we have used the street canyon dispersion submodel whereas the real site was located more openly. The parameter decay regulates the build-up of the dust depot and the shape of the resuspension concentration during primarily the cold season. Also the concentration levels are sensitive to the value of decay. More tests are needed studying primarily the values of the resuspension emission factor and the parameter decay, using existing and new PM-10 measuring data.

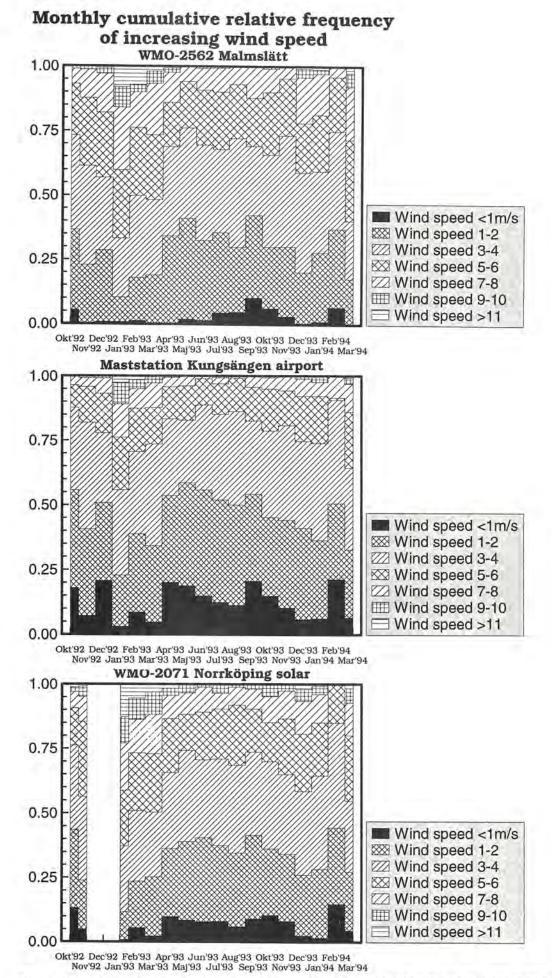


Figure 5.1. Monthly frequencies of wind speed at three sites for the whole test period.

Malmslätt includes data from Bråvalla.

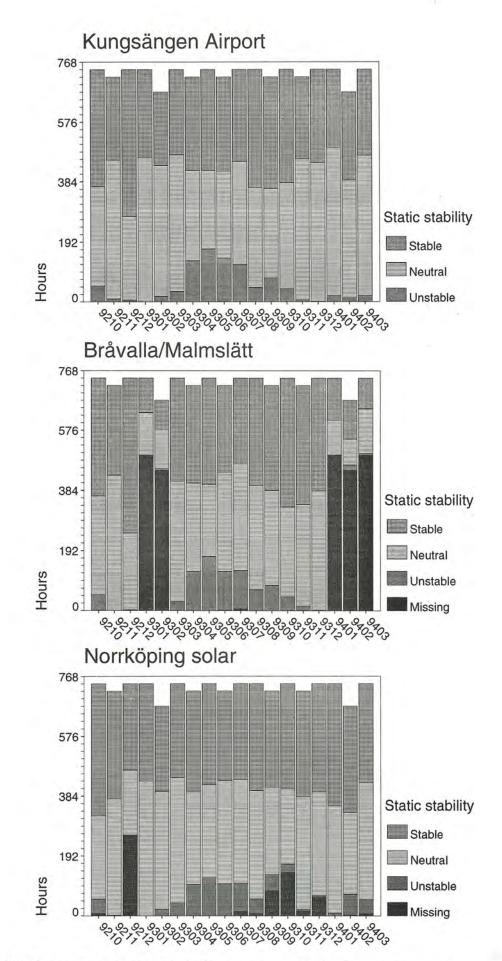


Figure 5.2. Monthly frequencies of static stability - stable, neutral and unstable stratification for three different data sets:

