NATIONAL INSTITUTE OF PUBLIC HEALTH AND THE ENVIRONMENT
BILTHOVEN, THE NETHERLANDS

Report no. 723301 007

Description and analysis of ambient fine particle concentrations in the Netherlands

J.T. van der Wal, L.H.J.M. Janssen.

September 1996

This investigation was commissioned by the Ministry of Housing, Spatial planning and Environmental protection (VROM) with respect to the project 723301 Urban Air Quality / Fine Particles.

RIVM, National Institute for Public Health and the Environment, P.O.Box 1, 3720 BA Bilthoven,
The Netherlands, tel. +31 (0)30-2749111, fax +31 (0)30-2742971

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Abstract

The spatial- and temporal variations of PM10 concentrations in the Netherlands, as measured in the National Air Quality Monitoring Network in the period 1993-1994, were analysed. Concentration gradients are small and differences in concentrations between measuring stations amounted to ca. 20% or less. Locally sources like traffic and urban emissions and industrial, agricultural and natural sources may increase PM10 concentrations by about 10%, with the regional background as a baseline. Because PM10 is a sum parameter for fine particles, the composition of PM 10 may vary more than its concentration pattern in the Netherlands, which is mainly determined by large-scale weather systems.

Most observed PM10 concentrations vary are found between 20 and 50 $\mu g/m^3$ during the year, with little difference between summer and winter. During episodes PM10 concentrations may increase to 4 to 5 times the annual average (>200 $\mu g/m^3$). Kalman-filtering is a time-varying linear regression technique. It was found to be an adequate method in modelling the daily variations of PM10. The final model used wind direction, temperature and duration of precipitation as explanatory variables.

SAMENVATTING

Het is al lang bekend dat kleine deeltjes (fijn stof of PM10; deeltjes met een aërodynamische diameter < 10 µm) een nadelig effect hebben op de gezondheid. Ondanks emissie-reductie maatregelen die in het verleden al genomen zijn, blijven de niveaus van fijn stof boven de Nederlandse normen (i.e. 40/140 µg/m^3, het jaar- en dag maximum resp.). Het nemen van aanvullende beleidsmaatregelen wordt bestudeerd en momenteel bereid de EU een dochterrichtlijn voor m.b.t. luchtverontreiniging door Fijn Stof. Deze analyse van de ruimtelijke en temporele variaties van PM10 niveaus, kan een bijdrage leveren aan het bepalen van het rendement van aanvullend beleid.

In deze analyse is gebruik gemaakt van luchtverontreinigingsgegevens (waaronder PM10-gegevens) zoals gemeten in het Landelijk Meetnet Luchtkwaliteit, dat wordt beheerd door het RIVM. Meteorologisch gegevens zijn afkomstig van het KNMI. In het Landelijk Meetnet worden drie typen meetstations onderscheiden: regionale (achtergrond) stations, stadsstations and straatstations.

De analyse geeft aan dat concentratie-verschillen tussen stations gering zijn. In Nederland kunnen verschillen tussen stations in het relatief schone noordwestelijke deel en het meer door buitenlandse en lokale bronnen beïnvloede zuidoosten oplopen tot maximaal 20% van de gemeten concentraties.
Lokaal kunnen verschillende bronnen zoals verkeers- en stedelijke emissies, industriële, agrarische en natuurlijke bronnen de PM10-concentratie verhogen met ca. 10% t.o.v. de regionale achtergrond. Wij vonden een verhoging van circa 5-6 µg/m^3 t.o.v. de regionale achtergrond ten gevolge van zeezout in kustgebieden, verkeer en industriële emissies in stedelijke en industrie-gebieden en lokale agrarische activiteiten op sommige regionale stations.
Omdat PM10 een "somparameter" is voor fijn stof, kan de samenstelling van fijn stof meer variëren dan het concentratiepatroon in Nederland dat voor een belangrijk deel wordt bepaald door grootschalige weerpatronen.
PM10-concentraties variëren tussen de 20-50 µg/m^3 in een jaar met weinig verschil tussen de zomer en de winter.
Een analyse van de meteorologische variabelen windrichting, windsnelheid, temperatuur, hoeveelheid regen en duur van de regenperiode, relatieve vochtigheid, zoninstraling, luchtdruk en menghoogte liet zien dat een aantal van deze variabelen hoog gecorreleerd is en dat de ruimtelijke variatie van deze variabelen over Nederland gering is. Om de dagelijkse variaties van PM10 te modelleren zijn daarom windrichting, temperatuur and neerslagduur zoals gemeten in De Bilt genomen om de dagelijkse variaties van PM10 in Nederland te beschrijven.
Het Kalman-filter is een tijdsafhankelijke lineaire regressie methode. De dagelijkse variaties van PM10 kunnen tamelijk goed worden beschreven met het Kalman-filter en windrichting, temperatuur en neerslagduur als verklarende variabelen. Variaties in de windrichting kunnen de trend van PM10 in een range van -10 tot +40 µg/m^3 beïnvloeden; variaties in temperatuur in een range van -10 tot +30 µg/m^3 en neerslagduur in een range van +2 tot -10 µg/m^3.
Gedurende episodes met droog weer, een lage temperatuur (<0°C) en een oostelijke wind kunnen PM10-concentraties oplopen to 100-150 µg/m^3. Hoge windsnelheden kunnen de PM10-concentratie verder verhogen met zo'n 100 µg/m^3 ten gevolge van opwaaiend stof.

SUMMARY

For a long time fine particles (PM10; particles with an aerodynamic diameter <10µm) have been known for their adverse health effects. Despite emission reduction measures in the past, fine particle levels are still above national guidelines (i.e. 40/140 µg/m^3, the yearly and daily averaged maximum values, resp.). Additional policy measures are studied and the EU is preparing a daughter directive on Fine Particle Pollution. This analysis of the spatial and temporal variations in PM10 levels may contribute to determine the effectiveness of abatement policy.

Air pollution data (including PM10 concentrations) as measured in the National Air Quality Monitoring Network and meteorological data supplied by the Royal Dutch Meteorological Institute have been used. In the monitoring network three types of monitoring stations are distinguished: rural (regional background), town and street stations.

Concentration differences between stations are small, upto ca. 20%, between clean background stations in the north-west and measuring stations in the south which are more strongly influenced by large-scale transport from foreign source areas and local sources. Local sources such as traffic and urban emissions, industrial, agricultural and natural sources may contribute to the measured concentrations and PM10 concentrations may increase by about 10%, with the regional background as a baseline. We found an increase of about 5-6 µg/m^3 with respect to the regional background due to sea salt in coastal areas, traffic and industrial emissions in urban areas and local agricultural activities at some rural stations.

Because PM10 is a sum parameter for fine particles, the composition of PM 10 may vary more than its concentration pattern in the Netherlands, which is mainly determined by large-scale weather systems.

Most observed PM10 concentrations are between 20 and 50 µg/m^3 during the year, with little difference between summer and winter.

An analysis of the meteorological variables wind direction, wind speed, temperature, amount of precipitation, duration of precipitation, relative humidity, insolation, atmospheric pressure and mixing height showed that most variables are highly correlated and that the spatial correlation for most variables over the area of the Netherlands is high. To model the daily variations of PM10, we therefore selected wind direction, temperature and duration of precipitation as measured in De Bilt to model the daily variations of PM10 over the Netherlands.

Kalman-filtering is time-varying linear regression technique. The results show that the daily variations of PM10 can be modelled reasonably well using the Kalman-filter and wind direction, temperature and duration of precipitation as explanatory variables.

Variations in wind direction could influence the trend in the PM10 concentration between -10 and + 40 µg/m^3; variations in temperature could influence PM10 concentrations between -10 and + 30 µg/m^3 and duration of precipitation could influence PM10 concentrations in a range between +2 and - 10 µg/m^3.

During episodes with dry weather, low temperatures (<0°C) and wind blowing from easterly directions PM10 concentrations may increase to 100-150 µg/m^3. Very high wind speeds may increase PM10 concentrations further by upto 100 µg/m^3 due to wind blown dust.

1. INTRODUCTION

For a long time fine particles have been known for their adverse health effects (Brimblecombe, 1987). In the past decades several emisson reduction measures have been taken to reduce ambient concentrations. However because of industrial and societal development as well as meteorological factors, both long-term-average and short-term-episodic fine particle concentrations are still high (Mage *et al.*, 1996; Pryor and Barthelmie, 1996). Concern about the present levels has risen, as several epidemiological studies (Dockery *et al.*, 1993; RIVM, 1994a; Annema *et al.*, 1994, Annema *et al.*, 1996; RIVM, 1995, Pope *et al.*, 1995; Ricci *et al.*, 1996) point out that health effects can occur. Observed health effects include premature deaths, increased occurrence and/or severity of respiratory complaints leading to an increased medicine use and increased risk for hospital admittance. Besides the risk for people suffering from afflictions of the pulmonary tract, there is also a risk for those with cardiovascular diseases (Schwartz and Dockery, 1992; Pope *et al.*, 1995; RIVM, 1995). An important source of uncertainty in human health studies on the concentration–response relationship, is the way in which mortality is corrected for weather influences (RIVM, 1995). Mortality data are very sensitive to meteorological parameters. The issue of how to compensate for the confounding effect of the weather in epidemiological and emission trend studies is also mentioned by Wilson (1995), Pryor and Barthelmie (1996), Lippmann and Ito (1995), Lipfert (1994) and Lipfert and Wyzga (1995a and 1995b).
Due to the association of fine particle air pollution with adverse health effects, the EU is preparing a Framework Directive on air pollution and a daughter directive on fine particle pollution, which will be decided on in 1997. An integrated assessment project has been started in the Netherlands to support both national policy and the EU Directive. This report is part of that project and presents an analysis of the contribution of sources and meteorology to concentrations of fine particles in the Netherlands. It may also serve to determine the effectiveness of abatement policies.

To contribute to an answer to these questions, this study analyses monitoring data for fine particles (PM10) and other air pollutants from the National Air Quality Monitoring Network in conjunction with meteorological data as supplied by the Royal Dutch Meteorological Institute.

2. MATERIALS AND METHODS

2.1 Air Quality Monitoring Network and RIL+-database

The Air Research Laboratory (LLO) of the National Institute of Public Health and the Environment (RIVM) operates a countrywide Air Quality Monitoring Network (LML). Several air pollutant components such as PM10, sulphate, nitrate, sulphur dioxide and nitrogen oxides are monitored in the LML. All the data from the National Air Quality Monitoring Network are stored in the RIL+database at the RIVM. The meteorological data are supplied by the Royal Dutch Meteorological Institute (KNMI) and added to the RIL+database. Meteorological parameters include for example, temperature, wind direction, air pressure and precipitation.

In the LML fine particles are measured using a β-radiation monitor. This monitor works by measuring the extinction of a radioactive source (β-radiation) by the dust settling on the filter (Swaan and van Elzakker, 1994). The size of the particles entering the apparatus is controlled by the design of the sampling head. The employed sampling head has a 50% cut-off point for particles with a diameter of 10 μm. The secondary aerosol constituents are measured by chemical analysis of filters from low volume samplers (LVS). This method is described in Swaan and van Elzakker (1995). Volatile components (benzene) are measured by collection on activated charcoal tubes followed by gas chromatography after elution (Somhorst and van Elzakker, 1995).

2.2 Fine particles and meteorology database

The relevant data for the study period were extracted from the central RIL+database using standard reports. The period analysed covers the years 1993 and 1994. A database which contains data on the concentrations of several air pollutant components was constructed: PM10 (particulate matter with an aerodynamic diameter <10 μm), secondary aerosols (sulphate, nitrate, ammonium and chloride) and several gaseous or volatile air pollutants (SO_2, NO_x, benzene) as measured in the Dutch Air Quality Monitoring Network and meteorological data (T, wind direction, wind velocity, amount and duration of precipitation, atmospheric pressure, relative humidity and insolation) as supplied by the KNMI. Also included was mixing layer height as measured using a LIDAR at the RIVM in Bilthoven. This measurement is explained in Salemink and van Maanen (1985), with the addition of a variance analysis to the procedure as described there. After extraction for further analysis the database was maintained using MS-Excel 5.0 (Microsoft 1994). More detailed information on the handling of the data is given in §2.3. The basic structure of the data files is discussed in Appendix B.

The LML stations used in this study are those where PM10 is measured. Other components are also measured at these and other sites in the monitoring network. In the LML three kinds of monitoring stations exist: rural, urban and street. Most of the LML stations were in operation at the start of the study period or earlier. Exceptions are:
Amsterdam - Noord (21/2/93), Breukelen - snelweg (18/5/94), De Zilk (28/4/93), Apeldoorn - Stationsstraat (5/3/93), Rotterdam - centrum (7/9/93), Dordrecht (21/9/93), Eindhoven - Genovevelaan (16/3/93), Vredepeel (2/2/93). The date between parentheses is the starting date for PM10 measurements. For every LML station a twin KNMI station was selected to supply the most relevant meteorology. The LML stations are listed in Table 1,

which contains information on their location, the kind of station and distances between the twinned stations. Table 2 lists the KNMI stations used in this study with their location. In some of the tables in this report an abbreviation will be used to indicate the station. This abbreviation is a combination of the first five letters of the station name directly followed by the station number. The names and numbers used are those given in Table 1 and Table 2. An example: station name Witteveen and station code 928 combine to give Witte928 as an abbreviation.
Figure 1 is a map of the Netherlands showing the location and type of the stations.

Table 1: Stations in National Air Quality Monitoring Network (LML) where PM10 is measured.

Station code	*Station type*	*Station name*	*Latitude degr. N*	*Longitude degr. E*	*Amersfoort coordinate x*	*y*	*Code Twin KNMI station*	*Distance [1] (km)*
928	rural	Witteveen	52°48'46"	6°40' 8"	2414	5369	290	62
538	rural	Wieringerwerf	52°48'17"	5° 2'56"	1322	5352	235	22
520	town	Amsterdam - Noord - 20	52°23'38"	4°55' 8"	1231	4896	240	12
444	rural	De Zilk	52°17'52"	4°30'35"	952	4791	240	20
641	street	Breukelen - snelweg	52°12'10"	4°59'20"	1277	4683	240	18
639	street	Utrecht - Const. Erzeijstr.	52° 4' 8"	5° 7'15"	1367	4533	260	6
722	rural	Eibergen	52° 5'31"	6°36'21"	2385	4566	290	27
728	street	Apeldoorn - Stationsstraat	52°12'54"	5°58' 1"	1946	4697	275	17
724	rural	Wageningen	51°58'26"	5°38'56"	1730	4428	275	19
404	town	Den Haag - Centrum	52° 4'33"	4°17'13"	796	4547	210	14
433	street	Vlaardingen - macro	51°54'43"	4°19'33"	820	4364	344	10
418	town	Rotterdam - Centrum	51°54'50"	4°28'44"	925	4365	344	7
437	rural	Westmaas	51°47'13"	4°27' 3"	904	4224	344	21
441	town	Dordrecht	51°48'17"	4°40' 8"	1055	4242	100348	25
230	rural	Houtakker	51°31' 8"	5° 8'56"	1384	3922	350	16
318	rural	Braakman	51°17'42"	3°44'56"	408	3685	310	21
236	street	Eindhoven - Genovevalaan	51°28'11"	5°28'22"	1609	3866	370	4
131	rural	Vredepeel	51°32'27"	5°51'10"	1873	3947	375	16
133	rural	Wijnandsrade	50°54'10"	5°52'55"	1898	3237	380	8

[1]: Distance between LML station and twinned KNMI station.

Table 2: KNMI-stations used for meteorological parameters.

Station code	*Station name*	*Latitude degr. N*	*Longitude degr.E*	*Amersfoort coordinate x*	*y*
235	De Kooy	52°55' 1"	4°46'58"	1140	5480
240	Schiphol	52°18'43"	4°47'52"	1148	4805
260	De Bilt	52° 5'34"	5°11'24"	1415	4560
290	Twente	52°12'36"	6°53'56"	2570	4764
275	Deelen	50°51' 0"	5° 7'12"	1890	4532
210	Valkenburg	51°39' 7"	4°24'28"	890	4650
344	Rotterdam	51°47' 5"	5°38'41"	901	4429
100348	Cabauw	51°58' 8"	4°55'40"	1234	4423
350	Gilze-Rijen	51°58'11"	4°45'53"	1235	3975
310	Vlissingen	51°27' 0"	3°36' 0"	300	3860
370	Eindhoven	51°27' 0"	5°27' 0"	1570	3845
375	Volkel	51°39' 7"	5°41'52"	1765	4070
380	Maastricht	50°55' 8"	5°46'44"	1825	3255

2.3 Data handling

The original data were available as hourly values and extracted from the RIL+database in this form. The extracted data were imported into MS-Excel 5.0 (Microsoft, 1994) and stored as spreadsheets. This program was also used for calculation and selection of subsets of data for different parts of the analyses. The following steps of data handling are important:

- correction for sampling efficiency of the β-radiation monitor,
- time-synchronisation of LML and KNMI data,
- calculation of daily average values from hourly average values with special attention to wind direction and speed,
- classification of wind direction for use with statistical techniques,
- classification of LML stations into four distinct aggregated stations: Background, Agriculture, Town and Street.

The hourly values for PM10 as extracted in November 1995 from the RIL+system were not corrected for sampling efficiency. The original databases of hourly values as they are archived for this study on backup tapes (Appendix A) are still in this uncorrected form. The archived databases with daily averaged values have the corrected values. All data presented in the report have been corrected for sampling efficiency by multiplying the stored values by a correction factor (1.33), based on experimental observations that the Low Volume Sampler inlet configuration used results in a systematic underestimation of fine particles. Some of this evidence is reported in Holländer *et al.* (1990). Since December 1995 all PM10 data extracted from the RIL+database system are correct, in the sense that the output is corrected for sampling efficiency of the β-radiation sampling apparatus.

The hourly values of the LML data are in local time, which is Central European Time (CET). The KNMI data are stored in UTC/GMT or CET-1 hour. All KNMI data have been shifted by one hour prior to analysis to synchronise LML and KNMI data.

The hourly data on fine particles and meteorology were studied first. Later daily values were calculated from the hourly values. The daily values have been used for most of the analyses. For most variables the daily values are arithmetic means of 24 hourly values. For the meteorological variables 'duration of precipitation' and 'amount of precipitation' the sum of 24 hourly values was taken as the daily value. The averages for wind direction and wind speed were calculated in a more complex manner. A wind vector was defined with the wind direction as the direction and the wind speed as the length. The vectors of hourly values were added vectorially to yield an average wind direction and speed for the day. This method is customary in meteorology.

In the meteorological database the wind direction data stored are rounded to the nearest ten degrees, where both 0 and 360 degrees are used. These values are not identical. Zero degrees (0°)stands for variable wind and/or very low wind speeds, in other words situations where it is difficult to assign an actual direction to the wind. The other wind direction 360° means wind blowing from the North.

The circular nature of the wind direction is problematic when trying to apply statistics, since these techniques usually assume a linear characteristic for the variables. To overcome this problem several ways of treating wind direction have been tried in the analyses. Instead of the normal classification of the wind direction according to degrees (0-360), it can be classified using measured levels of PM10 as a weighting factor. The PM10 concentrations used for this purpose are presented as windroses and will be discussed later. Classifications of wind direction are:

- WindSector which subdivides the wind into four sectors of 90 degrees. To avoid ambiguity in allocating the four main wind directions to a sector the starting point is shifted ten degrees. The sector around SE , starting with 100 degrees (10 degrees past East) to 190 degrees (just past South) was given sector number 1. This first sector has the highest PM10 concentrations. The next sector, with lower PM10 concentrations, was SW. It was given number 2, NE = 3 and the cleanest sector NW = 4. This classification has been used to investigate the importance of other variables, such as Temperature or 'Duration of precipitation', within a given sector;
- WindRank: assigns a rank number (1-37) to each wind direction (0;10;20;30 ... 360) based on the average concentration of PM10 observed with winds from that direction going from high to low;
- LinearWind: the compass is divided into 12 sectors of 30 degrees each. Start with the sector having the highest average concentrations (80-90-100) EAST, which gets number 1, and then continue clockwise (2,3, ..., 12) from there;
- WindClass: based on the windrose; every wind direction (n=37) is awarded a class number. This class number is 1 for the cleanest wind direction (<20 µg/m3), 2 for 15 to 20 $\mu g/m^3$ and so forth. Following this the average Class is calculated for each of the 12 sectors as described for LinearWind.

An example of the resulting classifications of wind direction are given in Table 3.

At some point during the study a decision was made not to continue with all 19 LML-stations (§3.3.5). Instead, a choice was made to analyse only four distinct types of station. Two of these types are identical with the categories 'urban' and 'street' as used in the LML. Within the rural stations it was found that not all stations exhibit similar behaviour. Some have a pattern that is interpreted as typical of a background station; some others, however, show rather high levels of PM10 and are thought to be influenced by local sources, probably of agricultural origin. The types of station recognised are referred to as Background, Agriculture, Town and Street. Each aggregated station is made up of two LML stations typical of the category by averaging their data. Table 4 gives the details.