- Kungsängen Airport
- Bråvalla/Malmslätt
- Norrköping solar

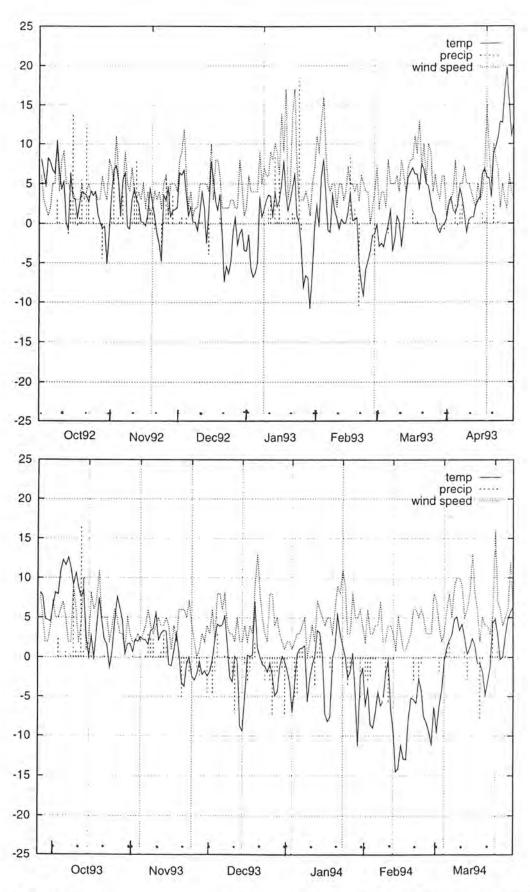
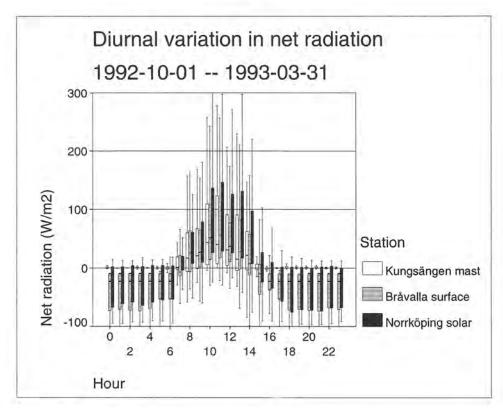
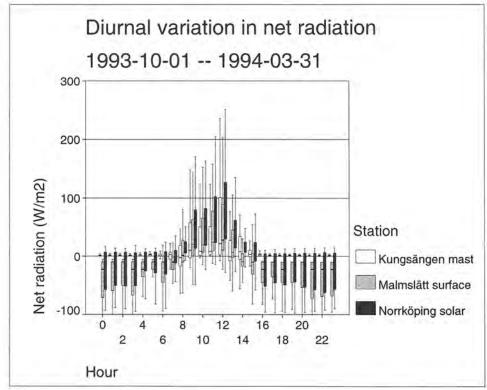


Figure 5.3. Daily mean temperature (C), daily precipitation (mm) and wind speed (ms<sup>-1</sup> at 13 UTC) at Malmslätt Airport near Linköping for the two cold seasons within the test period. The bars directed upward give rainfall and the bars downward give snowfall. The dots above the time axis indicate the 1:st, 11:th, 21:st etc of each month.





Diurnal variation in net radiation  $(W/m^2)$  at the three stations Kungsängen, Bråvalla and Norrköping solar. The bars show the interval between the 25-percentile  $(F_{25})$  and 75-percentile  $(F_{75})$ , with a mark for the median  $(F_{50})$ . As an extension from each bar, a "T-line" shows the highest/lowest observed value, excluding "outliers" (values further away from  $F_{25}$  and  $F_{75}$  than  $1.5 \times |F_{75} - F_{25}|$ ).

- Oct 1992-March 1993
- Oct 1993-March 1994

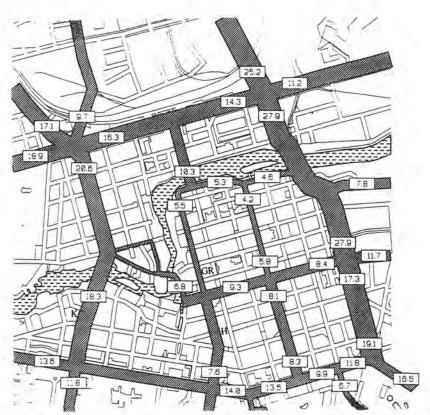


Figure 5.5. Map of major traffic flows in central Norrköping 1993. The figures are in thousands of vehicles per week day (Monday to Friday). Reference: Trafiken i Norrköping 1993.

The PM10 measuring sites considered here have been marked: Kungsgatan (K) Hörngatan (H) G:la Rådstugugatan (GR)

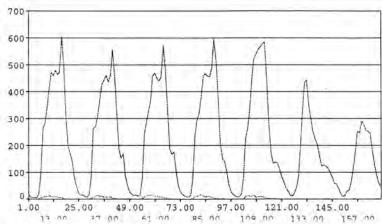


Figure 5.6. Hourly variation of the number of vehicles per hour at G:la Rådstugugatan in Norrköping during the average week of three weeks in October to December 1993. Only private cars (curve with large numbers) and heavy vehicles (curve with small numbers) are included.

Vertical axis: Number of vehicles per hour.

Horizontal axis: Hour during the week from Monday to Sunday.

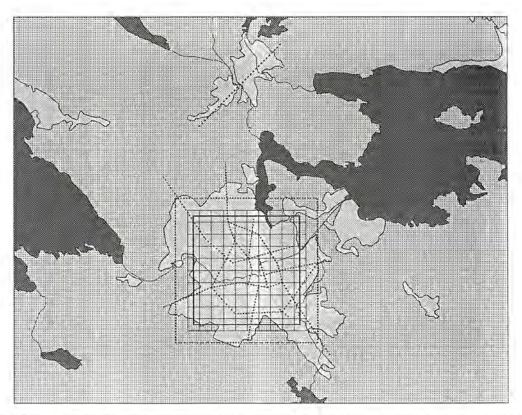


Figure 5.7. Line and area sources in Norrköping. The grid net shows the basic structure of area sources and the hatched lines give line sources. The sources are adapted to the concentration calculation area marked with a thick frame.

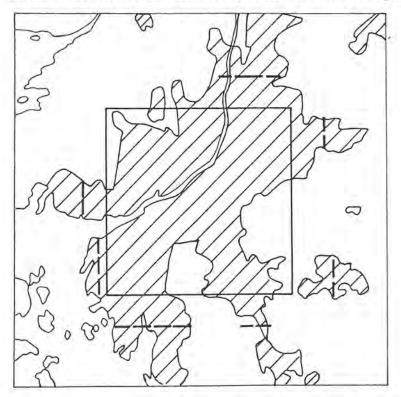


Figure 5.8. Calculation area for Gothenburg. Suburbs outside the calculation area are treated as line sources (thick dashed lines).