A choice was made to limit the number of stations in each aggregation to two to facilitate comparison between aggregated stations. For Background the two stations used are the only two who have data for both years and show a clear background behaviour. Adding stations 444 De Zilk with data missing for the first four months of 1993 would have made the comparison between both years and between stations more difficult. For the aggregation of Agriculture the stations with the two highest PM10 concentrations were selected.

Table 3: Example of wind direction coding

Station: WindDirection (degrees)	Background PM10 (µg/m3)	Wind Rank	Wind Sector	Linear Wind	Wind Class
0	35.82	16	3	11	2.75
10	19.85	35	3	12	1.67
20	19.18	36	3	12	1.67
30	26.94	22	3	12	1.67
40	28.51	20	3	1	4.33
50	32.79	18	3	1	4.33
60	42.37	11	3	1	4.33
70	43.20	10	3	2	7.67
80	51.14	6	1	2	7.67
90	59.24	2	1	2	7.67
100	45.15	9	1	3	8.00
110	61.16	1	1	3	8.00
120	48.99	8	1	3	8.00
130	58.85	3	1	4	8.33
140	50.01	7	1	4	8.33
150	51.63	5	1	4	8.33
160	54.17	4	1	5	6.33
170	37.05	15	2	5	6.33
180	40.64	12	2	5	6.33
190	38.03	13	2	6	4.67
200	37.19	14	2	6	4.67
210	34.88	17	2	6	4.67
220	28.52	19	2	7	3.00
230	25.16	25	2	7	3.00
240	27.62	21	2	7	3.00
250	25.87	24	2	8	2.33
260	24.24	28	4	8	2.33
270	24.41	27	4	8	2.33
280	26.19	23	4	9	2.33
290	20.87	34	4	9	2.33
300	23.85	29	4	9	2.33
310	22.33	33	4	10	2.00
320	22.75	30	4	10	2.00
330	22.75	31	4	10	2.00
340	25.06	26	4	11	2.75
350	22.35	32	3	11	2.75
360	17.83	37	3	11	2.75

Table 4: LML stations used for the aggregated stations Background, Agriculture, Town and Street

Station name	LML Stations	
Background	928 Witteveen	538 Wieringerwerf
Agriculture	230 Houtakker	131 Vredepeel
Town	520 Amsterdam - Noord	404 Den Haag - centrum
Street	639 Utrecht - Erzeijstr.	236 Eindhoven - Genoveveln.

2.4 Statistical methods

Statistical methods used in this study include:

- simple descriptive statistics (mean, variance, correlation coefficient),
- Principal Component Analysis,
- (Multiple) Linear Regressions and
- Kalman-filtering (a time-variant linear regression method).

The spatial pattern of fine particles was analysed using Principal Component Analysis. A concise description of this technique and its use for this purpose is given in §2.5. The time series were analysed using (multiple) linear regression and Kalman-filtering. The use of Kalman-filtering is discussed in §2.6.

All statistical calculations in this report have been performed assuming the data to be normally distributed. This assumption has not been explicitly tested and no transformation of data has been applied to make the data conform to a normal distribution.

2.5 Spatial analysis using Principal Component Analysis

Before starting a Principal Component Analysis (PCA) it is convenient to standardise the data of each station to have zero mean and unit variance. This yields a matrix **C** of dimension n x m, where n is the number of stations and m is the number of observations in the time series. From **C** a correlation matrix R (n x n) can be calculated by:

$$\mathbf{R} = 1/m * \mathbf{C} * \mathbf{C}^T, \qquad (1)$$

where $\mathbf{C}^T$ is the tranpose m x n matrix of matrix **C**.

Based on the correlation matrix **R** of between-stations correlations, PCA can be used to gain insight into the spatial behaviour of air pollution fields. From matrix **R** a n x n matrix **E** is calculated which consists of the n orthogonal Eigenvectors $[e_k]$ of **R**. The corresponding Eigenvalues are denoted by λ_k. The Eigenvectors and Eigenvalues can be interpreted in terms of variance reduction because when using standardised concentrations, the fraction of the total variance explained by the *k*th Eigenvector is equal to λ_k /n. Matrix **C**, which consists of the original and correlated aerosol concentrations, can now be written as the product of the orthogonal matrix $\mathbf{E}^{-1}$ multiplied by the matrix **Q** which consists of new and uncorrelated concentrations:

$$\mathbf{C} = \mathbf{E}^{-1} * \mathbf{Q} \quad \text{or} \quad \mathbf{C} = \mathbf{E}^T * \mathbf{Q} \qquad (2)$$

Equation (2) implies that concentration matrix **C** can be considered as a product of matrix **Q**, the rows of which vary only with time, and matrix $\mathbf{E}^T$, the columns of which vary only in space. The n columns of $\mathbf{E}^T$ can be considered to represent n uncorrelated air pollution patterns. The first pattern represents the mutual coherence (covariance) of the n stations, the measure of which is determined by the magnitude of the corresponding Eigenvalue λ_1. As each element e_{ik} of the *k*th Eigenvector $[e_k]$ belongs exclusively to a particular station, it is possible to draw n maps, each representing a characteristic air pollution pattern. The description given here is based on Janssen *et al.*(1989), which contains further references.

2.6 Kalman-filtering as a tool to analyse time series

Kalman-filtering is a structural time-series model. The filter was developed in the sixties by R.E. Kalman for use in industrial process control. It has become popular as a result of the good statistical properties of its estimators for unknown parameters. Kalman-filtering has since been used in many fields. The approach has become most popular in the field of econometrics as a result of work done by Harvey (1989 and 1993).

The advantage of structural time-series models is that components such as a trend, a cyclic signal and the influence of explanatory variables can all be modelled additively and as such can be studied separately. Furthermore, the parameters in the model are allowed to change with time. This means that in the regression model used, the weighting factors are time-dependent (Visser, 1996).

Structural time-series models are modular. The model for a time-series, y_t, can be seen as the addition of four components:

y_t = $trend_t$ + $cycle_t$ + influence of explanatory $variables_t$ + $noise_t$

The trend is a low-frequency part of the time-series. It can be simple (e.g. a constant or a linear trend) or be made flexible or time-variant by adding a noise source to the trend model (Visser 1996). The number of noise sources depends on the trend model. In this study a constant value for the trend has been used alongside its time dependent version referred to as Stochastic Level. A Stochastic Level trend has one noise source.
Cyclic signals can be filtered from a time series by including a cyclic component in the model. The period length must be constant and only one cyclic signal can be estimated at one time. The signal may have any possible form and may change with time by adding a noise source. Note that this modelling of a cyclic signal is not the same as Fourier analysis, where a signal is decomposed into a number of sine functions.

Explanatory variables can be accommodated by adding a regression model to the time-series model. When combining the regression part with a stochastic level trend without adding any noise sources the time-series model simplifies to a multiple linear regression model. The model becomes flexible by adding noise sources to the weights of one or more of the explanatory variables (Visser, 1996). The flexibility is controlled by giving a value for the amount of noise allowed. Alternatively optimal values can be calculated by the software. The values allowed for added noise sources range from 0 (= no flexibility) to 10 (extreme flexibility). Commonly, values smaller than 1 are used e.g. 1.0×10^{-4}. The values used in this study for the different noise sources were determined by iteratively testing different values and evaluating the results.

The Kalman-filter is based on an iterative process referred to as filtering. Based on observations for time t, it generates a prediction for the observations for t+1. By comparing the prediction with the observed value of y_t the filter is adjusted. This continues for all observations y_t resulting in N single-step prediction errors for t=1,...,N. The Kalman-filter generates model estimates by minimising the sum of squared single-step prediction errors. Besides estimates for the components of the structural model the Kalman-filter also gives standard deviations (per time step) for the trend, the cycle and the weights of the regression. Using the standard deviations confidence intervals can be constructed with $\pm 1\sigma$ for 68% and $\pm 2\sigma$ for 95% confidence.

When all observations are available prior to analysis, improved model estimates for y_t can be calculated by smoothing (Visser, 1996). Smoothing is a process where the iterative filtering process is repeated with one crucial difference. Smoothing not only uses the previous observations but also the following observations to adjust the original filtered model results. The results presented here have been generated using the smoothing process.

Predictions with confidence limits can be generated for future values of y_t, t=N+1, N+2, ..., N+L using the Kalman-filter. When explanatory values are part of the model two situations are possible: (i) values of explanatory variables are available for the prediction period and can be used to generate improved predictions or (ii) no such values are available. In both situations estimates can be made.

In the present study the following structural model was used:

$$PM10= L(t) + C(t) + a_1(t)x_1 + a_2(t)x_2 + a_3(t)x_3 + a_4(t)x_4 + ... + a_n(t)x_n + noise(t) \qquad (4)$$

where
$L(t)$ = a Stochastic Level trend model,
$C(t)$ = a cyclic signal,
$a_i(t)$ = weight for explanatory variable x_i in the regression model.

To each term a noise source can be added to increase the flexibility of the model. Without added noise sources and a cyclic signal the structural model is equivalent to multiple linear regression analysis, where L and a_i are constant.

Kalman-filter calculations were done using KALFIMAC 5.0 software (Visser, 1996). The analysis as calculated by KALFIMAC is controlled by an option file. Examples of option files as used in this study are included in Appendix B. In Appendix C a sample is given of a data file, with a description of the data in the columns.

Integrated in the KALFIMAC software are statistical tests which indicate whether the assumptions that are made by using the Kalman-filter are not violated. The tests include the calculation of an autocorrelation function, a Port-Manteau test, a Runs-test and an integrated test for Normality, Homoscedasticity and Independence (Jarque and Bera, 1980). All tests are performed on the residuals of the time-series model. The most important assumption that should not be violated is the independence of the residuals. If the residuals are normally distributed this is good, if this is not the case the Kalman estimators are still expected to perform well.
The Port-Manteau test tests whether the residuals are 'white noise' i.e. behave randomly. The Runs-test tests if the number of upward and downward runs in the residuals is randomly distributed. If this is the case, the number of runs will be within the statistical limits. If the number of runs is less, this is evidence of a trend still present in the residuals. If the number of runs is higher, a cyclical pattern may be present.

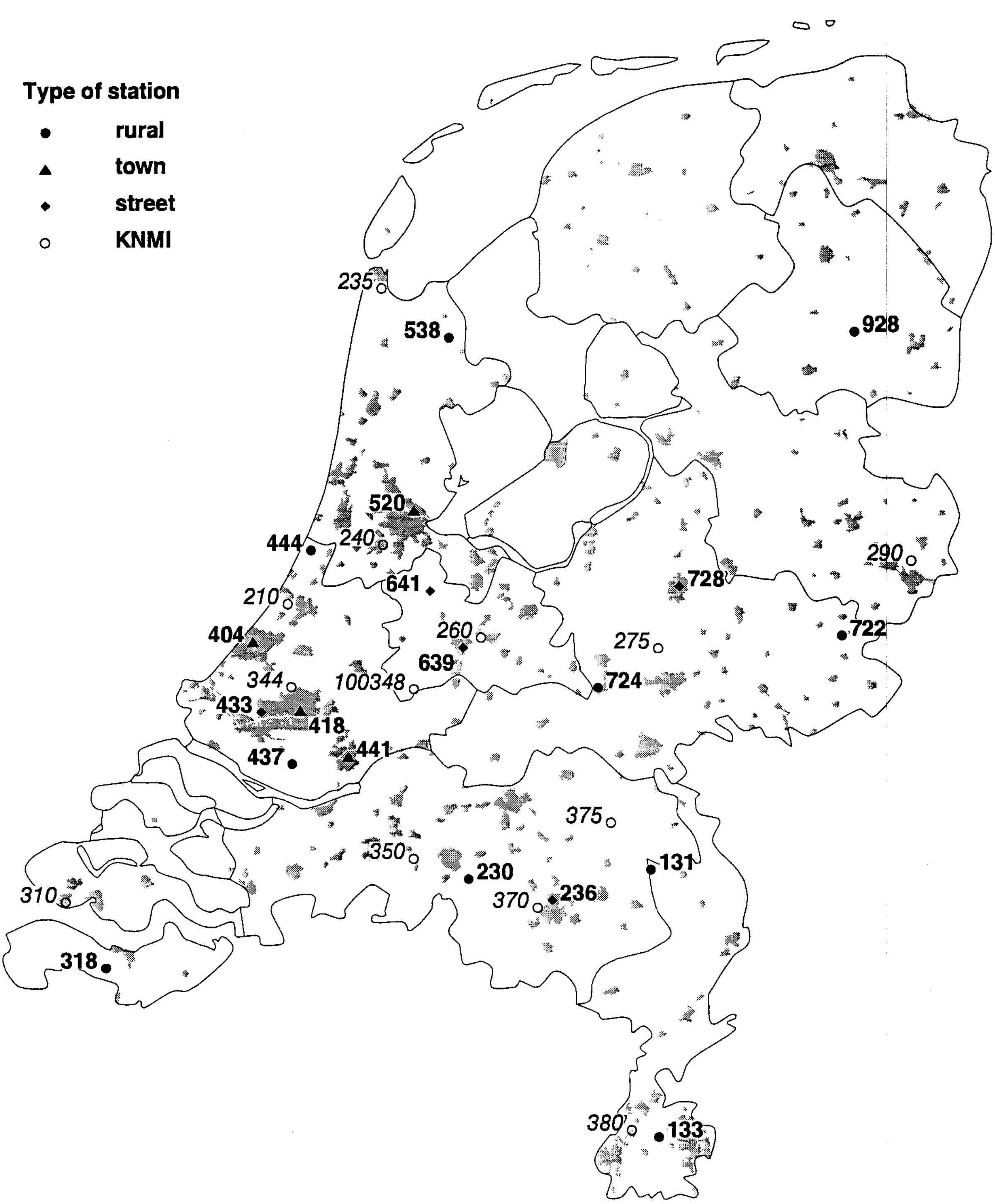

Figure 1: Map of the Netherlands showing the locations and types of the LML and KNMI stations. Numbers indicated on the map are the station codes from Table 1 and Table 2.

3. RESULTS

Measurements of fine particles from the LML Air Quality Monitoring Network in the Netherlands are analysed for temporal and spatial variability. An important part of the analyses performed also uses meteorological data. First, a descriptive analysis of the recorded PM10 levels is given. Next, the analysis results with a spatial component are presented followed by the results of time-series analyses.

3.1 Descriptive analyses

Using the hourly values, plots were made to obtain a first impression of the relationship between levels of PM10 and meteorological variables. A sample of time-series plots is presented in §3.1.1, while in §3.1.2 a short description is given, characterising distinctive periods within the two years under study. Included are descriptions of the course of PM10 levels during the day (§3.2) for summer and winter. Part of this section is concerned with the difference between weekdays and weekends in observed PM10 levels during the day.

3.1.1 Plots of hourly values

To gather an initial understanding of the relationship between levels of PM10 and meteorological variables, plots were made of the hourly values for all variables. Presented is one set of plots for a selected month. This month - November 1993 - has been selected to illustrate some of the points under study. By combining the information from all graphs an understanding of the relationship between the weather and fine particles was gained.

The PM10 levels of the first nine days of November are elevated (50-100 μg/m^3). The elevated levels start during the last few days of October, with colder nights, light north-easterly to easterly winds (< 5 m/s), high atmospheric pressure and no precipitation. The episode continues as long as the weather stays like this. Some rainfall on the seventh starts to bring the PM10 levels down. The levels of PM10 in this period are possibly elevated through stagnation.

The next week has low PM10 levels (5-30 μg/m^3). South to south-westerly winds prevail, they are also stronger (5-10 m/s). On the 15th even stronger winds blow, a storm with wind speeds over 20 m/s. In this week precipitation falls.

The next two weeks again have above normal levels, similar to the first week (50-100 μg/m^3). This coincides with frosty weather, light easterly winds (around 5m/s), while it stays mainly dry. This episode lasts until the beginning of December. At that time it stops as the temperature rises above 0 °C and the winds turn to south-west bringing precipitation. This second episode is a likely example of high levels of fine particles caused by transport from source areas over a long distance.

These episodes can be typified as a stagnation episode (first week) and a transport episode (third and fourth week). This is substantiated by benzene levels for the same periods. Benzene is a marker for the (local) influence of traffic. In the stagnation period benzene levels were high in both rural areas (ca. 5 μg/m^3) and in street stations (8-10 μg/m^3). During the transport episode levels were 2 and 5 μg/m^3, respectively, which is elevated relative to normal levels but only half of what they were during the stagnation episode.

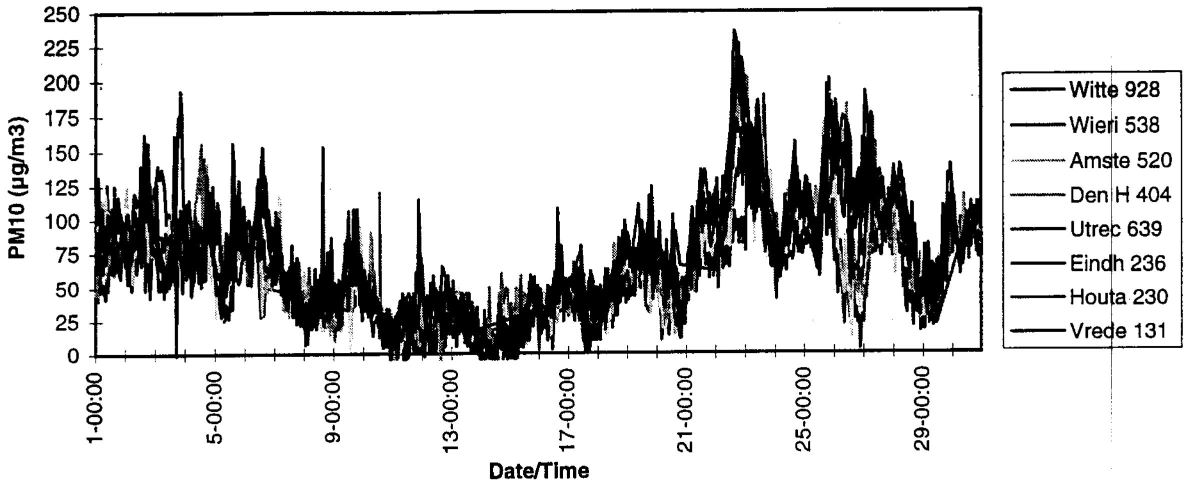

Figure 2: PM10 in November 1993.

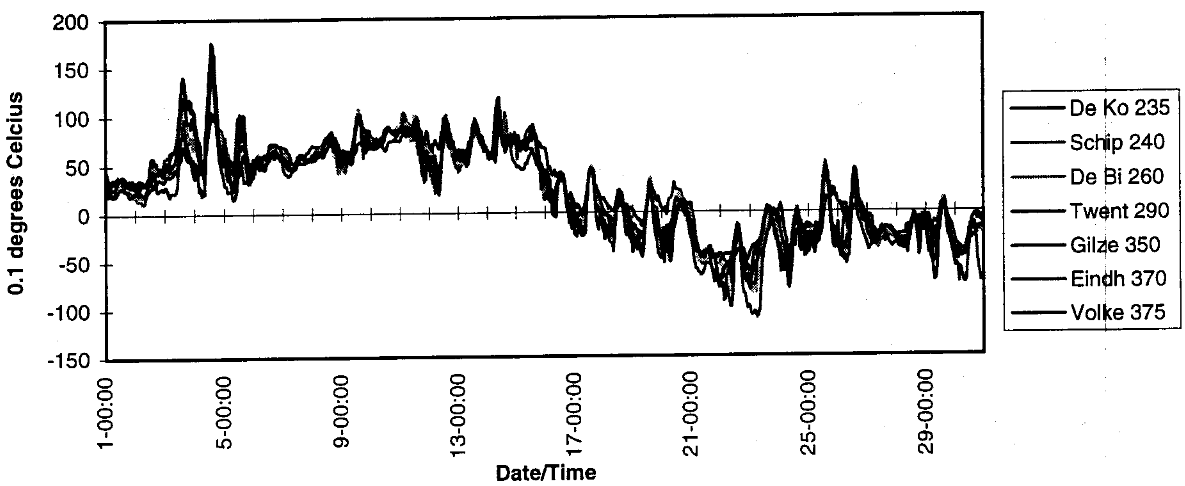

Figure 3: Temperature in November 1993.