# Water balance at Bråvalla/Malmslätt Winter 1992/93

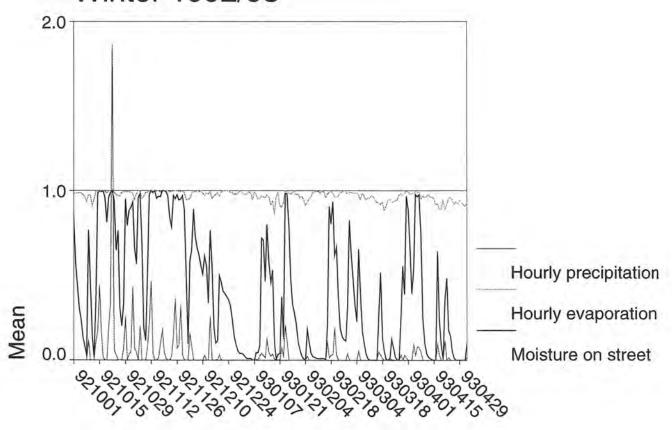


Figure 5.9 a. 1992/93. Daily values of precipitation in mm in average per hour (upward from bottom) and evaporation (downward from top). 1.0 means that the total daily precipitation was 24 mm. Moisture on street is in mm.

# Depot accumulation at Bråvalla/Malmslätt Winter 1992/93

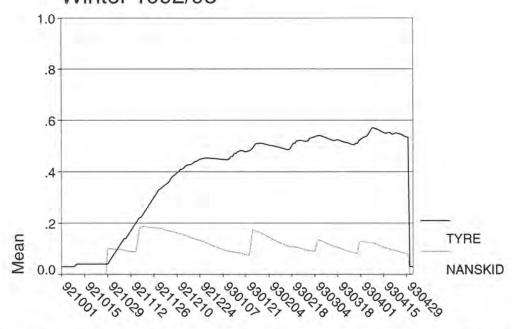


Figure 5.9 b. 1992/93. Curves for the depots of tyre and road particles (tyre) and normalized number of sanding/salting occasions (nanskid) (each with maximum 1.0).

# Resuspension factors at Bråvalla/Malmslätt

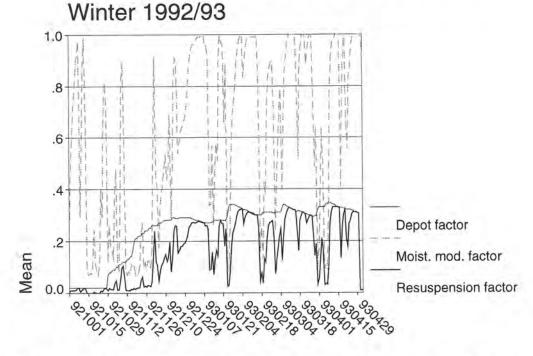


Figure 5.9 c. 1992/93. Curves for the factors in Equation 5.15. The depot factor is d. The moisture modification factor is fq. The resuspension factor is their product fqe.

# Water balance at Malmslätt Winter 1993/94

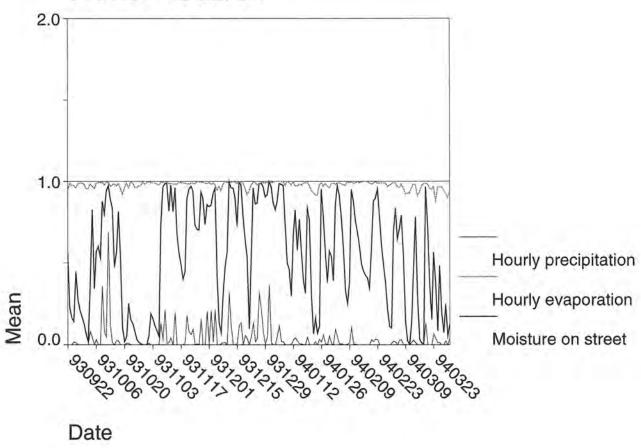


Figure 5.10 a. 1993/94. Daily values of precipitation in mm in average per hour (upward from bottom) and evaporation (downward from top). 1.0 means that the total daily precipitation was 24 mm. Moisture on street is in mm.

# Depot accumulation at Malmslätt

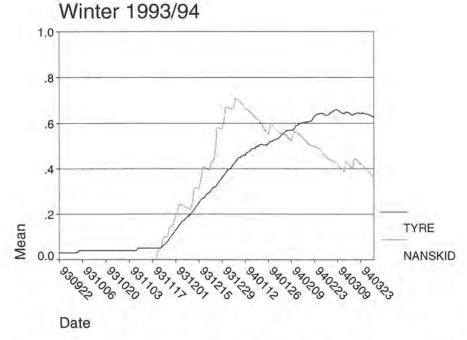


Figure 5.10 b. 1993/94. Curves for the depots of tyre and road particles (tyre) and normalized number of sanding/salting occasions (nanskid) (each with maximum 1.0).

### Resuspension factors at Malmslätt

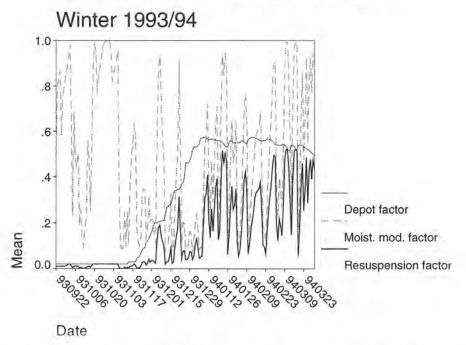


Figure 5.10 c. 1993/94. Curves for the factors in Equation 5.15. The depot factor is d. The moisture modification factor is fq. The resuspension factor is their product fqe.

## Daily mean concentrations of PM10 Norrköping - Kungsgatan

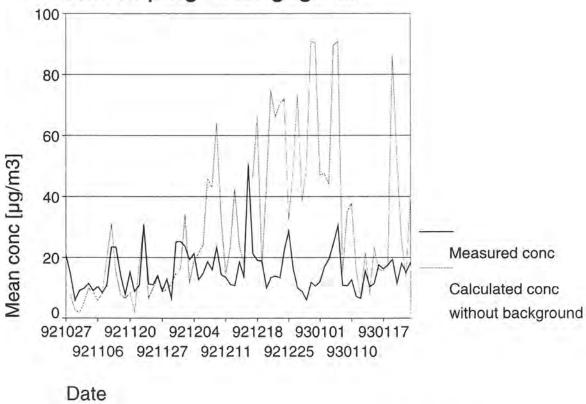


Figure 5.11. Daily mean concentrations at Kungsgatan in Norrköping calculated by the local PM-10 model. The data in Aspvreten are missing, so the results are compared to total measured concentrations.

## Daily mean concentrations of PM10 Norrköping - Hörngatan

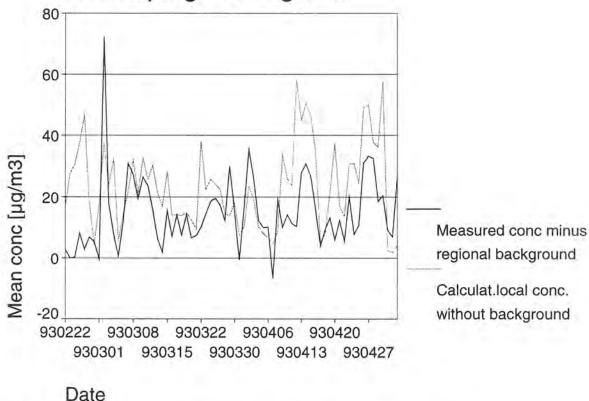


Figure 5.12. Daily mean concentrations at Hörngatan in Norrköping calculated by the local PM-10 model, compared with daily differences between measured concentration at Hörngatan and the background station in Aspyreten.

### Daily mean concentrations of PM10 Norrköping - G:a Rådstugugatan

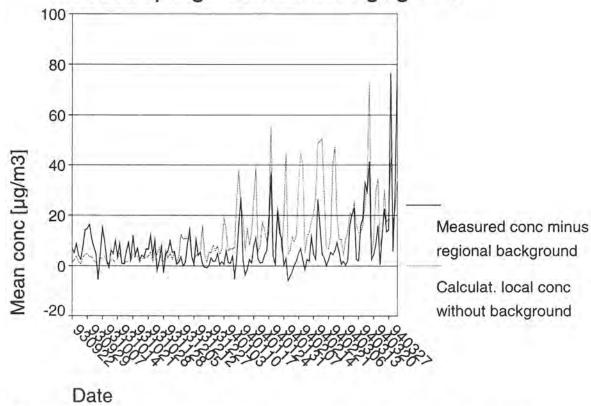


Figure 5.13. Daily mean concentrations at G:la Rådstugugatan in Norrköping calculated by the local PM-10 model, compared with daily differences between measured concentration at G:la Rådstugugatan and the background station in Aspvreten. Data at Asvreten are missing for some periods, e g in November 1993. The time scale is then reduced.

# Daily concentrations of PM10 Göteborg - Femman

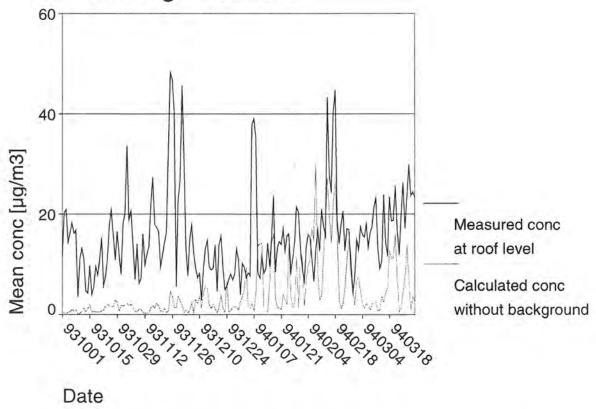


Figure 5.14. Daily mean concentrations at roof level in central Gothenburg (Femmanhuset) calculated by the local PM-10 model. The results are compared to the measured concentrations.

#### 6. RESULTS FROM THE REGIONAL MODEL PLUS THE LOCAL MODEL

Figures 6.1 and 6.2 compare measured values at G:la Rådstugugatan in Norrköping with calculated values from local model added to MATCH model. There is a good covariation between measured and calculated values. In Figure 6.1, the underestimate for November-December 1993 seems to be caused by the MATCH-Europe model, of reasons discussed in Section 4. This can be seen in comparing Figures 4.9, 4.10 and 5.13. In Figure 6.2 the level is better modeled, caused by an underestimate in the MATCH-Sweden model (Figure 4.13 and 4.14) and an overestimate in the local model (Figure 5.13).