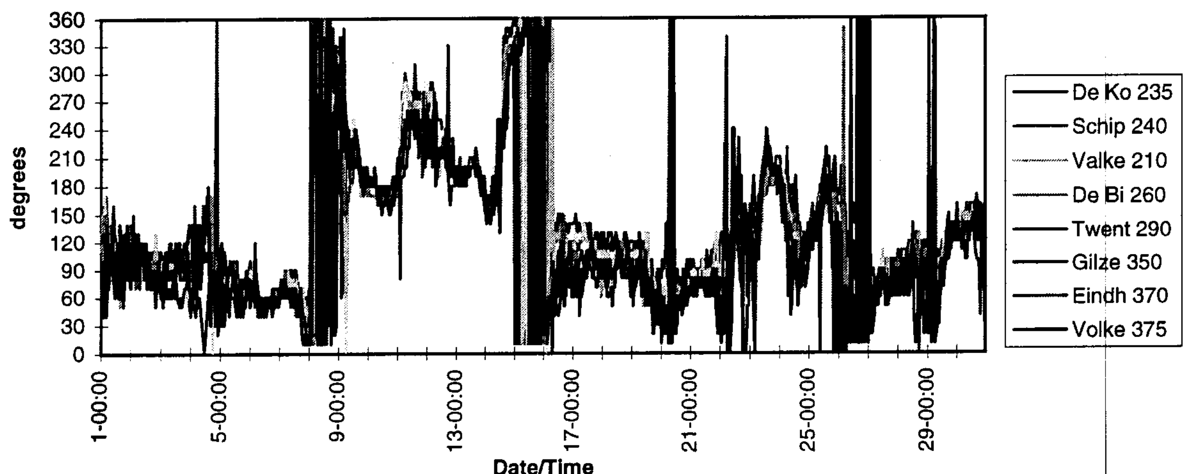

Figure 4: Wind direction in November 1993.

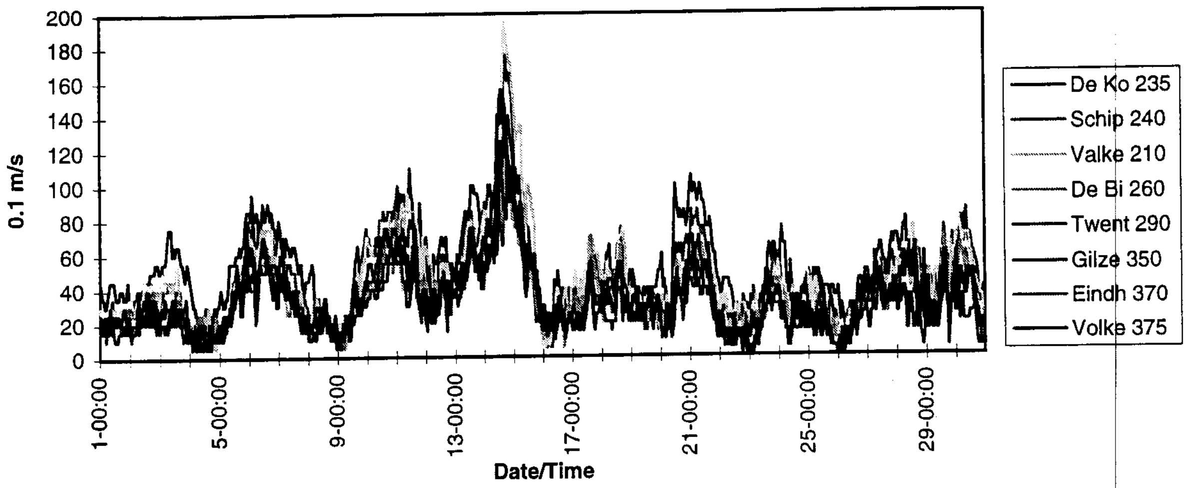

Figure 5: Wind speed in November 1993.

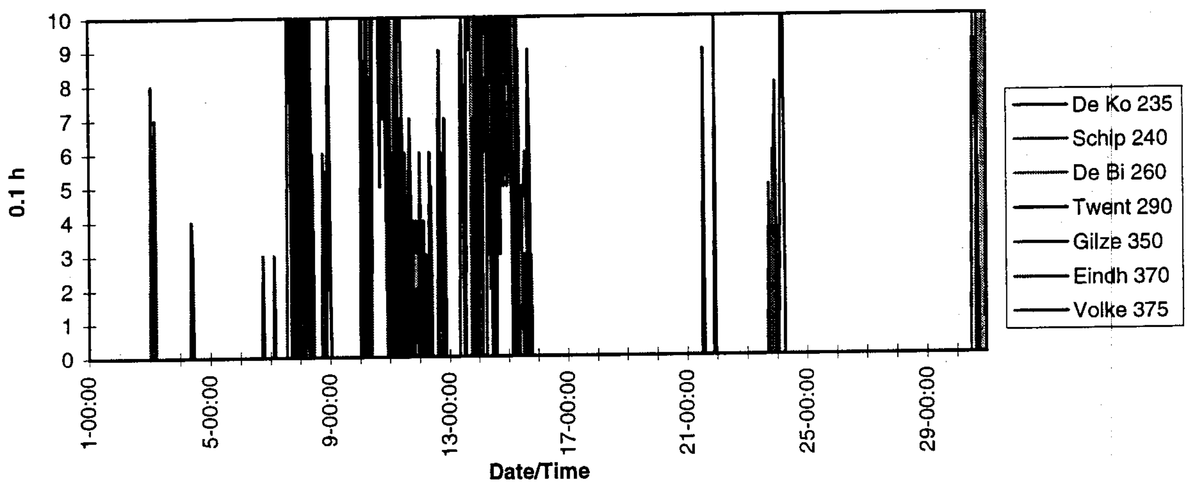

Figure 6: Duration of Precipitation in November 1993.

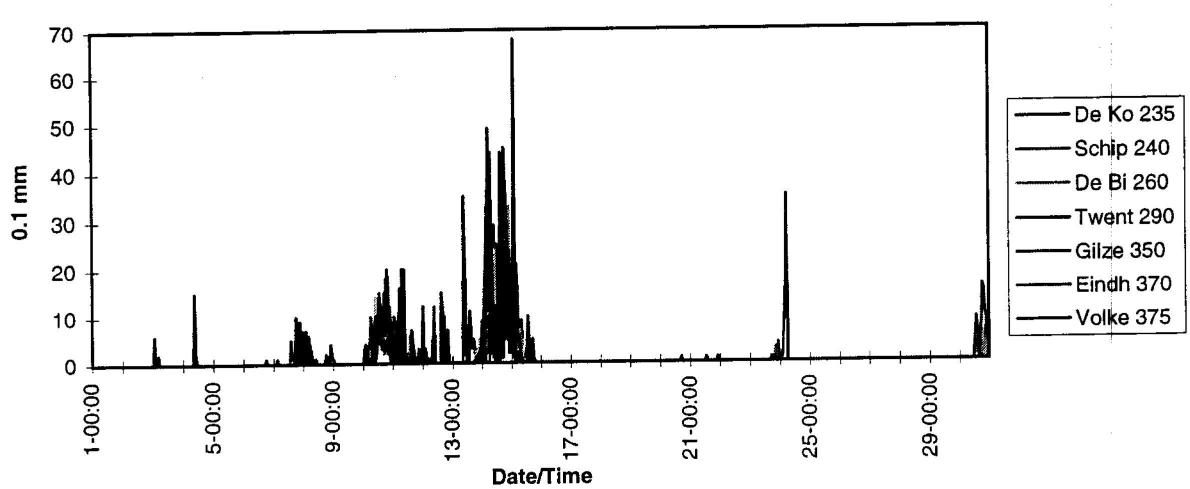

Figure 7: Rainfall in November 1993.

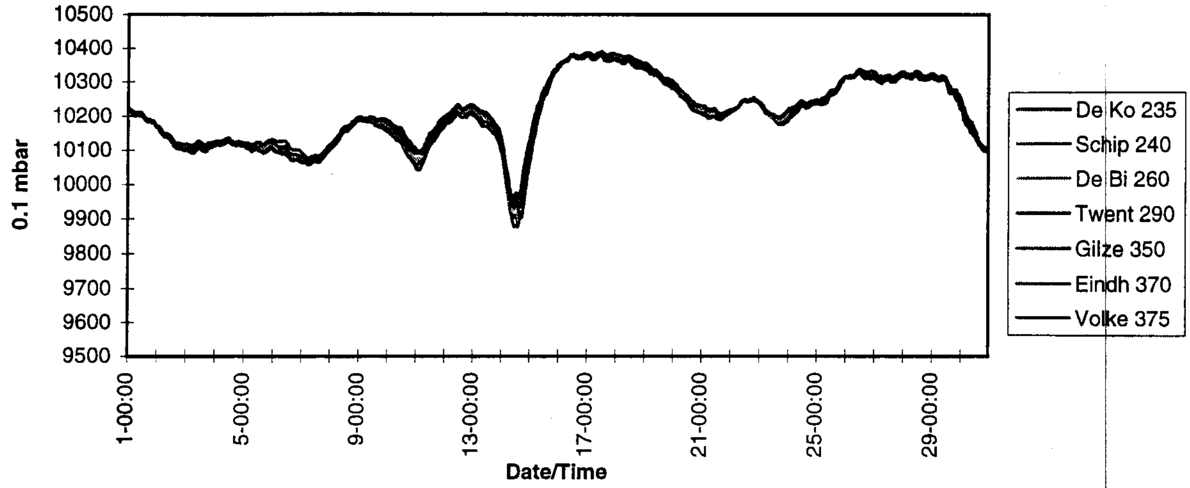

Figure 8: Atmospheric pressure in November 1993.

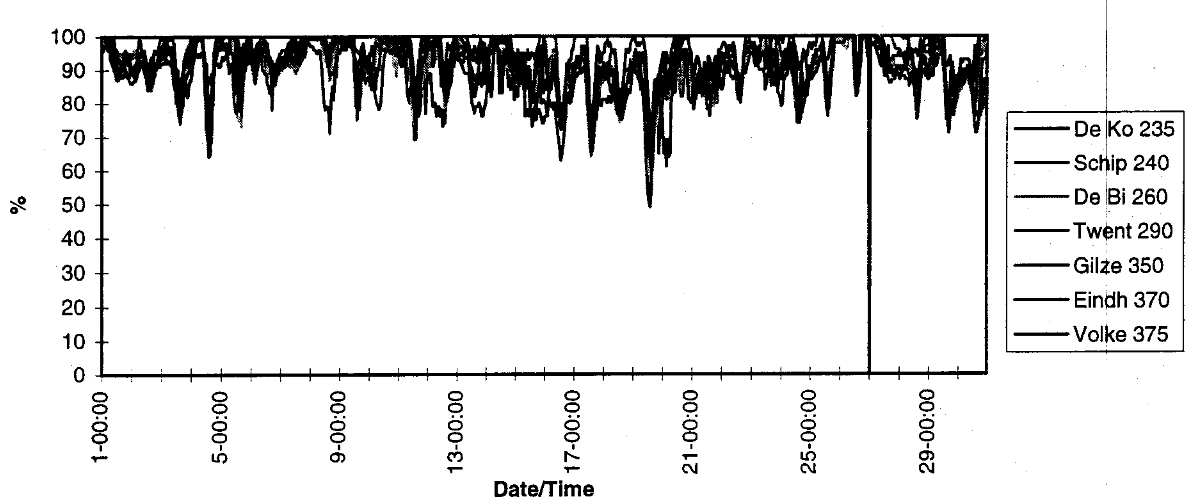

Figure 9: Relative humidity in November 1993.

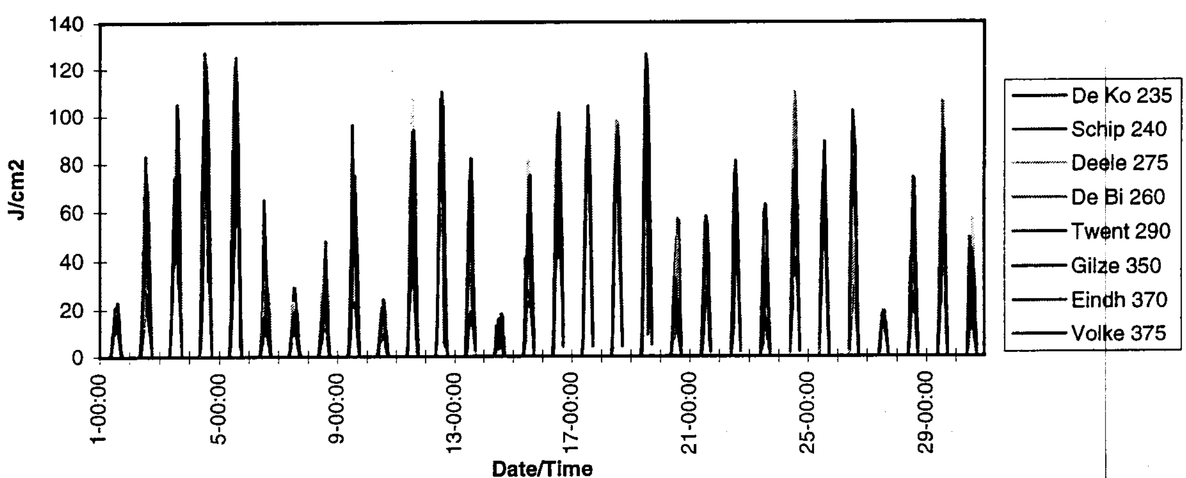

Figure 10: Insolation in November 1993.

3.1.2 Description of selected periods

Based on the complete set of graphs of hourly values as presented in the previous section, periods with special characteristics are identified. Some episodic periods with higher than normal levels of PM10 (>50 μg/m^3) and some periods with low levels are listed in chronological order.

1 Jan. '93, over 400 μg/m^3, extremely high levels
In combination with frosty weather, low wind speeds, high atmospheric pressure and no precipitation the fireworks associated with New Year's Eve add a lot of dust. This anthropogenic episode is very punctual. In 1993 this coincided with a weather pattern that aggravated the problem (Noordijk, 1993), in 1994 the weather did not make matters worse (Noordijk, 1994).

9-15 Feb. '93, ≈ 130 μg/m^3, high levels
High pressure and light winds between east and south. Dry with frost during the nights.

16-24 Feb. '93, ≈ 25 μg/m^3, low levels
Strong winds between south-west and north, mostly around 10 m/s, with some storms up to 20 m/s bring precipitation. Temperature around 5 °C, no frosts during the nights until the 21st.

1-27 Apr. '93, around 40 μg/m^3, mostly under 65 μg/m^3, normal levels scattering upwards
Temperature around 10 °C, winds varying from south to west to north and light (around 5m/s). Regular rain, mostly local. On the 12th higher values are recorded at some stations coinciding with a light easterly wind.

28 Apr. - 2 May '93 65-130 μg/m^3, some peaks up to 200, elevated levels
Warmer weather, initially steady winds (5 m/s) around north-east to east at the end turning west. No precipitation.

For the rest of the year the PM10 levels stay mostly below 65 μg/m^3 until November (Figure 2 through Figure 10). The summer is relatively cold and wet with mostly westerly winds.

7-13 Feb. 1994 mostly between 40 and 50 μg/m^3, normal levels.
A light wind slowly turns from south to west to north to north-east, high atmospheric pressure, no precipitation.

13-15 Feb. 1994 locally peak levels over 400 μg/m^3, high levels
The lowest recorded values during this period are around 45 μg/m^3. Frosty dry weather, high atmospheric pressure and a strong wind (10-15 m/s) from east to south-east.

16 Feb. 1994 ≈ 50 μg/m^3, normal level
Continuing frost and dry weather, but a light wind (<5 m/s) from the south

17-25 Feb. 1994 65-130 μg/m^3 , some peaks up to 200, elevated levels
Slow build up with cold weather (frost during the nights), light winds (< 5m/s) variable around east. Mainly dry but some precipitation from 21-24 Feb.

26 Feb. 1994 20-50 μg/m^3, normal to low levels
Start of a period lasting to somewhere in April with mainly westerly winds that blow stronger at 5-10 m/s. During this period there is regular precipitation. Temperatures are well above 0 °C.

1-31 Jul. 1994 ≈ 65 µg/m^3, slightly elevated, strong scatter.

During this month many days had warm summer weather (max. temp over 30 °C). The wind is mostly light (<5m/s) from variable directions. The atmospheric pressure is stable. Localised precipitation, most likely in the form of thunderstorms bring up to 10 mm per hour of rain in some places. The recorded PM10 levels appear to repeat a rise and fall pattern during the day. This behaviour is already present during part of June and persists into August.

For the rest of the year no exceptional levels of PM10 are recorded. Recorded values are mostly under 50 µg/m^3.

It appears that episodes with high PM10 levels coincide with the following weather conditions:

- winds from directions around east to south-east,
- low temperatures (around freezing or lower),
- dry weather.

It can be concluded that the most extreme levels of PM10 are recorded during the winter. Elevated levels do occur during summer but are less extreme. Levels of fine particles are higher than normal during summer under the following conditions:

- high temperatures
- dry weather.

3.2 Fluctuations on a diurnal basis

Since human activity and some meteorological parameters are governed by the clock, the fluctuation of PM10 during the day and night was studied by plotting the hourly observations averaged over a three month period for every hour of the day. This was done for: Winter: January, February + December for both 1993 and 1994 and likewise for Summer: June, July, August for 1993 and 1994. To ascertain whether weekdays are different from the weekend similar calculations were done for: Autumn: September, October, November for both years. In this case, weekdays (Mon-Fri) were averaged separately from weekends (Sat-Sun). The results are presented below.

In the following sections average levels for the period will be given in summarised tables using the following station grouping:

- Rural-low: Witteveen 928, Wieringerwerf 538, De Zilk 444;
- Rural-average: Eibergen 722, Wageningen 724, Westmaas 437, Wijnandsrade 133;
- Rural-high: Houtakker 230, Braakman 318, Vredepeel 131;
- Town: Amsterdam-Noord 520, Den Haag-centrum 404, Rotterdam 418, Dordrecht 441;
- Street: Breukelen 641, Utrecht-Erzeijstraat 639, Apeldoorn-Stationsstraat 728, Vlaardingen-macro 433 and Eindhoven-Genovevelaan 236.

3.2.1 Diurnal pattern in the summer

In the summer of 1994 (June, July and August) a distinct diurnal pattern is found for rural stations in the LML network (Figure 11). During the night the levels are high (between 25 and 80 µg/m^3) and lower (between 20 and 40 µg/m^3) during the day. Levels of PM10 drop sharply around 06.00 (about 1.5 hours after sunrise). From around 13.00 levels slowly begin to rise again. This rise increases considerably around 17.00. The highest levels of the day are reached around midnight.

At the stations Houtakker, Vredepeel and Wageningen the recorded levels are consistently higher (30-70 μg/m^3) than at other stations. The stations De Zilk, Wieringerwerf and Witteveen have the lowest values (20-40 μg/m^3).

Town stations (Figure 12) that measure the background level of PM10 in urbanised areas, record levels comparable with those measured at 'average' rural stations (Figure 11) between 20 and 50 μg/m^3. Peak levels during the day are recorded late in the evening. This also holds for Dordrecht, Rotterdam and Amsterdam. Den Haag has a different pattern. There the recorded levels are highest during the day.

At street stations the pattern during the day is as shown in Figure 13. The recorded levels, comparable with town (Figure 12) and 'average' rural stations (Figure 11), are in the range 20-50 μg/m^3. The PM10 course clearly shows an hourly pattern during the day with higher values during the night.

The previous year (1993) does not clearly show the pattern during the day as found in 1994. This will be discussed later. The rural stations with the highest and the lowest levels are the same. Levels in 1993 are on average lower than in 1994 (Table 5). The difference between 1993 and 1994 is around 15 μg/m^3 for rural stations; somewhat less for the cleanest stations (14 μg/m^3) and a bit more (17 μg/m^3) for rural stations with generally higher levels of PM10. For town stations the difference between 1993 and 1994 is comparable with that of the rural stations: around 15 μg/m^3 higher levels in 1994. Street stations show the largest difference with 1994 having levels that are almost 20 μg/m^3 higher than those of 1993.

Table 5: Levels of PM10 in μg/m^3 for 1993 and 1994 during the summer (June, July and August)

		1993		1994	
		Mean	**S.E.**	**Mean**	**S.E.**
Rural					
	Low	19.0	0.5	33.4	0.7
	Average	22.2	0.6	40.7	1.1
	High	29.8	0.8	47.9	1.5
Town		20.4	0.4	35.9	0.7
Street		22.0	0.4	40.2	0.7

3.2.2 Diurnal pattern in the winter

During the winter months rural stations show no clear diurnal cycle (Figure 14). Levels of fine particles do not vary much during the day. The recorded levels lie between 25 and 50 μg/m^3, with most stations within the narrower range of 40 to 45 μg/m^3. De Zilk is the cleanest station, with levels ranging from 25 to 40 μg/m^3. Other clean stations are Wijnandsrade and Witteveen: 30-45 μg/m^3. Stations where high levels are measured are Houtakker en Braakman with 40-50 μg/m^3.

In Figure 15 it can be seen that town levels lie between 40 and 50 μg/m^3. There is only a very slight pattern where daytime levels are a bit higher at 45-50 μg/m^3 than night-time levels at 40-45 μg/m^3. The daytime levels are also higher than levels recorded at rural stations. This difference is largest for Den Haag and smallest for Dordrecht.

On the street PM10 levels are between 30 and 45 μg/m³, but most stations measure values between 40 and 45 μg/m³, as shown in Figure 16. Only Breukelen-snelweg is lower and remains mainly below 40 μg/m³. The course during the day is low. Usually the lowest levels of the day occur around 05:00 to 06:00 in the morning. After that levels rise somewhat. This rise remains almost constant for most of the day. In Apeldoorn, Utrecht and Vlaardingen levels show a small dip around lunch-time. The highest levels are measured between 16:00 and 19:00.