Figures 6.3 and 6.4 compare measured values at roof level of Fernmanhuset in central Gothenburg with calculated values from the local model added to the MATCH model. It is possible to separate the contributions from the two models for different periods by comparing with Figure 4.11, 4.15 (MATCH model) and Figure 5.14 (local model). The MATCH model contributes significantly in most of the cold season 1993-94. The local model is important in January to March 1994 due to increased contributions from local resuspension.

As can be seen in Section 3.2 and Appendix 1, a number of episodes of high concentrations arising from different kinds of sources can be found in the data. The episode of 24 November to 2 December 1993 is an episode with high contributions from long-range transport. This can also be seen from the model results: the MATCH model contributes strongly and the local model very weakly. On the episode of 16 to 17 February 1994 there seem to be important contributions from both long-range transport and local resuspension.

## Daily mean concentrations of PM10 Norrköping - G:a Rådstugugatan

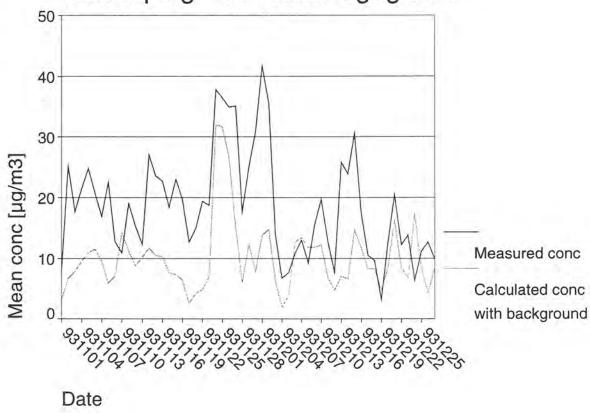


Figure 6.1. November - December 1993. Sum of daily mean concentrations at G:la Rådstugugatan in Norrköping calculated by the local PM-10 model and the MATCH-Europe model compared with measured values.

## Daily mean concentrations of PM10 Norrköping - G:a Rådstugugatan

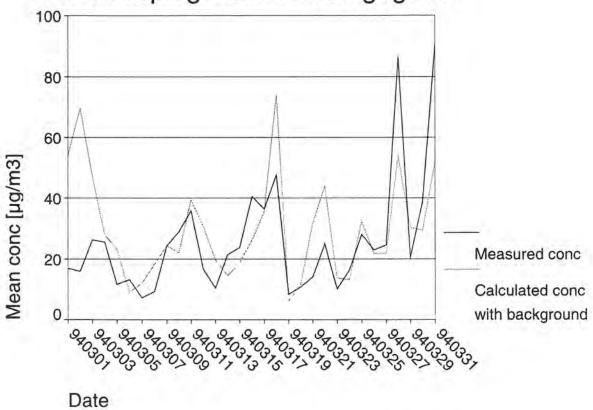


Figure 6.2. March 1994. Sum of daily mean concentrations at G:la Rådstugugatan in Norrköping calculated by the local PM-10 model and the MATCH-Sweden model compared with measured values.

## Daily mean concentrations of PM10 Göteborg - Femman

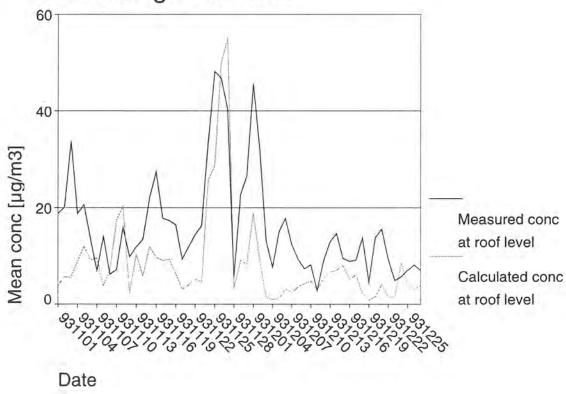


Figure 6.3. November - December 1993. Sum of daily mean concentrations at Femmanhuset in Gothenburg calculated by the local PM-10 model and the MATCH-Europe model compared with measured values.

### Daily mean concentrations of PM10 Göteborg - Femman

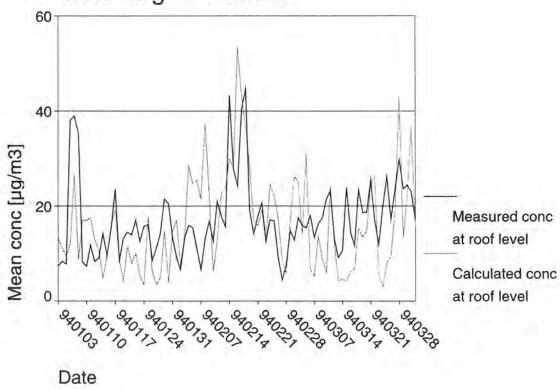


Figure 6.4. January - March 1994. Sum of daily mean concentrations at Femmanhuset in Gothenburg calculated by the local PM-10 model and the MATCH-Sweden model compared with measured values.