Table 6: Levels of PM10 in μg/m³ for 1993 and 1994 during winter (January, February + December)

		1993		1994	
		Mean	S.E.	Mean	S.E.
Rural					
	Low	26.5	0.6	37.4	0.7
	Average	29.8	0.4	39.5	0.3
	High	31.5	0.4	44.9	0.4
Town		27.0	0.7	45.2	0.4
Street		24.3	0.8	44.7	0.5

When comparing the winter period of 1993 with that of 1994 it can be noticed that:

- The observed average levels are higher in 1994 than they were in 1993 (Table 6). The difference is around 10 μg/m³ for rural stations with low levels. On stations with high levels the difference between the years is also greater, about 13 μg/m³.
- The same stations have low levels in both years: De Zilk, Witteveen, Wijnandsrade and Wieringerwerf. Likewise for the stations Houtakker, Vredepeel, Braakman and Westmaas, where high levels were measured in these years.
- In both years there is no hourly pattern for PM10 levels during the day on rural stations.
- The pattern, as described above for town stations in 1994, was the same in 1993 i.e. levels during the day a little higher than during the night. The difference in PM10 between the years for town stations is of the same magnitude (around 10 μg/m³) as that for the rural stations.
- The differences between 1993 and 1994 are largest at street stations. At street stations the recorded levels of the winter of 1994 are about 20 μg/m³ higher than those of 1993.

The difference in levels between 1993 and 1994 during the winter can be attributed to more frequent easterly winds in 1994. Especially the episode in February 1994 is important in this respect. With easterly winds the anthropogenic influence on PM10 levels increases and at the same time the levels increase as well.

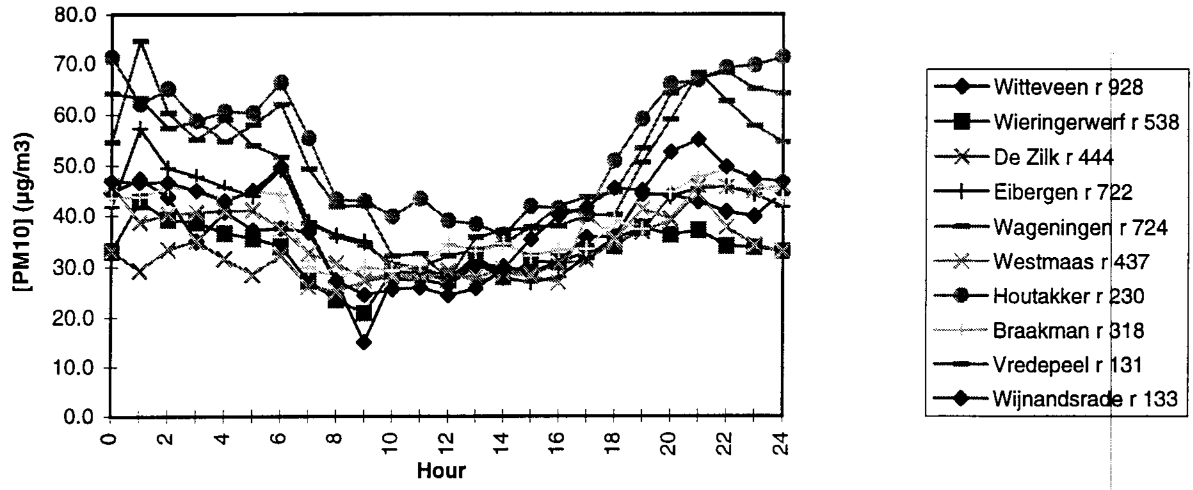

Figure 11: Course of PM10 during the day, rural stations in summer 1994.

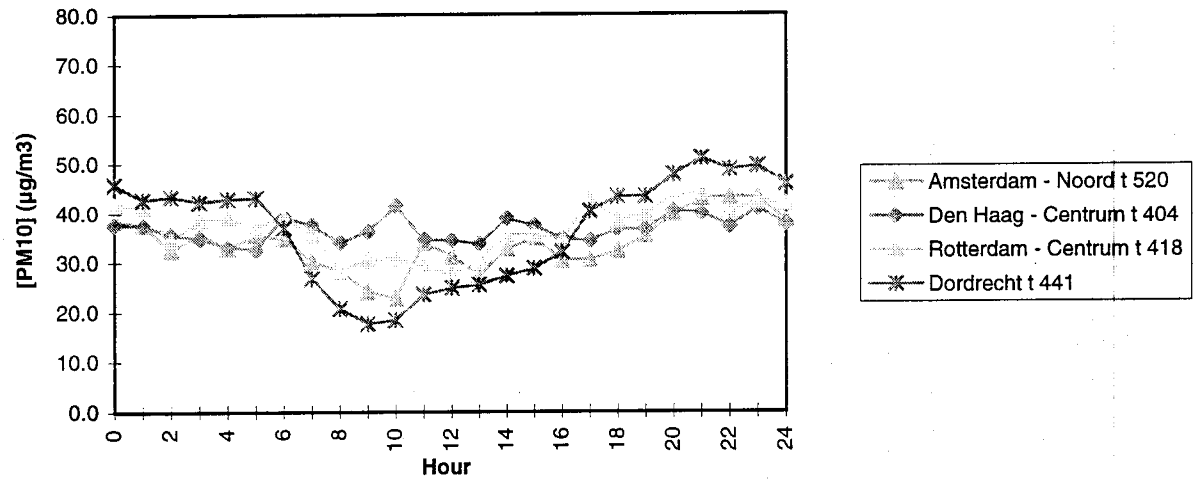

Figure 12: Course of PM10 during the day, town stations in summer 1994.

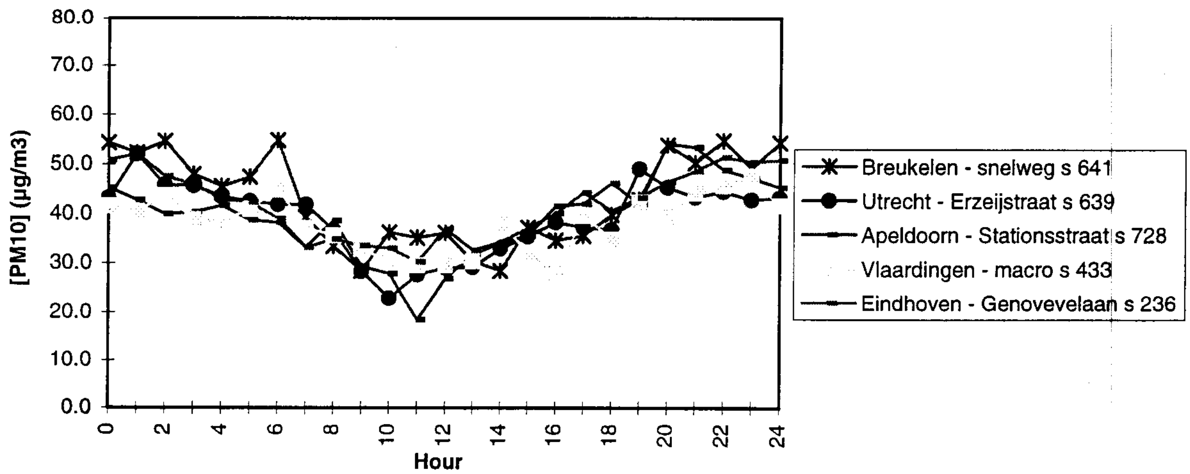

Figure 13: Course of PM10 during the day, street stations in summer 1994.

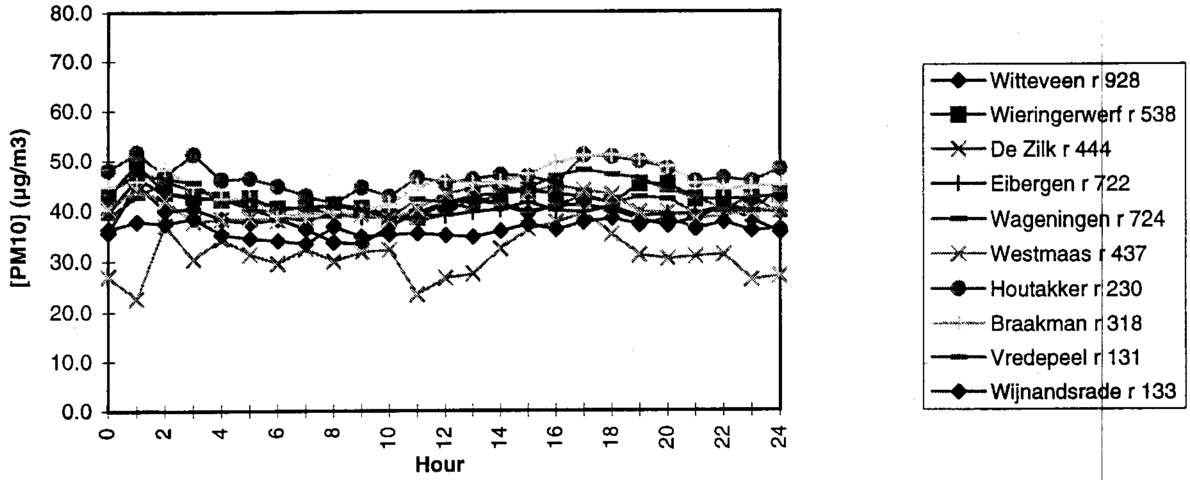

Figure 14: Course of PM10 during the day, rural stations in winter 1994.

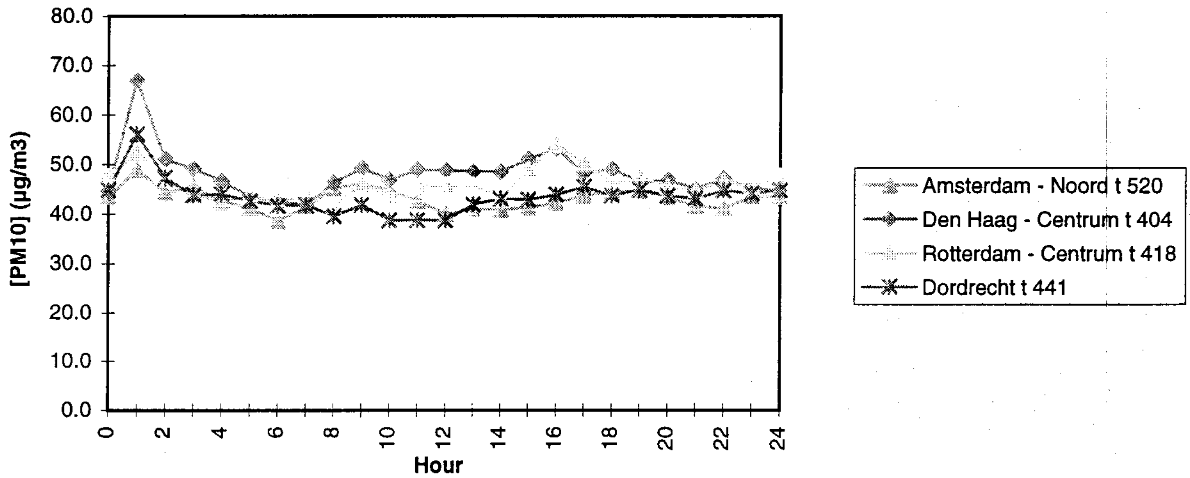

Figure 15: Course of PM10 during the day, town stations in winter 1994.

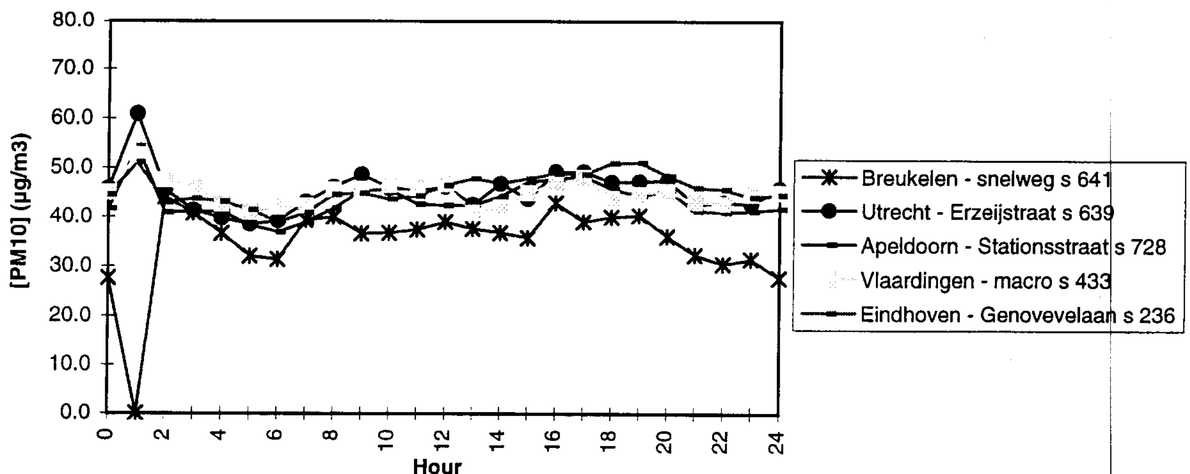

Figure 16: Course of PM10 during the day, street stations in winter 1994.

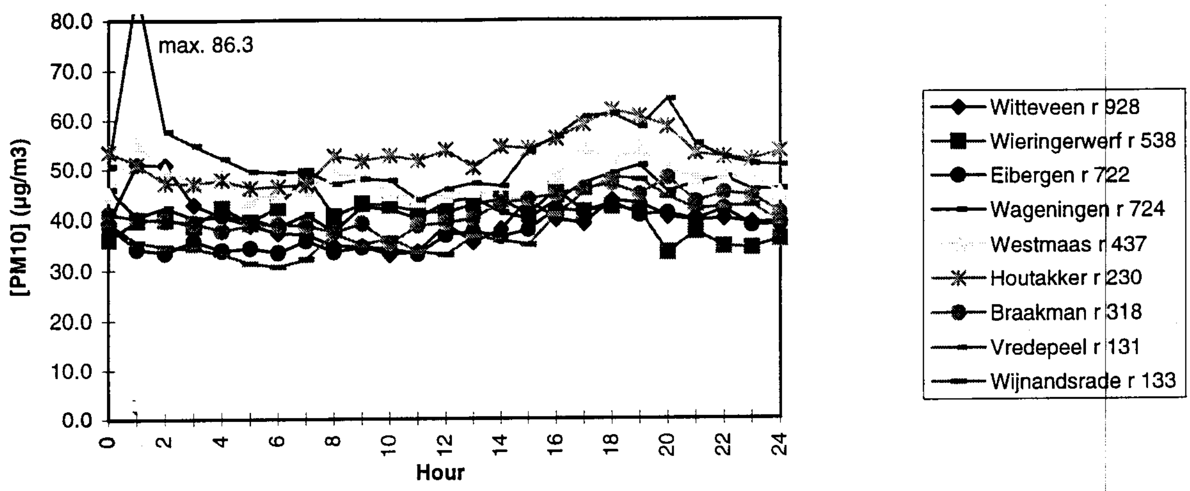

Figure 17: Course of PM10 during the day, weekdays (Mon-Fri) in autumn 1993, rural stations.

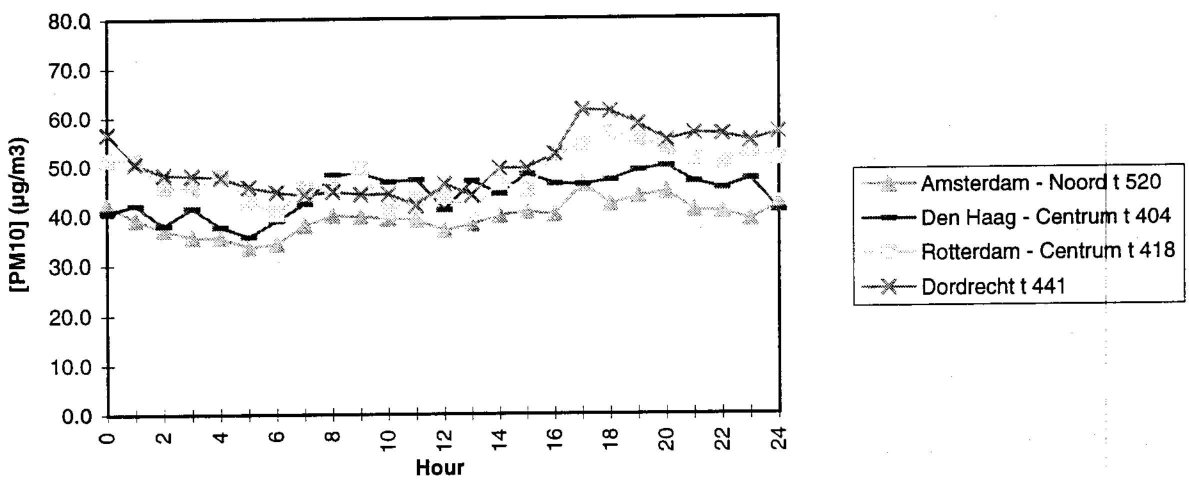

Figure 18: Course of PM10 during the day, weekdays (Mon-Fri) in autumn 1993, town stations.

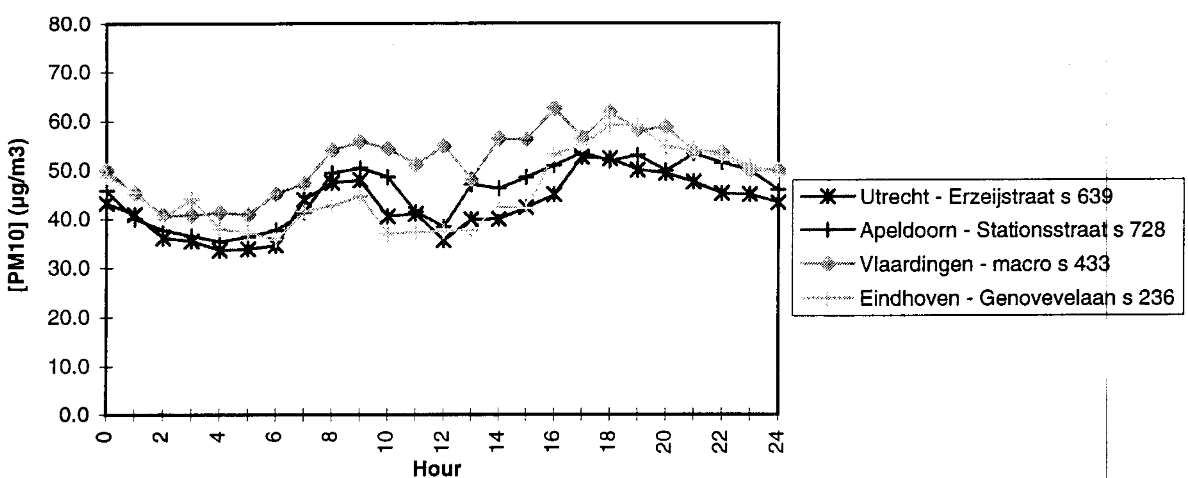

Figure 19: Course of PM10 during the day, weekday (Mon-Fri) in autumn 1993, street stations.

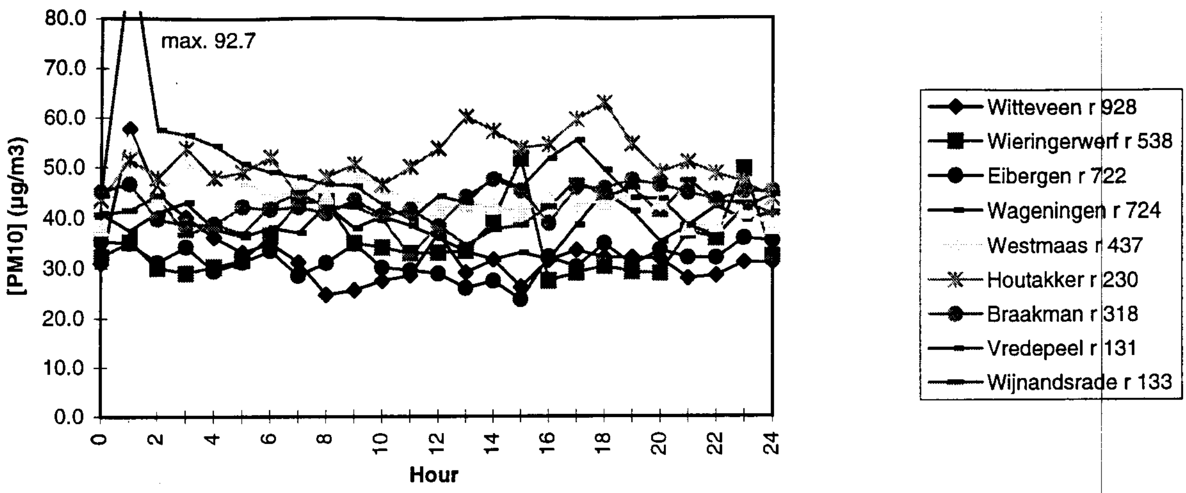

Figure 20: Course of PM10 during the day, weekends (Sat-Sun) in autumn 1993, rural stations.