#### TOTAL MODEL SYSTEM

In the model system presented in this study, we have estimated the background PM-2.5 concentrations in two different and independent ways, by means of the regional scale models MATCH-Europe and MATCH-Sweden. For the time being, we have used these PM-2.5 concentrations as a first estimate of the background PM-10 concentrations. Future improvements are needed to include the total PM-10 concentrations in the MATCH system.

MATCH-Europe has the advantage that it can be applied to archived meteorological data, as well as run in real-time. Even a few days forecasts, based on weather data from HIRLAM or ECMWF weather prediction models, can be produced. Emission data for the whole of Europe is needed.

MATCH-Sweden, which needs emission data for Sweden and background air chemistry data from Scandinavian background stations, cannot be run in real-time. On the contrary, this model version can only be applied to studies for time periods at least 1-2 years back in time. Based on data from the operational yearly studies within the frame of the national environmental supervision, it is rather simple to perform the above presented calculations of PM-2.5 particulate concentrations. The simplicity of the application of PM-2.5 estimates for the whole of Sweden is the advantage of using MATCH-Sweden.

The local PM-10 model needs continuous meteorological data series, like hourly or three-hourly meteorological data from a synoptic or airport observing station. Other kinds of data series like observations from a radiation station or special meteorological masts are also possible to use. The source inventory data for the local street are very important. The PM-10 sources in other parts of the city can possibly be mapped simpler than made in this study.

At present, the MATCH model and the local model are run separately and the resulting concentrations are added hour by hour as illustrated in Figure 7.1. The boxes with thick frames refer to basic model or data treatment, while the boxes with thin frames stand for data. Here the concentrations produced by the MATCH model are processed together with "Local model DISPERSION" to give "Added concentrations". Finally "Postprocessed results" ready for use are formed by processing the added concentrations.

For practical application, investigating the consequences of different emission alternatives, it is possible to merge the two model systems for running in parallel in a more efficient manner than shown in Figure 7.1.

#### INHALABLE PARTICLES

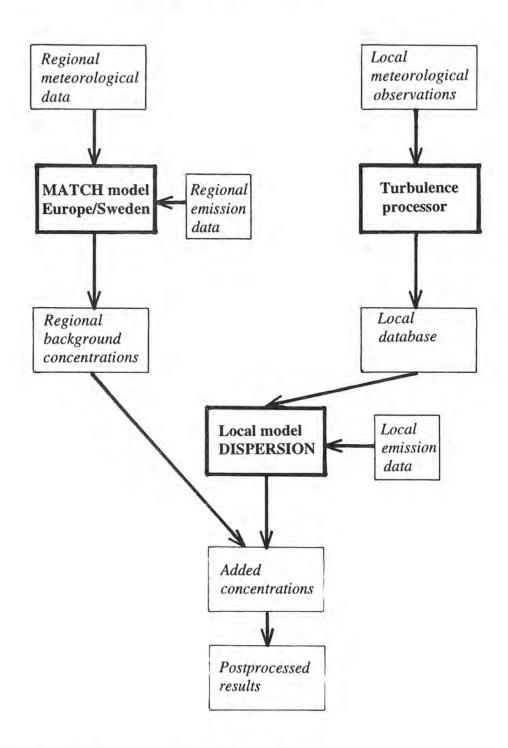


Figure 7.1. Flux diagram for the total PM-10 model.

#### Acknowledgements

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Air and precipitation chemistry data for background stations in Sweden, Norway, Finland, Denmark and Lithuania were provided by The Swedish Environmental Research Institute (IVL), The National Environmental Research Institute in Denmark and The Institute of Applied Environmental Research at Stockholm University in Sweden. Mr Gunnar Nyquist of this institute has been very kind in reviewing part of the manuscript.

The data on PM-10 and tracer elements in Norrköping have been provided by The Institute of Applied Environmental Research, The Division of Nuclear Physics of the Technical University in Lund and the Environmental Office of the Community of Norrköping. The data in the other cities have been supplied by Miljöteknik and the Community of Gothenburg, Stockholms Luft och Bulleranalys, The Community of Sundsvall, Environmental Office, The Community of Umeå, Environmental Office. The data of Aspvreten were supplied by The Institute of Applied Environmental Research.

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## Appendix 1. Episodes of high PM-10 concentration during the total model test period 1 October 1992 to 31 March 1994.

16 December 1992. PM-10 concentration in Norrköping: 51  $\mu$ gm<sup>-3</sup>. The surrounding days have rather low values: 15 - 20  $\mu$ gm<sup>-3</sup>. Gothenburg has a weaker maximum this day.

7 to 15 March 1993. The highest values occur in the beginning and end of the episode. High values in most urban and background stations.

2 to 3 April 1993. In Norrköping and Stockholm between 50 and 60  $\mu gm^3$ . Aspvreten about 25  $\mu gm^3$ .

26 to 28 April 1993. In Norrköping 50 to 62  $\mu$ gm<sup>-3</sup>. Vavihill has a low maximum but Aspvreten is about 25  $\mu$ gm<sup>-3</sup>. 27 to 28 April were warm days: Gothenburg had a maximum of 26 C. Weak winds and sunny.

24 November to 2 December 1993. In Norrköping occurred PM-10 values of 30 to 40  $\mu gm^{-3}$  and still larger in Gothenburg. High values at background stations.

25 January 1994. Norrköping: 43 μgm<sup>-3</sup>. No high values elsewhere.

16 to 17 February 1994. In Norrköping about 40  $\mu gm^{-3}$ . Rather high values in Gothenburg and Aspvreten.