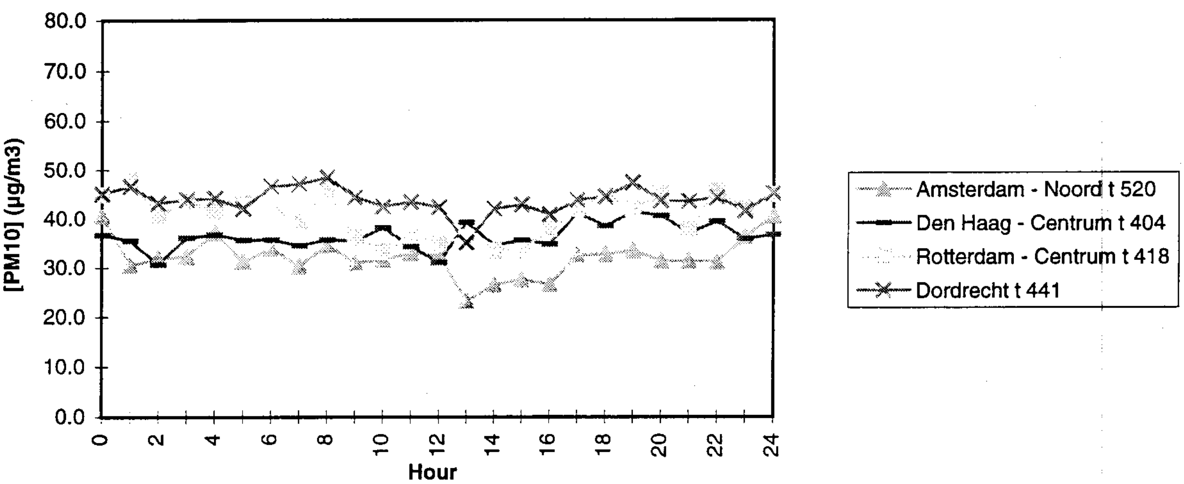

Figure 21: Course of PM10 during the day, weekends (Sat-Sun) in autumn 1993, town stations.

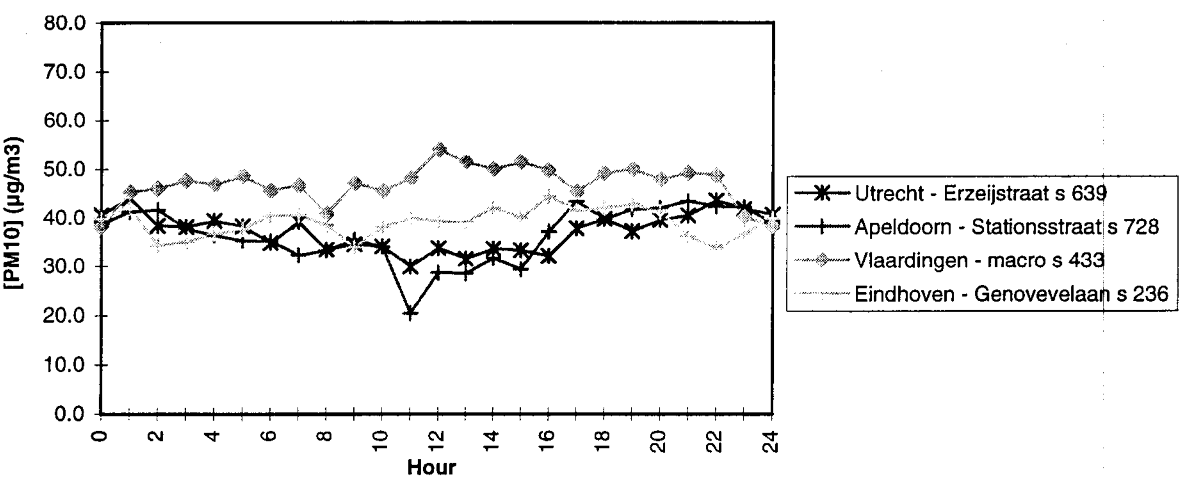

Figure 22: Course of PM10 during the day, weekends (Sat-Sun) in autumn 1993, street stations.

3.2.3 Diurnal pattern in the autumn (weekdays versus weekends)

Levels of PM10 in 1993 are lower in the weekend than on weekdays as is shown in Figure 17 to Figure 22 and Table 7. The average concentration difference is largest, more than 5 $\mu g/m^3$, for the rural stations with the lowest levels like Witteveen and Wieringerwerf. The difference is less for the other stations. From Monday to Friday the highest concentrations occur between 17:00 and 19:00.

In town, the weekend shows only a small difference with weekdays, as can be seen in Figure 21 and Figure 18 respectively. The recorded levels in Amsterdam-Noord are less than those in other cities (Rotterdam-Centrum, Dordrecht and Den Haag-Centrum). The average difference in concentration between weekdays and weekend is about 3 $\mu g/m^3$, which is comparable with the 'average' rural stations in Table 7.
For street stations the course of PM10 for weekdays is shown in Figure 19 and in Figure 22 for the weekend. The lowest concentrations are measured during the night. Levels of fine particles rise around 08:00 and again around 16:00. In both 1993 and 1994 the highest levels on workdays are recorded between 17:00 and 19:00. This pattern is possibly attributable to traffic. The average difference in PM10 between weekdays and weekend is about 6 $\mu g/m^3$, which is comparable in magnitude with the findings at rural stations with low levels of PM10, as can be seen in Table 7.

When comparing the results of 1993 and 1994 (Table 7) it is noticed that:
- Systematic differences between weekdays and weekends are caused by human activity, which is the controlling factor for anthropogenic emissions.
- When the air masses come from areas with many anthropogenic sources, levels are higher (1993) and the afore-mentioned difference is found.
- When the air masses are from a clean origin, with few anthropogenic sources, levels are low (1994) and little difference between weekdays and weekends is recorded.

Table 7: Levels of PM10 in $\mu g/m^3$ for 1993 and 1994 for weekdays and weekends in autumn (September, October and November)

		1993				1994			
		Mon-Fri		Sat-Sun		Mon-Fri		Sat-Sun	
		Mean	S.E.	Mean	S.E.	Mean	S.E.	Mean	S.E.
Rural									
	Low	40.1	0.6	33.8	0.9	23.5	0.4	25.1	0.4
	Average	41.5	0.6	38.7	0.6	27.7	0.4	28.9	0.5
	High	49.2	0.9	47.7	0.9	32.2	0.6	33.3	0.7
Town		41.5	0.6	38.3	0.6	30.2	0.4	30.0	0.5
Street		46.3	0.7	39.9	0.6	30.5	0.4	31.1	0.5

3.3 Spatial analysis

The spatial variation of PM10 concentrations is investigated using Principal Component Analysis (PCA) in §3.3.1 and by analysing windroses as presented in §3.3.2. This spatial analysis is used in the apportionment of PM10 to different sources, which is treated in §3.3.3. Also part of the spatially oriented analyses in this report are the correlations between the KNMI stations for the meteorological variables and between the LML stations for PM10 levels, as reported in §3.3.4 and §3.3.5, respectively.

3.3.1 Principal Component Analysis and spatial patterns in PM10

Table 8 summarises the results of the Principal Component Analysis of PM10 data. The large amount of variance explained by the first principal component, shows that a strong correlation in time exists between changes in PM10 concentrations at all LML stations. Here, time can be seen as representing meteorology. Changes in the weather occur almost simultaneous at all stations. This indicates that concentration variations are determined by large scale (meteorological) phenomena. The second component indicates a north-south gradient. The third principal component shows a contrast between regional (background) stations, on the one hand, and town/street stations, on the other hand. An explanation for this systematic difference is the presence of local sources in urbanised areas.

Table 8: Summary of Principal Component Analysis using PM10 data

	First component	Second component	Third component
Variance explained	85 %	7 %	4 %
Score < 0	-	Wieringerwerf Witteveen	Den Haag - Centrum Amsterdam - Noord Utrecht - Erzeijstr. Apeldoorn - Stationsstr. Rotterdam - Centrum
Score > 0	Largest: Vredepeel Houtakker Smallest: Wijnandsrade	Wijnandsrade Braakman	Vredepeel Wijnandsrade Houtakker Witteveen

The assumed presence of local sources in urbanised areas is associated with only 4% of the total variance. Their magnitude can therefore not be determined from analysing only total PM10 levels. The strong temporal correlation and the weak north-south gradient on the second component completely mask local sources in the yearly average values (Table 9).

Table 9: Average yearly PM10 values (μg/m³) for all LML stations in 1993 and 1994

		Yearly average	Yearly average
Station [1]		1993	1994
Witteveen r	Witte928	36.0	33.1
Wieringerwerf r	Wieri538	32.3	34.2
De Zilk r	De Z444	~	30.7
Eibergen r	Eiber722	36.4	37.1
Wageningen r	Wagen724	43.0	41.5
Westmaas r	Westm437	41.9	32.3
Houtakker r	Houta230	46.6	47.5
Braakman r	Braak318	44.2	41.1
Vredepeel r	Vrede131	49.7	43.2
Wijnandsrade r	Wijna133	39.1	37.4
Breukelen-snelweg s	Breuk641	~	42.3
Utrecht-Erzeijstraat s	Utrec639	42.4	39.8
Apeldoorn-Stationsstraat	Apeld728	39.2	39.0
Vlaardingen-macro s	Vlaar433	40.7	39.5
Eindhoven-Genovevelaan	Eindh236	41.8	41.9
Amsterdam-Noord t	Amste520	41.0	37.1
Den Haag-Centrum t	Den H404	41.0	41.2
Rotterdam-Centrum t	Rotte418	41.2	40.6
Dordrecht t	Dordr441	41.2	37.5

[1]: r = rural, t = town, s = street.

3.3.2 Wind direction

In order to study the relationship between the wind direction and the associated levels of PM10, windroses have been constructed for all 19 LML stations using meteorological data from the nearest KNMI station. Table 12 summarises for which periods windroses have been made. Windroses were made not only for the whole year but for winter (Jan.-Mar. + Oct.-Dec.) and summer (Apr.-Sep.) separately as well.
To illustrate the influence of wind direction on the PM10 levels the following windroses are presented: Rural 1993, Wijnandsrade 133; Town 1993, Amsterdam-Noord 520; and Street 1993, Apeldoorn-Stationsstraat 728 (Figure 23 through Figure 25). Next are two windroses that illustrate the difference between summer and winter. Selected for this was the rural station Eibergen 722 (Figure 26 and Figure 27). The frequency of winds from a given direction is given for four sectors for winter (Table 10) and summer (Table 11) separately.

Table 10: Frequency distribution for Wind direction in winter.

North-west 14%	14% North-east
South-west 48%	24% South-east

Table 11: Frequency distribution for Wind direction in summer.

North-west 25%	24% North-east
South-west 36%	15% South-east

From Figure 23 to Figure 31 it becomes clear that in the Netherlands south-easterly winds carry the heaviest load of fine particles. This is much more pronounced during the winter than during the summer. The reason why during the summer of 1994 North-westerly winds were carrying more fine particles than during winter is unknown. This could be a point for further study. A role can be played by sea salt and other local sources, that are influenced by the dry summer weather of 1994. If local sources are not capable of explaining the found differences, trajectories may help to find an explanation based on transport from foreign areas.

Table 12: Summary of windroses made to study the relationship between PM10 levels and wind direction.

Station	1993	1993 Summer	1993 Winter	1994	1994 Summer	1994 Winter
Witteveen 928	+	+	+	+	+	+
Wieringerwerf 538	+	+	+	+	+	+
Amsterdam-Noord 520	+	+	+	+	+	+
De Zilk 444	-	-	-	+	+	+
Breukelen-snelweg 641	-	-	-	+	+	+
Utrecht-Erzeijstraat 639	+	+	+	+	+	+
Eibergen 722	+	-	-	+	+	+
Apeldoorn-Stationsstraat 728	+	-	-	+	+	+
Wageningen 724	+	-	-	+	+	+
Den Haag-centrum 404	+	+	+	+	+	+
Vlaardingen-macro 433	+	-	-	+	+	+
Rotterdam-centrum 418	+	-	-	+	+	+
Westmaas 437	+	-	-	+	+	+
Dordrecht	+	-	-	+	+	+
Houtakker 230	+	-	-	+	+	+
Braakman 318	+	-	-	+	+	+
Eindhoven-Genovevelaan 236	+	+	+	+	+	+
Vredepeel 131	+	+	+	+	+	+
Wijnandsrade 133	+	-	-	+	+	+

To analyse the influence of wind speed on PM10 concentrations, the windroses (Figure 23 to Figure 27) were made with a subdivision based on the wind speed (Table 13). As can be seen from the windroses presented there is no clear influence of wind speed on PM10.

Table 13: Wind speed classes as used in generating windroses.

Wind speed (0.1 m/s)	Beaufort-scale	Description
[0 - 50]	up to 3 Bf.	moderate wind
<50 - 100]	4-5 Bf.	fairly strong wind
<100 - 150]	6 (-7) Bf.	strong wind
<150 - ...]	7 Bf. and stronger	storm

Finally, the windroses for 1993 and 1994 taken together are given for the Background, Agriculture, Town and Street stations in Figure 28 to Figure 31. This is to aid in the interpretation of the results of both multiple linear regression and Kalman-filtering. In this case, data from the KNMI station in De Bilt have been used for all four stations (see §3.3.4). In the graphs 95% confidence limits around the mean are indicated. The relatively wide range for winds between east and south are a result of the contrast between observations during summer and winter (cf. Figure 26 and Figure 27).

Wijnandsrade 133 WindDirection (degr.)	PM10 All Data.	PM10 [0-50]	PM10 <50-100]	PM10 <100-150]	PM10 <150-..]
0	55	55	-	-	-
10	23	23	-	-	-
20	28	28	-	-	-
30	37	42	27	-	-
40	60	60	-	-	-
50	70	68	83	-	-
60	56	56	-	-	-
70	50	47	78	-	-
80	55	55	-	-	-
90	47	47	-	-	-
100	72	72	-	-	-
110	-	-	-	-	-
120	64	64	-	-	-
130	54	54	-	-	-
140	49	49	-	-	-
150	72	72	-	-	-
160	28	28	29	-	-
170	55	55	-	-	-
180	25	25	-	-	-
190	26	30	20	-	-
200	31	35	22	-	-
210	28	35	20	-	-
220	33	50	22	21	-
230	34	50	23	6	-
240	36	48	23	21	-
250	36	40	11	29	-
260	28	27	30	-	-
270	44	49	21	-	-
280	26	25	32	-	-
290	37	37	-	-	-
300	25	25	-	-	-
310	21	-	21	-	-
320	10	10	-	-	-
330	28	28	-	-	-
340	23	23	-	-	-
350	41	49	26	-	-

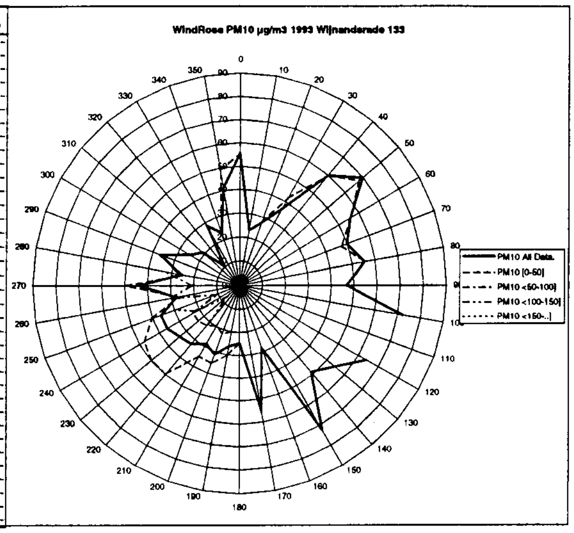

Figure 23: Windrose 1993 Rural, Wijnandsrade 133.

Amsterdam-Noord 520 WindDirection (degr.)	PM10 All Data.	PM10 [0-50]	PM10 <50-100]	PM10 <100-150]	PM10 <150-..]
0	29	29	-	-	-
10	26	26	22	-	-
20	18	18	-	-	-
30	26	27	20	-	-
40	32	31	35	-	-
50	33	33	33	-	-
60	51	48	54	-	-
70	54	49	66	69	-
80	43	43	-	-	-
90	50	44	81	-	-
100	44	46	25	-	-
110	69	69	-	-	-
120	64	64	62	-	-
130	68	68	-	-	-
140	51	-	51	-	-
150	66	69	52	-	-
160	74	74	-	-	-
170	44	58	17	-	-
180	54	58	32	-	-
190	59	79	24	-	-
200	49	63	26	-	-
210	40	45	25	-	-
220	50	55	43	-	-
230	38	48	27	19	-
240	35	50	26	30	-
250	34	32	36	34	-
260	28	28	27	-	-
270	39	43	30	43	-
280	30	32	26	-	-
290	27	26	32	-	-
300	26	20	25	41	-
310	27	27	-	-	-
320	31	35	6	-	-
330	26	27	24	33	-
340	17	11	35	-	-
350	16	13	21	-	-
360	10	16	4	-	-

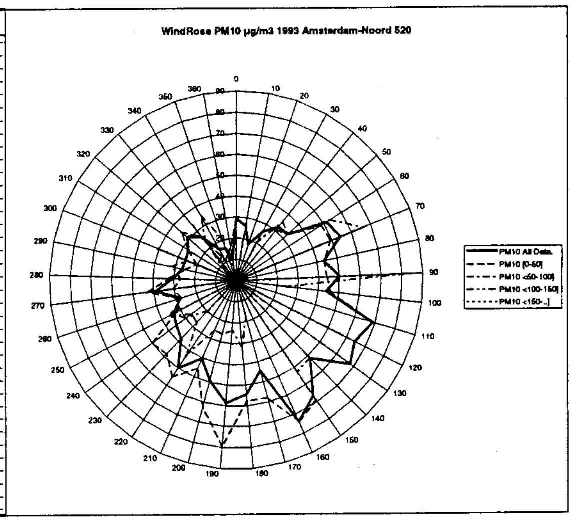

Figure 24: Windrose 1993 Town, Amsterdam-Noord 520.

Apeldoorn-Stationsstraat 728 WindDirection (degr.)	PM10 All Data.	PM10 [0-50]	PM10 <50-100]	PM10 <100-150]	PM10 <150-..]
0	25	25	-	-	-
10	30	30	-	-	-
20	28	28	-	-	-
30	26	27	25	-	-
40	41	45	38	-	-
50	51	51	-	-	-
60	50	43	80	-	-
70	64	64	-	-	-
80	58	56	68	-	-
90	49	49	-	-	-
100	60	60	-	-	-
110	100	100	-	-	-
120	59	59	-	-	-
130	36	36	-	-	-
140	52	56	32	-	-
150	77	77	-	-	-
160	33	33	-	-	-
170	46	49	31	-	-
180	37	38	29	-	-
190	54	65	22	-	-
200	25	27	17	-	-
210	42	46	21	-	-
220	28	33	15	-	-
230	31	33	20	-	-
240	32	33	26	-	-
250	34	35	30	22	-
260	29	32	20	-	-
270	28	28	22	-	-
280	36	36	-	-	-
290	24	24	-	-	-
300	20	20	-	-	-
310	29	29	-	-	-
320	19	19	-	-	-
330	24	24	-	-	-
340	26	26	-	-	-
350	25	25	-	-	-
360	29	28	35	-	-

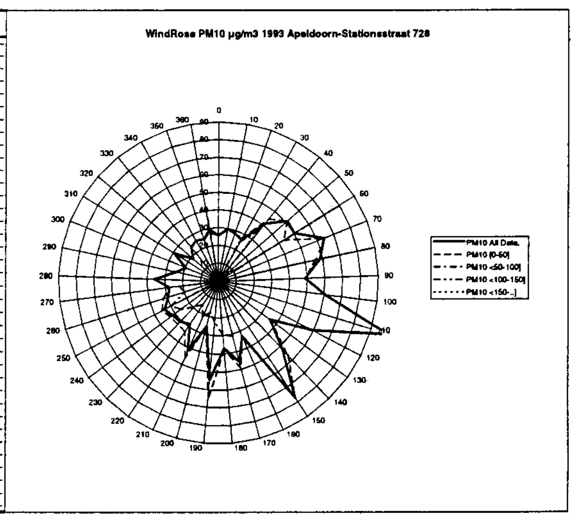

Figure 25: Windrose 1993 Street, Apeldoorn-Stationsstraat 728.

Eibergen 722 WindDirection (degr.)	PM10 Summer	PM10 [0-50]	PM10 <50-100]	PM10 <100-150]	PM10 <150-..]
0	69	69	-	-	-
10	21	16	41	-	-
20	36	34	42	-	-
30	28	28	-	-	-
40	36	37	32	-	-
50	31	25	42	-	-
60	42	45	27	-	-
70	42	42	-	-	-
80	37	37	-	-	-
90	40	41	38	-	-
100	25	25	-	-	-
110	45	45	-	-	-
120	40	40	-	-	-
130	57	57	-	-	-
140	36	36	-	-	-
150	63	63	-	-	-
160	37	44	23	-	-
170	44	44	-	-	-
180	26	26	-	-	-
190	43	44	41	-	-
200	31	31	-	-	-
210	25	28	20	-	-
220	31	28	46	-	-
230	54	54	-	-	-
240	36	37	32	-	-
250	31	30	49	-	-
260	36	36	-	-	-
270	25	25	-	-	-
280	28	28	-	-	-
290	36	36	-	-	-
300	49	49	-	-	-
310	9	9	-	-	-
320	60	60	-	-	-
330	51	51	-	-	-
340	21	21	-	-	-
350	49	57	33	-	-

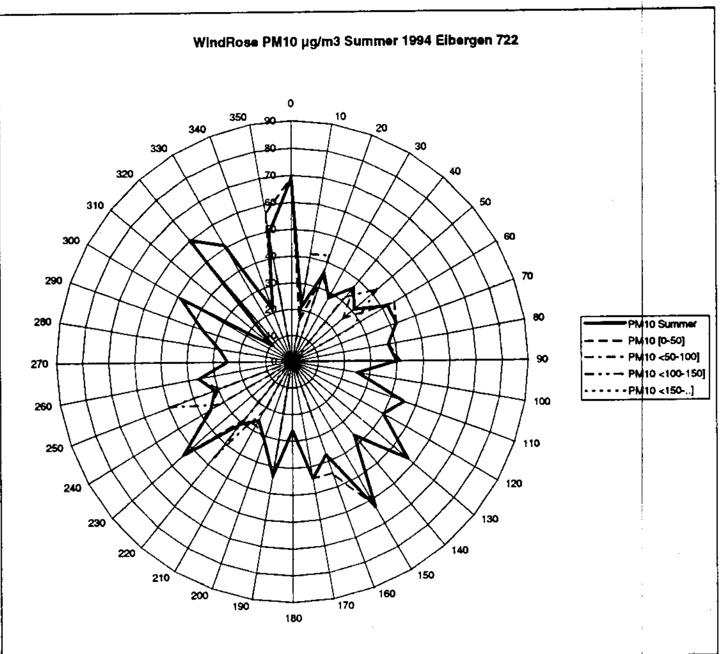

Figure 26: Windrose summer 1994 Rural, Eibergen 722.

Eibergen 722 WindDirection (degr.)	PM10 Winter	PM10 [0-50]	PM10 <50-100]	PM10 <100-150]	PM10 <150-..]
0	-	-	-	-	-
10	81	81	-	-	-
20	-	-	-	-	-
30	79	79	-	-	-
40	29	29	-	-	-
50	56	56	-	-	-
60	22	16	28	-	-
70	68	74	44	-	-
80	68	68	-	-	-
90	66	57	85	-	-
100	54	61	27	-	-
110	43	43	-	-	-
120	49	49	-	-	-
130	52	52	-	-	-
140	54	54	-	-	-
150	46	44	54	-	-
160	35	35	-	-	-
170	71	135	50	-	-
180	38	34	40	-	-
190	31	33	23	-	-
200	32	31	33	-	-
210	33	38	27	-	-
220	37	42	26	-	-
230	26	35	19	-	-
240	31	33	29	-	-
250	23	25	19	-	-
260	27	30	20	-	-
270	11	11	13	-	-
280	25	25	-	-	-
290	15	16	15	-	-
300	23	23	-	-	-
310	10	10	-	-	-
320	-	-	-	-	-
330	25	25	-	-	-
340	8	8	-	-	-
350	24	24	-	-	-

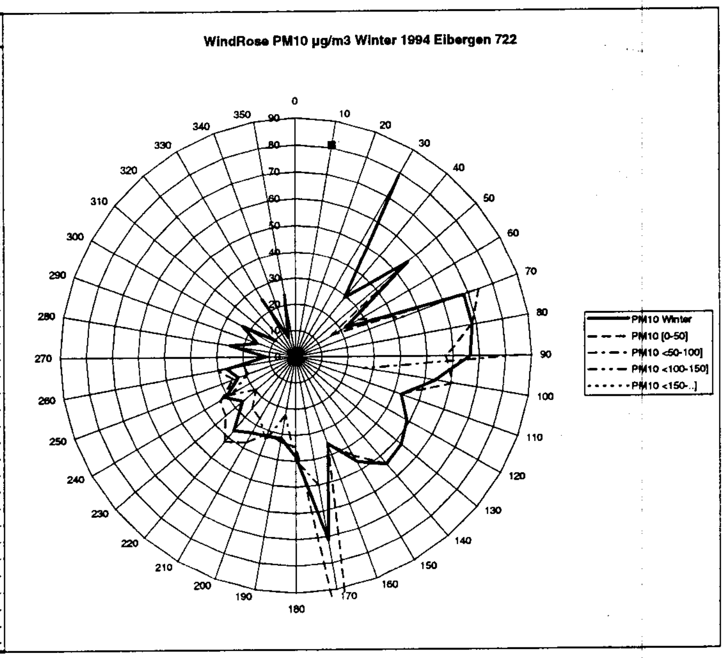

Figure 27: Windrose winter 1994 Rural, Eibergen 722.

Background				
WindDirection (degr)	PM10	n	s	± 95% conf.
0	35.8	3	4.8	5.4
10	19.9	12	5.5	3.1
20	19.2	16	12.5	6.1
30	26.9	19	15.7	7.0
40	28.5	16	12.9	6.3
50	32.8	11	19.0	11.3
60	42.4	21	24.7	10.6
70	43.2	17	27.7	13.2
80	51.1	17	31.9	15.2
90	59.2	16	64.4	31.5
100	45.1	13	23.2	12.6
110	61.2	11	38.0	22.5
120	49.0	23	34.9	14.3
130	58.9	16	21.1	10.3
140	50.0	19	21.0	9.4
150	51.6	12	27.9	15.8
160	54.2	10	35.4	21.9
170	37.0	15	11.8	6.0
180	40.6	24	28.3	11.3
190	38.0	27	19.8	7.5
200	37.2	21	22.7	9.7
210	34.9	40	23.7	7.3
220	28.5	48	14.8	4.2
230	25.2	49	11.1	3.1
240	27.6	42	10.6	3.2
250	25.9	37	10.9	3.5
260	24.2	28	9.2	3.4
270	24.4	23	9.1	3.7
280	26.2	19	12.6	5.7
290	20.9	11	7.0	4.1
300	23.9	11	18.0	10.7
310	22.3	12	9.4	5.3
320	22.8	12	13.4	7.6
330	22.7	15	15.7	7.9
340	25.1	8	9.8	6.8
350	22.3	11	11.7	6.9
360	17.8	5	9.4	8.2
Average	33.7	710	23.7	1.7

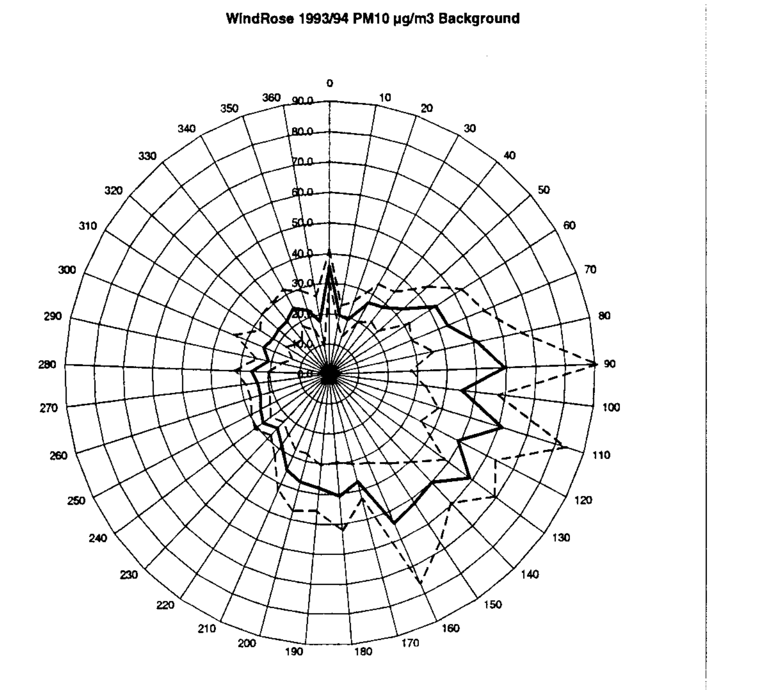

Figure 28: Windrose 1993/1994 Background.

Agriculture				
WindDirection (degr)	PM10	n	s	± 95% conf.
0	54.7	3	7.5	8.5
10	29.0	13	14.1	7.6
20	41.6	16	27.1	13.3
30	48.3	21	26.3	11.2
40	52.3	17	27.0	12.9
50	58.1	11	21.6	12.8
60	66.6	22	31.5	13.2
70	63.9	17	24.7	11.8
80	76.2	18	38.0	17.6
90	65.5	18	22.5	10.4
100	61.1	14	22.2	11.6
110	71.1	11	36.7	21.7
120	68.1	23	59.3	24.2
130	76.5	16	26.9	13.2
140	60.0	20	31.8	13.9
150	55.5	12	31.2	17.6
160	51.3	11	33.0	19.5
170	41.3	15	17.8	9.0
180	40.7	24	24.4	9.8
190	44.3	28	20.8	7.7
200	40.1	22	29.8	12.4
210	39.2	41	28.2	8.6
220	37.1	48	22.2	6.3
230	33.6	49	18.9	5.3
240	39.2	42	20.2	6.1
250	33.0	38	14.1	4.5
260	40.5	28	25.3	9.4
270	35.8	24	13.2	5.3
280	39.9	19	23.0	10.3
290	33.6	11	15.8	9.4
300	32.9	11	22.1	13.1
310	37.9	13	20.1	10.9
320	41.1	13	34.2	18.6
330	38.5	15	24.6	12.5
340	31.9	8	21.0	14.6
350	37.6	12	20.9	11.8
360	34.1	6	23.5	18.8
Average	45.7	730	28.7	2.1

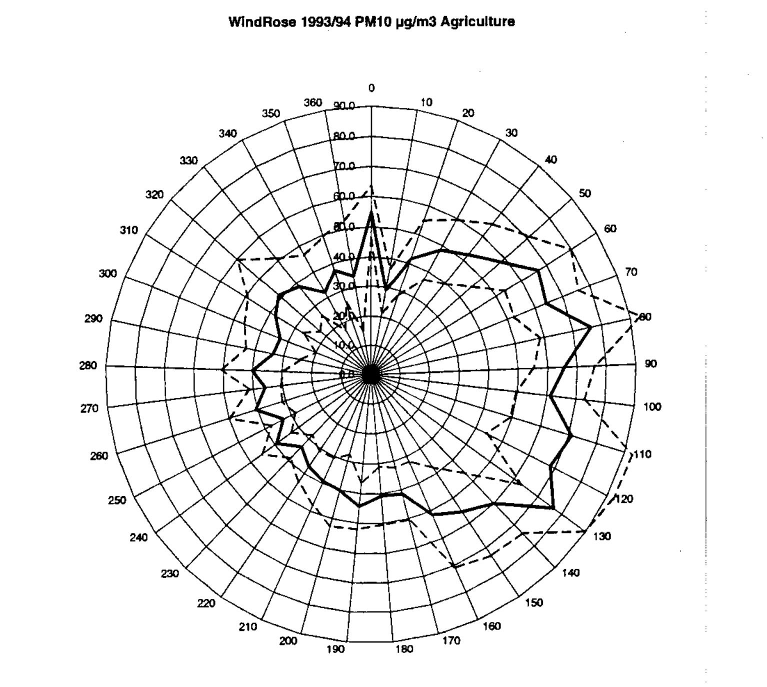

Figure 29: Windrose 1993/1994 Agriculture.

WindDirection (degr)	Town PM10	n	s	± 95% conf.
0	42.5	3	8.7	9.8
10	20.9	13	6.4	3.5
20	26.8	16	19.3	9.4
30	30.9	21	21.7	9.3
40	34.8	17	23.0	11.0
50	40.2	11	18.4	10.9
60	48.9	22	27.4	11.5
70	47.8	17	25.4	12.1
80	57.0	18	29.3	13.5
90	50.7	18	20.1	9.3
100	54.4	14	21.5	11.3
110	66.9	11	32.4	19.1
120	69.4	23	56.9	23.3
130	75.5	16	23.4	11.5
140	61.5	20	34.8	15.3
150	56.7	12	31.3	17.7
160	57.6	11	37.8	22.3
170	48.9	15	22.5	11.4
180	43.1	24	31.3	12.5
190	39.8	28	21.6	8.0
200	40.8	22	20.7	8.7
210	36.6	41	23.7	7.2
220	34.3	48	18.5	5.2
230	30.7	49	10.6	3.0
240	33.4	42	11.9	3.6
250	31.9	38	12.0	3.8
260	31.5	28	7.9	2.9
270	29.2	24	8.2	3.3
280	33.2	19	9.5	4.3
290	25.1	11	9.5	5.6
300	28.2	11	13.8	8.2
310	29.2	13	9.7	5.3
320	30.3	13	14.7	8.0
330	26.3	15	13.8	7.0
340	26.7	8	12.8	8.9
350	31.8	12	12.3	7.0
360	28.4	6	8.3	6.7
Average	39.7	730	25.1	1.8

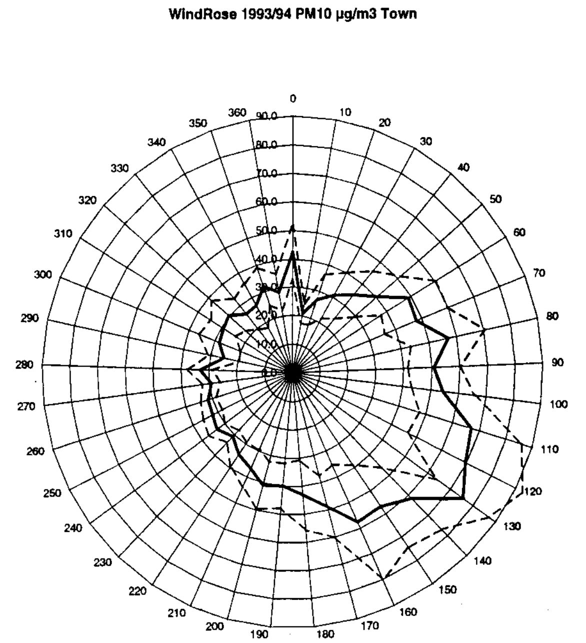

Figure 30: Windrose 1993/1994 Town.

WindDirection (degr)	Street PM10	n	s	± 95% conf.
0	48.7	3	11.1	12.5
10	25.4	13	8.9	4.9
20	35.0	16	23.9	11.7
30	42.3	21	24.2	10.3
40	43.9	17	26.5	12.6
50	48.6	11	20.2	11.9
60	59.3	22	29.4	12.3
70	54.9	17	23.4	11.1
80	65.5	18	35.1	16.2
90	55.5	18	16.0	7.4
100	56.8	14	21.8	11.4
110	65.9	11	39.8	23.5
120	66.6	23	55.0	22.5
130	76.7	16	25.6	12.5
140	59.4	20	32.5	14.2
150	55.3	12	30.1	17.0
160	54.2	11	35.5	21.0
170	47.5	15	22.0	11.1
180	40.5	24	27.7	11.1
190	42.9	28	24.2	9.0
200	38.8	22	21.4	8.9
210	39.6	41	26.0	8.0
220	34.3	48	19.9	5.6
230	31.2	49	13.3	3.7
240	36.3	42	15.6	4.7
250	31.7	38	12.1	3.8
260	33.9	28	18.1	6.7
270	32.3	24	12.2	4.9
280	33.4	19	15.3	6.9
290	28.5	11	8.2	4.8
300	29.9	11	19.2	11.4
310	33.5	13	13.6	7.4
320	35.0	13	16.0	8.7
330	30.8	15	15.7	7.9
340	27.5	8	14.0	9.7
350	34.6	12	21.0	11.9
360	33.7	6	21.0	16.8
Average	42.2	730	26.1	1.9

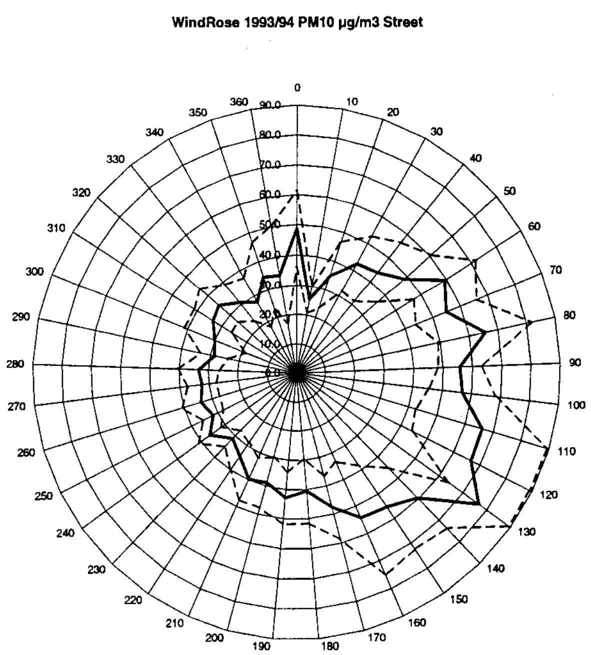

Figure 31: Windrose 1993/1994 Street.

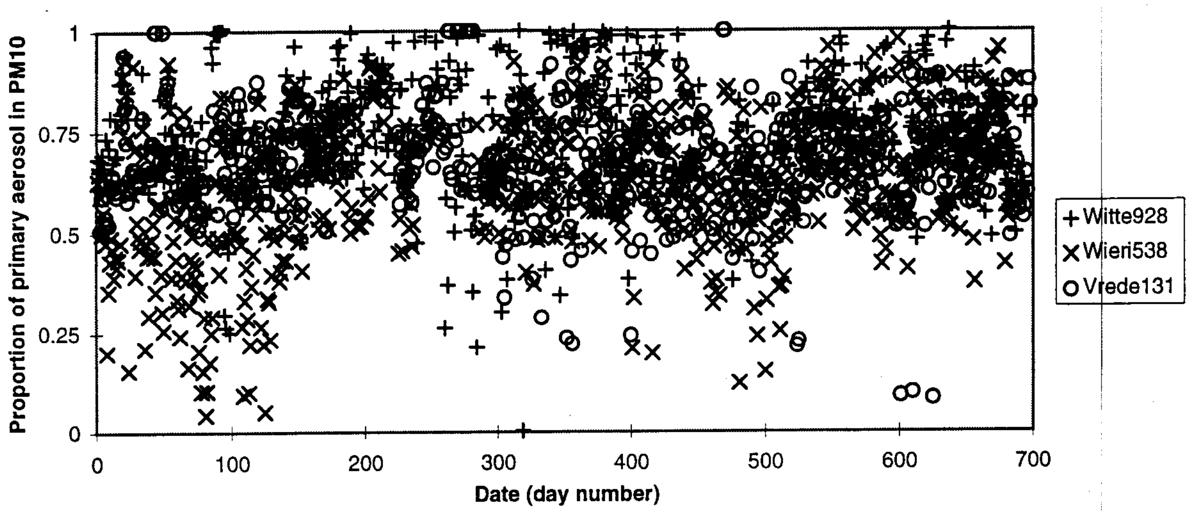

Figure 32: Ratio of primary to secondary aerosol at rural stations during the study period.

3.3.3 Source apportionment

As part of the spatial component of the behaviour of fine particles, the possibility of attributing specific portions of PM10 to specific sources is a question of interest. A few topics relating to this question, like benzene as a marker for traffic, the importance of sea spray and the ratio of primary to secondary aerosol, will be addressed in this paragraph.

The importance of traffic emissions for levels of fine particles can be studied by combining PM10 with benzene, where benzene is used as a marker for traffic (intensity). Simply by studying levels and correlations, a first-order approach to answer this specific question can be made (Table 14). Rural stations have lower PM10 and lower benzene levels than stations situated in cities. Street sites within cities have higher values than town stations for both, especially benzene. The correlation between PM10 and benzene becomes stronger in the same direction (rural - town - street).

Table 14: Mean levels of PM10 and benzene and their correlation for some LML stations

Station	[1]	PM10 (µg/m^3)	Benzene (µg/m^3)	Correlation coefficient
Witteveen	r	34.2	0.8	0.30
Rotterdam	t	40.0	2.5	0.50
Dordrecht	t	37.4	2.6	0.65
Utrecht-Erzeijstraat	s	40.6	4.7	0.65
Eindhoven-Genovevelaan	s	41.4	5.6	0.50
Apeldoorn-Stationsstraat	s	39.0	3.9	0.75

[1]: station type, r = rural, t = town, s = street.