16 to 18 March 1994. Norrköping: 40 to 50 μgm<sup>-3</sup>. No high values elsewhere.

28 March 1994. Norrköping: 86  $\mu gm^{-3}$ . Some hourly values are 300 to 400  $\mu gm^{-3}$ . Gothenburg: about 25  $\mu gm^{-3}$ .

31 March 1994. Norrköping: 91 µgm<sup>-3</sup>. Many hourly values above 100 µgm<sup>-3</sup>.

#### Appendix 2. Flow chart for the local dispersion model

Figure A2.1 a shows a module flow chart of the model. 1) The Setpar module reads general settings from the general scenario file. The calculation is initiated. Regional background concentrations are inserted into the grid. 2) The Input module reads source data from namelist files for different source types and stacks them on a large buffer. 3) The ChkInput module controls if user-specified data are within allowed dimensions for the model. 4) The PrintInput module controls output of user-specified data by calling source-specific printing subroutines. 5) The OpenMet module reads the sequential meteorological and turbulence data file with hourly or three-hourly observations until the start observation for the calculation period is reached. These five modules initiate the scenario calculation.

Figure A2.1 b shows a module flow chart of the hourly loop in the model. 6) The ReadMet module reads meteorological and turbulence data for the observation that is next to be used in the calculations. 7) The SurfParam module calculates the wind at the local site, for which the calculations are made (other than the observation site). Inversion layer height and surface layer height are calculated. 7b) An urban version of this module has been designed. It is described later in this section and in Appendix 3. 8) The Emission module calculates the emission from each source during the actual hour. 9) The Conc module calculates actual concentration fields for the specified sources. 10) The Statistics module calculates mean values and percentiles for each month, half year, year or total period. Exceedance statistics and time series are also output from this module. 11) The Initialize module clears the statistical matrixes for next iteration and 12) the Print module outputs the results to a table file and a plot file. The hourly loop continues until ReadMet stops it.

Figure A2.1 c shows a module flow chart for the Conc module with its subroutines for different source types. The ConcStreet module is in fact a street canyon model, which uses data from the other Conc subroutines as input at roof level of the street canyon.

### Program structure PM10-Dispersion Initialization

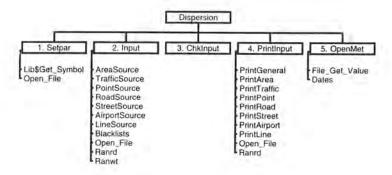


Figure A2.1 a. Module flow chart of the local scale PM-10 dispersion model. Initialization of the scenario calculations.

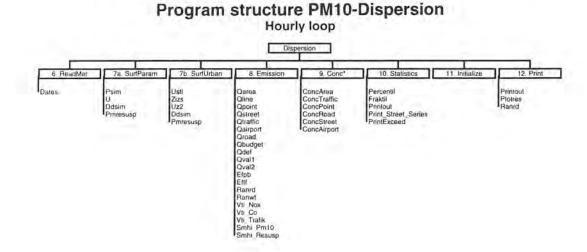


Figure A2.1 b. Module flow chart of the local scale PM-10 dispersion model. Hourly loop in the calculations of mass concentrations.

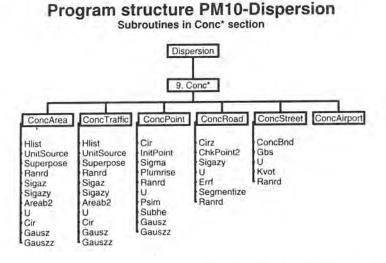


Figure A2.1 c. Module flow chart of the local scale PM-10 dispersion model. Subroutines in the calculations of mass concentration.

## Appendix 3. Calculation of urban sensible heat flux with corrections of net radiation and anthropogeneous heat emission

Total sensible heat flux:

$$H_{tot} = Rn(T_u^4) - E - G + A(T_u)$$

Rn = net radiation flux including longwave radiation from the ground due to urban temperature Tu.

There are two corrections:

I) Calculation of net radiation flux Rn.

Since Rn i Dispersion is calculated from observed rural temperature Tr at the airport, but should hold for Tu, is obtained

$$Rn(T_u^4) = Rn(T_r^4) + \sigma T_u^4 - \sigma T_r^4 \approx Rn(T_r^4) + 4 \sigma T_r^3 (T_u - T_r)$$

The average temperature difference due to urban effects is estimated as

$$T_u - T_r = 3 \text{ C}$$
 for  $T_r = -10 \text{ C}$   
 $T_u - T_r = 0 \text{ C}$  for  $T_r \ge 25 \text{ C}$ 

Then

$$T_u = T_r + (3/35)*(25 - \min(T_r, 25))$$
 and

$$Rn(T_u^4) = Rn(T_r^4) + 4 \sigma T_r^3 (25 - min(T_r, 25)) * 3 / 35$$
 W/m<sup>2</sup>.

II) Estimate of the anthropogenic surface heat flux A(Tu)

Taesler (1988) reports how the "Daily total urban district heat output" depends on outdoor temperature. In his Figure 1 is given a total heat delivery to the district heating system of 6 Gwh per 24 hours at -13 C and for a connection of 70 % of the residential buildings. For a city area of 20 km2 is obtained the heat emission 17.85 Wm-2 for -13 C, included also buildings not connected to the system.