Another way of distinguishing particles within PM10 is a division into primary (mainly carbonaceous) and secondary aerosol (sulphates/nitrates/ammonium). Also the contribution of sea salt (chloride plus an equivalent amount of sodium, stemming from sea spray) to PM10 can be distinguished. Figure 32 shows that at rural stations the ratio of primary to secondary aerosol fluctuates in a wide band around 0.6. A substantial contribution of secondary aerosol, around 40% on average, is in agreement with the small differences in total levels of fine particles countrywide.

From differences between the sum total for secondary aerosol and the same minus sodium chloride, the sea salt contribution to observed levels of PM10 is calculated. At coastal stations around 10% of yearly average PM10 concentrations (3-4 $\mu g/m^3$) can be sea salt. For stations further inland in the eastern parts of the country this contribution drops to less than 5% (Table 15).

Table 15: Sea spray contribution to PM10 at inland and coastal rural sites

Station	Primary PM10	(Primary - Sea salt) PM10	Sea salt
Witteveen 928	0.64	0.60	4%
Wieringerwerf 538	0.4	0.3	10%
De Zilk	0.6	0.5	10%

The ratio of primary to secondary aerosol is generally constant. The ratio can however deviate considerably from its usual value, around 0.6, during specific episodes.

One example of such an episode is the one that occurred in the middle of February 1994, when strong easterly winds blew, combining forces with severe frost and dry weather. The

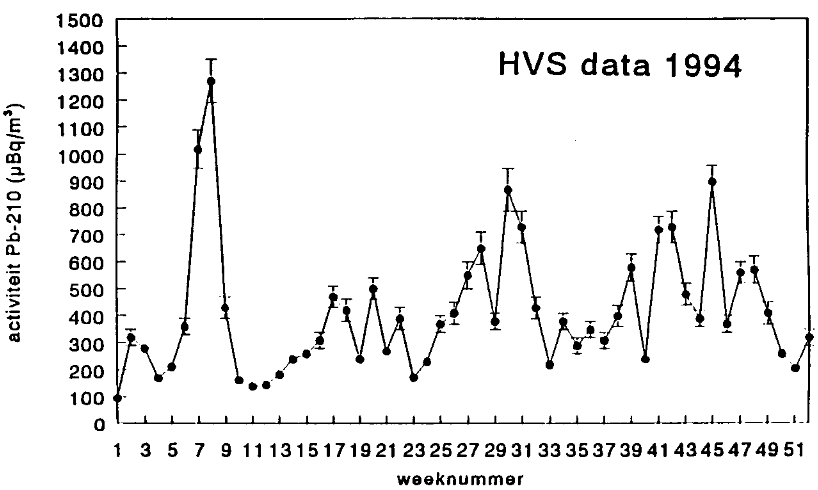

Figure 33: ^{210}Pb activity during 1994.

x-axis: week number in 1994, y-axis: ^{210}Pb-activity ($\mu Bq/m^3$)

importance of wind speed in this episode, is supported by Bloemen *et al.* (1995), who report an increase of PM10 during this episode for wind speeds over 12 m/s using hourly values. Much of this airborne dust is likely to be resuspended soil dust, resulting in a primary aerosol ratio of almost 1.00. The hypothesised resuspension of soil dust is substantiated by the fact that an unusually high amount of ^{210}Pb (Figure 33), an isotope which is a marker for soil-derived particles, has been measured during this episode (H. Reinen, personal communication).

3.3.4 Spatial correlations between the meteorological KNMI stations

Before starting the multiple-linear regression and Kalman-filtering it was decided to study the spatial correlation between the meteorological stations for all meteorological variables. If these stations correlate well it would not be necessary to use different weather information for the different stations.
Listed in Appendix C are the between-station correlations for all eight meteorological variables. For variables like wind direction and wind speed many stations have correlation coefficients of 0.80 or higher. The correlations for temperature, atmospheric pressure and insolation are 0.95 or higher for many stations. The variables with the lowest correlation coefficient are amount of precipitation and duration of precipitation. Of these two the amount of precipitation is the most variable, some correlations are even below 0.50.
There is a geographical influence on the correlations. The strength of the correlation decreases with increasing distance between the stations. Examples of stations that are far apart are De Kooy (north-west) and Maastricht (south-east) or Vlissingen (south-west) and Twente (east). This can best be seen in the tables on amount and duration of precipitation.

Based on these results it was decided to use data of just one weather station for further analysis. Bearing in mind the influence of geographical distance on the strength of the correlations the weather station De Bilt was selected. This station is located in the centre of the country, thus minimising the influence of geography.

Table 16: Descriptive statistics of meteorological parameters for KNMI station De Bilt for the whole period 1993-1994

	Temp.	*Duration of Precip.*	*Amount of Precip.*	*Rel. humid.*	*Atmosph. pressure*	*Insol-ation*	*Wind speed*	*MixHT*
	0.1 C	0.1h	0.1mm	%	0.1mbar	j/cm2	0.1m/s	m
Mean	101	46	45	83.0	10154	917	35	1395
Standard Error	2	2	3	0.4	4	28	1	27
Median	102	34	21	84.4	10158	701	30	1296
Mode	151	10	-1	92.8	10046	261	30	1006
Standard Deviation	61	41	63	10.4	96	749	18	585
Kurtosis	-0.3	2.4	5.5	0.7	0.2	-0.5	1.0	-0.4
Skewness	-0.1	1.6	2.2	-0.8	-0.2	0.8	1.0	0.5
Minimum	-77	1	-17	42	9833	16	0	219
Maximum	264	215	377	100	10406	2893	105	2888
n Observations	730	367	402	730	730	730	730	453

3.3.5 Spatial correlations between LML stations

As with the meteorological stations the correlations of the PM10 levels recorded at the LML stations is calculated. The correlation coefficients are listed in Table 18.

It can be seen that both town and street stations tend to share high correlations. Rural stations are less strongly correlated. Within the group of rural stations two subgroups can be distinguished. One is a group consisting of Witteveen 928, Wieringerwerf 538 and De Zilk 444, which are regional stations with relatively low levels of fine particles in the air. The other group consists of Vredepeel 131, Houtakker 230 and Braakman 318. These three all share a relatively high level of fine particles. Three more rural stations, Wageningen 724, Eibergen 722 and Westmaas 437, are also possible members of the second group.

Table 17: Descriptive statistics of PM10 (μg/m³) for the aggregated stations Background, Agriculture, Town and Street for the whole period 1993-1994

	Background	Agriculture	Town	Street
Mean	33.7	45.7	39.9	41.0
Standard Error	0.9	1.1	1.0	0.9
Median	27.2	37.8	31.8	33.1
Mode	19.9	14.6	19.9	37.5
Standard Deviation	23.7	28.7	27.2	25.5
Kurtosis	22.3	11.2	35.7	17.2
Skewness	3.3	2.2	4.0	2.8
Minimum	2.7	5.0	7.5	8.1
Maximum	287.9	309.6	379.1	304.5
n Observations	710	728	719	730

Based on the correlations it was decided to continue the analyses with four distinct aggregated stations. The aggregated PM10 stations have been labelled:

- Street;
- Town;
- Background (rural stations with low levels of fine particles) and
- Agriculture (rural stations with high PM10 concentrations, possibly as a result of agricultural activities).

The LML stations used as representatives for these types have been given in 4. To characterise the four aggregated stations Table 17 gives some descriptive statistics.

Table 18: Correlation matrices of PM10 on LML-stations in 1993 and 1994

Correlationmatrix PM10 1993

	Witte928	Wieri538	Amste520	Utrec639	Elber722	Apeld728	Wagen724	Den H404	Vlaar433	Rotte418	Westm437	Dordr441	Houta230	Braak318	Eindh236	Vrede131	Wijna133
Wijna133	.71	.60	.67	.76	.79	.71	.76	.70	.67	.84	.73	.85	.83	.79	.84	.87	1.00
Vrede131	.80	.71	.82	.87	.87	.84	.89	.81	.82	.93	.87	.94	.92	.87	.90	1.00	
Eindh236	.80	.72	.81	.88	.86	.89	.88	.84	.87	.94	.89	.94	.93	.87	1.00		
Braak318	.82	.78	.87	.90	.85	.85	.88	.88	.87	.93	.90	.94	.92	1.00			
Houta230	.84	.78	.85	.93	.89	.88	.94	.89	.89	.96	.93	.97	1.00				
Dordr441	.91	.93	.96	.97	.94	.96	.96	.97	.97	.99	.97	1.00					
Westm437	.82	.81	.89	.92	.84	.87	.93	.94	.96	.97	1.00						
Rotte418	.90	.93	.95	.97	.94	.95	.96	.97	.98	1.00							
Vlaar433	.80	.84	.91	.93	.84	.89	.92	.96	1.00								
Den H404	.84	.87	.93	.94	.84	.89	.92	1.00									
Wagen724	.89	.86	.94	.97	.94	.95	1.00										
Apeld728	.91	.84	.93	.95	.94	1.00											
Elber722	.90	.80	.88	.92	1.00												
Utrec639	.90	.86	.96	1.00													
Amste520	.90	.87	1.00														
Wieri538	.85	1.00															
Witte928	1.00																

Correlationmatrix PM10 1994

	Witte928	Wieri538	Amste520	De Zl444	Breuk641	Utrec639	Elber722	Apeld728	Wagen724	Den H404	Vlaar433	Rotte418	Westm437	Dordr441	Houta230	Braak318	Eindh236	Vrede131
Wijna133	.44	.46	.56	.45	.52	.68	.70	.68	.69	.60	.67	.66	.70	.70	.81	.75	.84	.80
Vrede131	.69	.70	.72	.65	.73	.80	.87	.83	.85	.70	.78	.77	.84	.83	.91	.76	.90	1.00
Eindh236	.55	.66	.79	.70	.76	.88	.85	.85	.85	.82	.86	.88	.88	.89	.91	.84	1.00	
Braak318	.56	.69	.80	.74	.69	.85	.72	.81	.76	.85	.90	.87	.90	.85	.85	1.00		
Houta230	.65	.69	.77	.73	.76	.85	.84	.84	.86	.76	.83	.82	.89	.85	1.00			
Dordr441	.61	.74	.88	.79	.81	.93	.80	.89	.82	.89	.93	.93	.93	1.00				
Westm437	.61	.75	.88	.87	.82	.91	.82	.88	.87	.88	.93	.93	1.00					
Rotte418	.61	.74	.90	.82	.80	.92	.78	.88	.82	.93	.95	1.00						
Vlaar433	.61	.72	.88	.81	.80	.92	.78	.88	.86	.91	1.00							
Den H404	.57	.73	.90	.83	.71	.90	.73	.87	.77	1.00								
Wagen724	.66	.71	.82	.78	.86	.87	.88	.89	1.00									
Apeld728	.70	.79	.91	.83	.83	.93	.89	1.00										
Elber722	.69	.75	.79	.69	.78	.84	1.00											
Utrec639	.65	.75	.91	.82	.84	1.00												
Breuk641	.76	.81	.84	.80	1.00													
De Zl444	.72	.87	.86	1.00														
Amste520	.69	.82	1.00															
Wieri538	.89	1.00																
Witte928	1.00																	

The labelling as Agriculture of the station aggregating two rural stations with high levels of fine particles, is not intended to signify that agricultural or bio-industrial activities are the sole source of the observed high levels. The presence of agricultural activity, among others pig farming, in the vicinity is known and can be traced by elevated levels of ammonia measured at Vredepeel 131. For comparison, yearly average ammonia concentrations (RIVM, 1994b and J. Aben, personal pommunication) of a few rural station are listed in Table 19 along with yearly average PM10 concentrations. Note that ammonia is short-lived in the atmosphere and is quickly transformed in to ammonium aerosol with a much longer lifetime. Annual average ammonium concentrations in the Netherlands are around 2 to 3 μg/m³ and do not show high spatial variability.

Table 19: Ammonia and PM10 concentrations at rural stations for 1993 and 1994

Station	$[NH_3]$ '93 (μg/m³)	[PM10] '93 (μg/m³)	$[NH_3]$ '94 (μg/m³)	[PM10] '94 (μg/m³)
Witteveen 928	2	36	2	33
Wieringerwerf 538	5	32	4	34
De Zilk 444	~	~	2	31
Eibergen 722	11	36	11	37
Vredepeel 131	21	50	21	43

3.4 Time-series analyses

As the data on PM10 and meteorology are available as a series of observations in time, it is logical to analyse them in ways that more or less explicitly use this information. In the following sections such analyses are presented.

3.4.1 Linear Regression.

Linear regressions have been calculated to look for a means of reconstructing - possibly even predicting - levels of PM10 based on meteorological parameters. To illustrate the relationship between fine particles and the meteorological parameters, graphs are presented plotting PM10 against each variable on a one-to-one basis. The presented graphs are based on data from the station labelled Background (Figure 34 to Figure 36). In the accompanying tables parameter values defining the regression lines equations for the other stations (Agriculture, Town and Street) are given.

In Figure 34 it can be seen that temperature has considerable influence on occurring levels of fine particles. Levels are low at normal temperatures between 5 and 20 °C but show a tendency to rise both at lower and at higher temperatures. A parabolic curve ($y=a.x^2 + b.x + c$) was fitted to the data as it can take on this form. The curves as calculated for all four stations are summarised in Table 20.

Table 20: Curve estimates ($y=a.x^2 + b.x + c$) for the relationship between Temperature (0.1 °C) and PM10 (μg/m³)

Station	a	b	c
Background	0.002	-0.5	55
Agriculture	0.003	-0.6	68
Town	0.002	-0.5	61
Street	0.002	-0.6	68

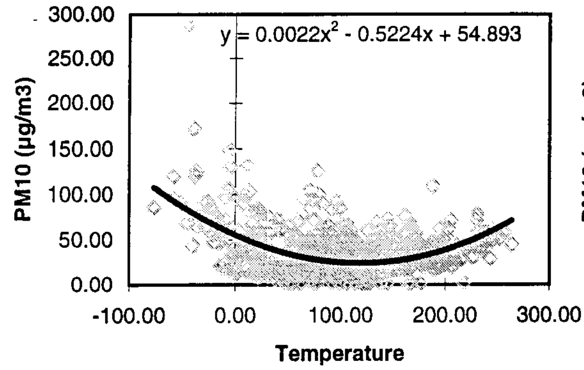

Figure 34: PM10 Background plotted against Temperature.

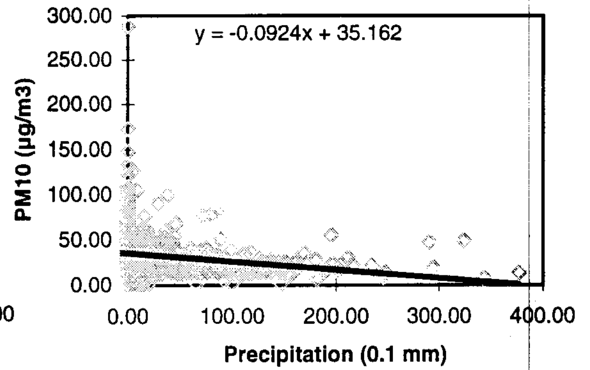

Figure 39: PM10 Background plotted against Amount of Precipitation.

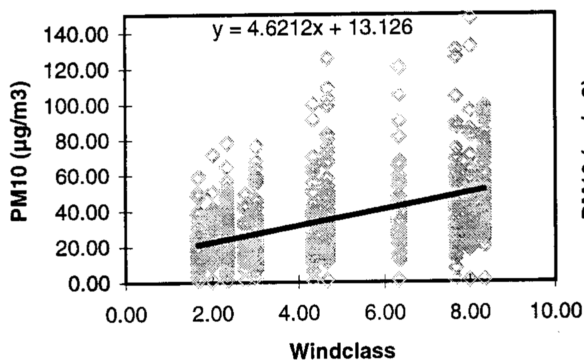

Figure 35: PM10 Background plotted against WindClass.

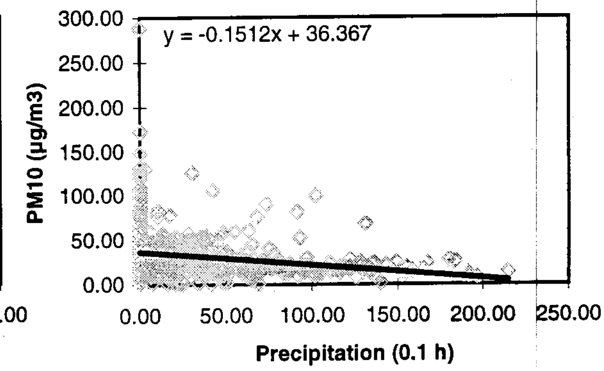

Figure 40: PM10 Background plotted against Duration of Precipitation.

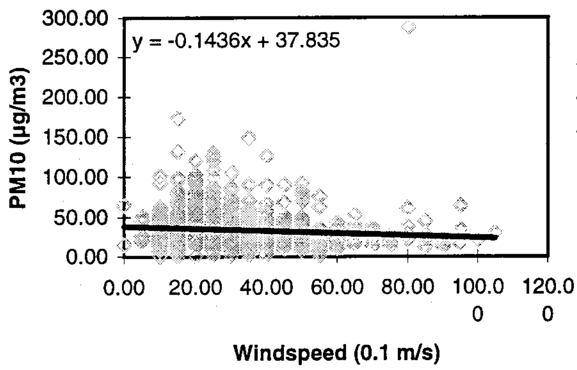

Figure 37: PM10 Background plotted against Wind speed.

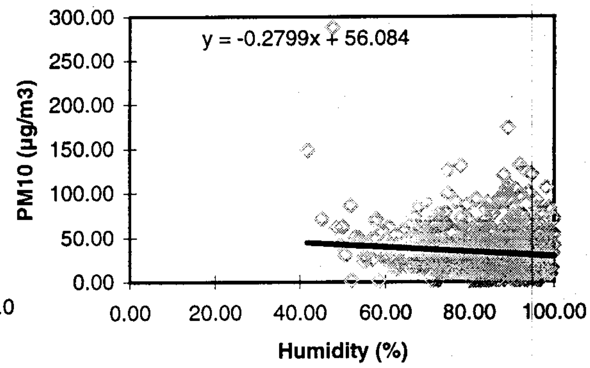

Figure 41: PM10 Background plotted against Relative Humidity.

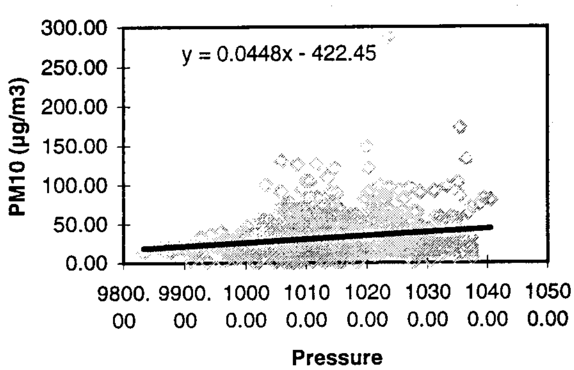

Figure 38: PM10 Background plotted against Atmospheric Pressure.

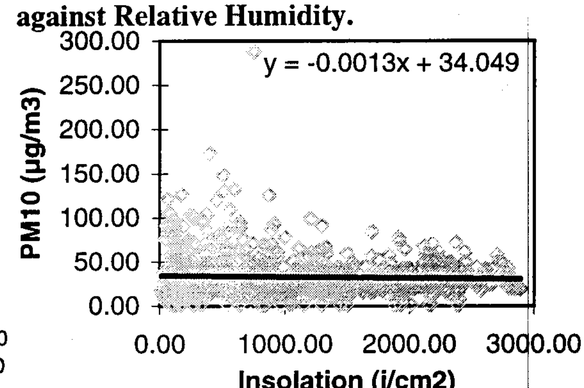

Figure 36: PM10 Background plotted against Insolation.

The influence of the direction of the wind as encoded in the parameter WindClass on levels of PM10 is plotted in Figure 35. Again there is considerable scatter of the observations around the estimated line. A listing of the estimates for calculating levels of PM10 based on WindClass are given in Table 21.

Table 21: Line estimates (y=a.x + b) for the relationship between WindClass and PM10 (µg/m^3)

Station	a	b
Background	4.6	13
Agriculture	4.9	13
Town	5.0	12
Street	4.9	13

With wind speed an inverse relationship with levels of fine particles is found (Figure 37). When the wind speed is high, levels of PM10 are usually lower than at lower wind speeds. In Table 22 the estimated parameters for a straight line fitted to the data are given. Observe that the slope (a) is considerably smaller than for WindClass.

Table 22: Line estimates (y=a.x + b) for the relationship between Wind speed (0.1m/s) and PM10 (µg/m^3)

Station	a	b
Background	-0.1	38
Agriculture	-0.4	60
Town	-0.2	47
Street	-0.3	54

Figure 38 illustrates the relationship between PM10 and Atmospheric Pressure. Scatter in the data is smallest for pressure around 1000 mBar or lower. At higher pressures the majority of the observations is not much higher, but a substantial minority is considerably higher. This minority could be associated with episodes. The parameters for this relationship are listed in Table 23.