Interpolation between

$$0 \text{ W/m}^2 \text{ at } 25 \text{ C}$$
 and  $17.85 \text{ W/m}^2 \text{ at } -13 \text{ C}$  gives  $A(T_u) = 0.470 (25 - \min (T_u, 25))$  W/m<sup>2</sup>.

Substitution of  $T_u = T_r + (3/35)^* (25 - \min(T_r, 25))$  above, gives multiplication by 1-(3/35) i.e.

$$A(T_r) = 0.430 (25 - min(T_r, 25)) W/m^2$$
.

This holds as area average for the total city area of Norrköping. In the central part, where the model will be used, it is assumed that the anthropogenic surface heat flux is three times as large as in the total area. (Roger Taesler, pers. comm.):

$$A(T_r) = 1.29 (25 - min (T_r, 25)) W/m^2$$
.

Total correction.

Since  $\sigma = 5.67 * 10^{-8}$  and if assumed that  $T_r^3 = 273^3$  is obtained

$$H_{tot} = Rn(T_r^4) - E-G + 4 \sigma T_r^3 (25 - min (T_r, 25)) * 3 / 35 + 1.29 (25 - min (T_r, 25)) =$$

$$= Rn(T_r^4) - E-G + (0.395 + 1.29)(25 - min (T_r, 25)) = Rn(T_r^4) - E-G + 1.69 (25 - min (T_r, 25))$$

Also should be added heat contributions from traffic and chimneys of units for local heating and industry. Therefore, the total correction is estimated as a factor two i. e.

Htot = 
$$(Rn(T_r^4) - E-G) + 2 * (25 - min(T_r, 25))$$

For the model this can be written

$$Htot_{urban} = Htot_{rural} + 2 * (25 - min (T_r, 25))$$

Htot<sub>rural</sub> then is the surface heat flux routinely calculated by the preprocessor in Dispersion without urban effects.

The following table shows calculated total corrections to surface sensible heat flux in central Norrköping:

$$\begin{array}{lll} T_r & 2*(25-min~(T_r,25)) \\ -10~C & 70~W/m^2 \\ 0~C & 50~W/m^2 \\ 10~C & 30~W/m^2 \\ 25~C & 0~W/m^2 \end{array}$$

Thus regard has been made to

- the size of built up area
- the correction should be greater in the densely built up city area (multiplication by 3 above)
- traffic part, buildings not connected to district heating system and industry (crudely)

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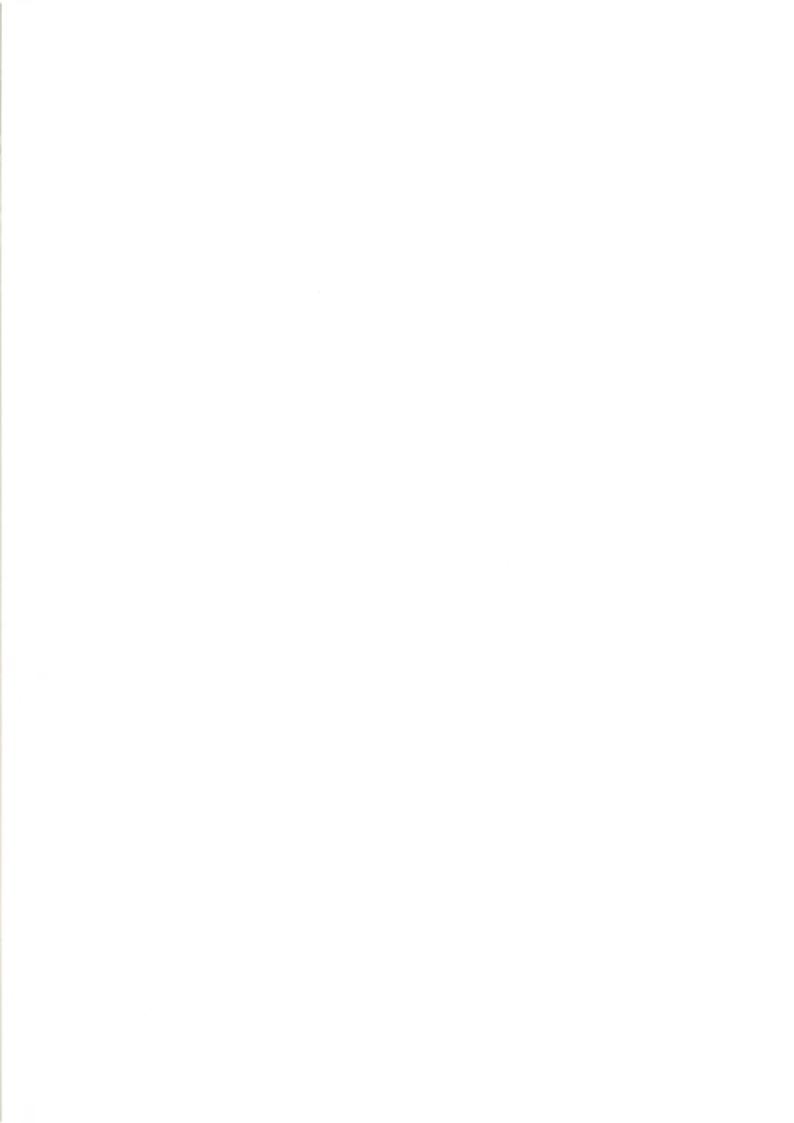
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