Table 23: Line estimates (y=a.x + b) for the relationship between Atmospheric Pressure (0.1 mBar) and PM10 (µg/m^3)

Station	a	b
Background	0.04	-422
Agriculture	0.12	-1203
Town	0.08	-810
Street	0.11	-1106

The amount of precipitation (mostly rainfall) and its influence on levels of PM10 is shown in Figure 39. The fitted straight line indicates that with increasing amounts of rain the levels of fine particles fall. The slope of this line is largely a result of the regular occurrence of high levels combined with no or very little precipitation, which forces the left-hand side upwards. When looking at the plotted data the recorded levels on days with 10 mm or more of precipitation are almost constant at around 25 µg/m^3. The parameters of the fitted lines are listed in Table 24.

Table 24: Line estimates (y=a.x + b) for the relationship between Amount of Precipitation (0.1 mm) and PM10 (μg/m^3)

Station	a	b
Background	-0.1	35
Agriculture	-0.3	52
Town	-0.2	44
Street	-0.2	47

In Figure 40 PM10 is plotted against duration of precipitation. As can be seen when comparing Table 24 with Table 25: Line the slope (a) is equal or somewhat steeper for the relationship with duration than with amount of precipitation.

Table 25: Line estimates (y=a.x + b) for the relationship between Duration of Precipitation (0.1 h) and PM10 (μg/m3)

Station	a	b
Background	-0.2	37
Agriculture	-0.3	55
Town	-0.2	52
Street	-0.3	55

The influence of relative humidity on levels of fine particles is not systematic. The data are very scattered (Figure 41) and the calculated slope is almost zero. The slope is similar for the other stations. Therefore no table listing the parameters for all stations is given. Also for insolation the slope is nearly zero, as can be seen in Figure 36 and no table is given.

3.4.2 Correlations between meteorological parameters/variables

Since independence of variables is desirable in many statistical methods, the correlations between the variables have been studied. This allows for the selection of a subset of the available variables that consist of largely independent variables. An approach like this, where the interrelation between variables are studied, is advocated by Thurston and Kinney (1995).

Table 26 gives the correlations for the meteorological parameters as calculated for the KNMI station De Bilt (see §3.3.4). The largest correlation is that between amount and duration of precipitation. Since the latter is less variable across the country (Tables are included in Appendix C), it is the variable of choice from this pair. Another group of linked variables is Insolation, Relative Humidity, Maximum Mixing Layer Height and Temperature. From this group Temperature is selected.

For the calculation of multiple linear regressions and Kalman-filtering the following basic set of three independent variables has been used: Temperature, WindClass and 'Duration of Precipitation'.

Table 26: Correlation matrices for meteorological variables at KNMI station De Bilt for 1993 and 1994

Correlation matrix Meteo De Bilt 1993	T	WD	WD	RH	RT	RF	P	Q	MH
MaxMixHt / MH	.48	.13	.00	-.27	.05	.16	-.21	.36	1.00
Insolation / Q	.63	-.13	-.22	-.74	-.40	-.27	.06	1.00	
Atmosph. Pressure / P	-.34	-.18	-.24	-.06	-.44	-.39	1.00		
Amnt. of Prec. / RF	.09	.13	.18	.33	.86	1.00			
Durat. of Prec. / RT	-.01	.17	.23	.41	1.00				
Humidity / RH	-.35	.11	-.10	1.00					
WindSpeed / WS	-.04	.07	1.00						
WindDirection / WD	.11	1.00							
Temperature / T	1.00								

Correlation matrix Meteo De Bilt 1994	T	WD	WD	RH	RT	RF	P	Q	MH
MaxMixHt / MH	.36	-.03	-.08	-.18	.00	.06	-.12	.30	1.00
Insolation / Q	.63	-.13	-.29	-.71	-.36	-.22	.11	1.00	
Atmosph. Pressure / P	.00	.06	-.34	-.01	-.30	-.29	1.00		
Amnt. of Prec. / RF	.03	.15	.18	.29	.80	1.00			
Durat. of Prec. / RT	-.09	.18	.25	.39	1.00				
Humidity / RH	-.30	.19	.07	1.00					
WindSpeed / WS	-.25	.00	1.00						
WindDirection / WD	.04	1.00							
Temperature / T	1.00								

3.4.3 Multiple-linear regression

Expanding the linear regression to take into account more than one variable at the same time results in multiple-linear regression. As can be judged from the strong scatter around the fitted lines in the previous paragraph there is considerable room for improvement by using more than one variable. The variables used for multiple-linear regression are:

- Temperature,
- WindClass,
- Duration of Precipitation,
- Wind speed and
- Atmospheric Pressure.

Because of strong correlations with Temperature, meteorological parameters like Relative Humidity and Insolation were not used. Another argument not to use these variables is that they are unreliable predictors for fine particles (cf. §3.4.1). Table 27 lists the results for some combinations of the selected variables for the Background, Agriculture, Town and Street stations. Also tested as a meteorological parameter was Mixing Layer Height. This was found to be a useful addition. The use of Mixing Layer Height, however, did not yield a worthwhile increase in the total amount of variance explained, because it is correlated with Temperature. Almost all of the variance explained by Mixing Layer Height was at the expense of Temperature. The interpretation of the column headed Cycle in Table 27 is the subject of a separate section (§3.4.4).

Table 27: Variances explained per station for different combinations of meteorological parameters in multiple-linear regression for PM10 with average and standard deviation of each parameter

		Variance Explained (%)						
Station	**No. of Var.**	**Temperature**	**Windclass**	**Duration of Pr.**	**Windspeed**	**Atm. Pressure**	**Cycle**	**Total**
Background	3	4.5	20.0	5.8 ~	~	~		26.1
	3	4.5	20.0	5.8 ~	~		0.2	26.1
	4	4.6	20.0	5.5	1.3 ~	~		26.7
	4	4.6	20.0	5.6	1.3 ~		0.3	26.7
	4	4.3	20.1	5.5 ~		1.3 ~		26.3
Agriculture	3	1.0	17.1	11.5 ~	~	~		24.8
	3	1.0	17.0	11.6 ~	~		1.0	24.8
	4	1.1	16.9	10.8	6.0 ~	~		27.8
	4	1.1	16.8	10.8	6.0 ~		1.1	27.7
	4	0.5	17.1	9.3 ~		9.1 ~		27.6
Town	3	9.1	22.2	6.0 ~	~	~		32.0
	3	9.1	22.2	6.0 ~	~		1.0	32.0
	4	9.2	22.1	5.7	1.9 ~	~		33.1
	4	9.2	22.0	5.7	1.9 ~		1.3	33.1
	4	8.7	22.3	5.1 ~		4.0 ~		33.0
Street	3	7.8	18.3	9.5 ~	~	~		30.0
	3	7.8	18.2	9.7 ~	~		1.7	30.0
	4	7.9	18.0	8.8	5.4 ~	~		33.1
	4	7.9	17.9	8.8	5.4 ~		1.8	33.1
	4	7.3	18.4	7.9 ~		7.9 ~		32.1
Average		101.47	4.27	23.25	34.72	10154.10		
S.D.		61.11	2.23	37.22	17.79	96.41		

The results given in Table 27 were generated using the KALFIMAC program. The calculations were made after standardising the variables to have zero mean and unit variance. To use the equations given in the next section measured values of these parameters should be standardised (standardised value=(observed-average)/standard deviation).

From Table 27 it can be seen that the direction of the wind as encoded in WindClass is the most important variable in the regression (17-22% var. expl.). It is also the variable that has the most constant explanatory value. Temperature and Duration of Precipitation are the next two in order of importance (0.5-9% and 5-12% variance explained respectively). Temperature performs best when used for the Town and Street stations; it has very little explanatory value for the Agriculture station. For this station Duration of Precipitation is more important than Temperature. The Street stations is also sensitive to Duration of Precipitation. This variable is less important for both the Background and the Town stations. The other two variables (WindSpeed and Atmospheric Pressure) in some cases explain as much as Temperature and Precipitation of the variance, but the gain in the total amount of variance explained is small. The distribution of the explanatory value for WindSpeed is similar to that of Duration of Precipitation. It is more important for the Agriculture and Street stations than for the Background and Town stations. Other variables not presented here, performed even less well when used in multiple-linear regression. Two explanations for this low performance are possible. One is that there is actually no relation with levels of PM10; the other is that such a relation exists but the same information carried by this new variable is already contained in a (correlated) previous variable.

Considering that models that use the smallest possible number to give a reasonable approximation of reality are preferable, a choice was made to present only results based on the combination of Temperature, WindClass and Duration of Precipitation. As can be seen in Table 27, adding other variables does not substantially improve the total amount of variance explained (ca. 25% for Background to ca. 33% for Street). Note that the percentage given as Total is less than the sum of the percentages for all variables, because the variables are correlated (In cases where variables are uncorrelated the summed amounts of variance explained per variable will be equal to the total amount of variance explained). Given next are multiple linear regression models of type $y=a_0+a_1.x_1+a_2.x_2+a_3.x_3$ for the stations Background, Agriculture, Town and Street (Table 28).

Table 28: Multiple-linear regression results, Constant and Weights for Temperature, WindClass and Duration of Precipitation with standard deviations

Stations	Constant a_0	Temperature a_1	WindClass a_2	Duration of Precipitation a_3
Background	33.7 ± 0.7	-3.7 ± 0.76	9.4 ± 0.8	-4.6 ± 0.8
Agriculture	45.7 ± 0.9	-2.1 ± 0.87	10.0 ± 0.9	-7.8 ± 0.9
Town	39.7 ± 0.7	-6.3 ± 0.74	10.2 ± 0.7	-5.0 ± 0.7
Street	42.2 ± 0.8	-6.3 ± 0.77	9.1 ± 0.8	-6.7 ± 0.8

By calculating the multiple-linear regression model not for the whole study period of two years, but for different subsets of the data the consistency of the values given in Table 28 was studied. In Table 29 an overview is given of the direction and significance of a variables contribution to the model based on data for station Background.

Table 29: Overview of significance and direction of link between variables in multiple-linear regression model and PM10 Background

Period	Constant	Temperature	WindClass	Duration of Precipitation	Atmospheric Pressure
1993/1994	+	+ ↘	+ ↗	+ ↘	-
Summer	+	+ ↗	+ ↗	+ ↘	+ ↘
Winter	+	+ ↘	+ ↗	+ ↘	-
South-East	+	+ ↘	-	-	-
South-West	+	-	+ ↗	+ ↘	+ ↗
North-West	+	+ ↗	-	+ ↘	-
North-East	+	+ ↘	+ ↗	+ ↘	+ ↘
Summer & South-East	+	-	-	+ ↘	-
Summer & South-West	+	+ ↗	+ ↗	+ ↘	-
Summer & North-West	+	+ ↗	-	+ ↘	+ ↘
Summer & North-East	+	-	-	-	+ ↘
Winter & South-East	+	+ ↘	-	-	-
Winter & South-West	+	+ ↘	+ ↗	-	+ ↗
Winter & North-West	+	-	+ ↗	-	+ ↗
Winter & North-East	+	+ ↘	+ ↗	-	-

+ : significant contribution to multiple-linear regression model.
- : insignificant contribution to multiple-linear regression model.
↘: negative link with PM10; ↗: positive link with PM10.

Note that not all variables in Table 29 contribute significantly to the model for all periods. For some variables, their influence is not the same in summer or winter. The best example for this is Temperature. Temperature has a positive regression coefficient when calculated for the summer period (warmer means higher levels of PM10). But it has a negative coefficient for the winter (colder means higher levels of PM10). Clearly, this means that in calculating a regression function for the whole year/period (winter and summer together) the resulting value is a compromise between the two. The winter influence is strongest since the whole-year regression coefficient is negative as well. The power of temperature as an explanatory variable is reduced by this changing behaviour during the year. This finding agrees with the picture given in Figure 34. Instead of using a parabola to describe the behaviour of PM10 with changing temperature, Kalman-filtering with time-variant regressions coefficients can be used (§3.5). WindClass (= wind direction) is always linked positively with PM10. Notice that meteorological parameters are often linked in so-called synoptic weather patterns or 'groβwetterlagen'.

3.4.4 Fluctuations in PM10 on a weekly basis

The existence of a weekly pattern in the levels of fine particles was studied, using the Cycle option of the KALFIMAC program. The program calculates a fixed contribution to occurring levels of PM10 based on the day of the week, which is for a cycle length of 7 time-steps. The results presented here are taken from the multiple linear regression calculations of the previous section. As can be seen in Table 27, the amount of variation explained by a weekly pattern is small (< 2%).

In Figure 42 to Figure 45 the patterns found for a specific contribution to levels of PM10 on a particular day of the week are presented for the stations Background, Agriculture, Town and Street. For Background (Figure 42) no day of the week makes a significant contribution to observed levels of fine particles. This is concluded from the fact that the two standard deviation limits indicated, which form a 95% confidence interval, include zero as a possible value.

For Agriculture (Figure 43) it was observed that on Sundays there is a significant negative contribution, meaning that on Sundays levels are slightly but significantly lower than on other days of the week. The Town and Street stations (Figure 44 and Figure 45) have a clear week pattern. On Fridays levels are elevated relative to the rest of the week. The increase is about 4 $\mu g/m^3$. On Sundays levels are reduced relative to working days by about 6 $\mu g/m^3$. These results agree with the results using more descriptive statistics (§3.2.3), where weekend levels where found to be lower than weekday levels.

Background	Cycle	C+2s	C-2s
Sat	-0.4	3.3	-4.1
Sun	-2.5	1.3	-6.2
Mon	1.0	4.7	-2.8
Tue	-0.2	3.5	-3.9
Wed	0.7	4.5	-3.0
Thu	0.2	3.9	-3.5
Fri	1.2	4.8	-2.5
Sat	-0.4	3.3	-4.1
Sun	-2.5	1.3	-6.2
Mon	1.0	4.7	-2.8
Tue	-0.2	3.5	-3.9
Wed	0.7	4.5	-3.0
Thu	0.2	3.9	-3.5
Fri	1.2	4.8	-2.5
Sat	-0.4	3.3	-4.1

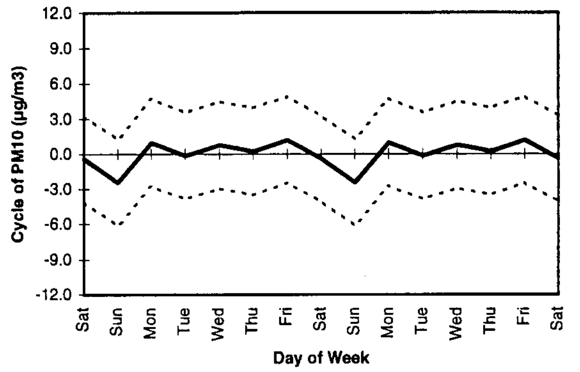

Figure 42: Weekly cycle of PM10 in Background.

Agriculture	Cycle	C+2s	C-2s
Sat	-0.2	4.3	-4.6
Sun	-5.4	-0.9	-9.9
Mon	-1.1	3.4	-5.6
Tue	0.2	4.7	-4.2
Wed	1.9	6.4	-2.6
Thu	1.1	5.6	-3.4
Fri	3.5	7.9	-1.0
Sat	-0.2	4.3	-4.6
Sun	-5.4	-0.9	-9.9
Mon	-1.1	3.4	-5.6
Tue	0.2	4.7	-4.2
Wed	1.9	6.4	-2.6
Thu	1.1	5.6	-3.4
Fri	3.5	7.9	-1.0
Sat	-0.2	4.3	-4.6

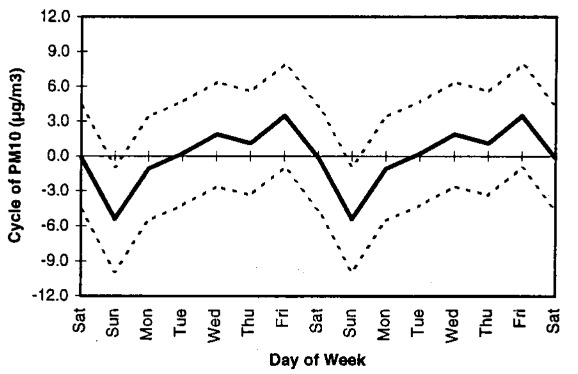

Figure 43: Weekly cycle of PM10 in Agriculture.

Town	Cycle	C+2s	C-2s
Sat	-1.8	1.9	-5.5
Sun	-5.0	-1.3	-8.7
Mon	0.1	3.8	-3.7
Tue	-1.3	2.4	-5.0
Wed	1.5	5.2	-2.2
Thu	2.1	5.8	-1.6
Fri	4.4	8.1	0.7
Sat	-1.8	1.9	-5.5
Sun	-5.0	-1.3	-8.7
Mon	0.1	3.8	-3.7
Tue	-1.3	2.4	-5.0
Wed	1.5	5.2	-2.2
Thu	2.1	5.8	-1.6
Fri	4.4	8.1	0.7
Sat	-1.8	1.9	-5.5

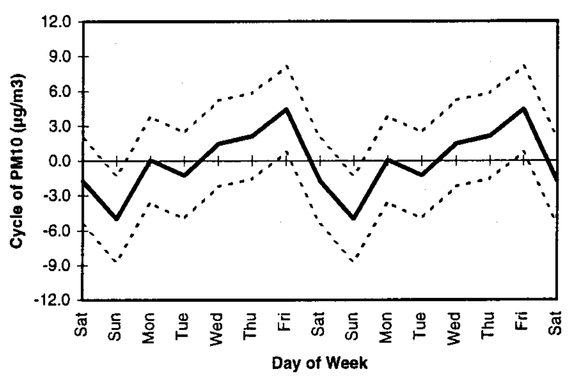

Figure 44: Weekly cycle of PM10 in Town.

Street	Cycle	C+2s	C-2s
Sat	-0.6	3.4	-4.5
Sun	-6.3	-2.4	-10.3
Mon	-0.5	3.4	-4.4
Tue	-0.6	3.4	-4.5
Wed	0.7	4.6	-3.2
Thu	1.8	5.8	-2.1
Fri	5.4	9.3	1.5
Sat	-0.6	3.4	-4.5
Sun	-6.3	-2.4	-10.3
Mon	-0.5	3.4	-4.4
Tue	-0.6	3.4	-4.5
Wed	0.7	4.6	-3.2
Thu	1.8	5.8	-2.1
Fri	5.4	9.3	1.5
Sat	-0.6	3.4	-4.5

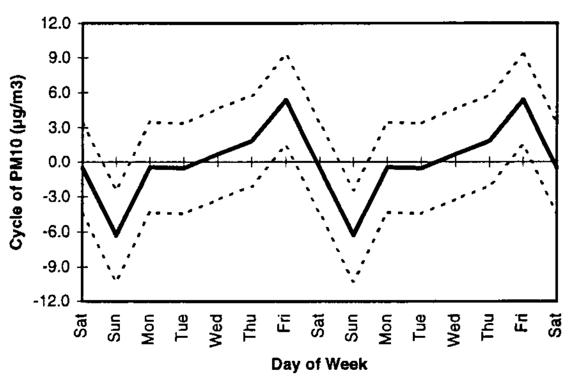

Figure 45: Weekly cycle of PM10 in Street.

3.5 Kalman filtering

As explained earlier Kalman-filtering can be seen as an expansion of multiple-linear regression. This technique has the possibility to let the influence of a variable in the regression model change with time. In this section results will be presented of Kalman-filtering with three variables: Temperature, WindClass and Duration of Precipitation. Included in the model were a trend, a cycle and explanatory variables. By allowing L(t) in Equation (**4**) on page 22 to change with time, a variable trend is estimated. The cycle is the same weekly pattern as presented in § 3.4.4 and present in Equation (**4**) as C(t). The Stochastic Level trend, Temperature and WindClass have been allowed to change slowly during the course of the year. The amount of flexibility allowed has been determined by trying various values for the noise sources and subsequently evaluating the results. The value used for the noise sources is 0.001 for Trend, Temperature and WindClass. For Duration of Precipitation and Cycle no flexibility has been allowed. The resulting amounts of variance explained by introducing time variability to different potential sources is given in Table 30.

Table 30: Variances explained per station for different options for the Kalman-filter with meteorological parameters and PM10 with average and standard deviation of each parameter

Station	No. of Var.	Initial variance	Var. before Trend (%) [1]	Variance explained (%) after Trend [2]				
				Temperature	Windclass	Duration of Pr.	Cycle	Total
Background	3	560.3	100	22	27	5.1	-	44
	3		105	22	28	4.5	-	45
	3		100	22	27	5.2	0.2	44
	3		105	22	28	4.5	0.3	45
Agriculture	3	823.0	100	21	23	10.4	-	43
	3		107	22	23	9.4	-	44
	3		100	21	23	10.5	1.0	43
	3		107	22	23	9.5	1.1	44
Town	3	630.2	100	23	28	5.4	-	45
	3		111	19	29	5.0	-	44
	3		100	23	28	5.5	1.3	45
	3		111	20	29	5.1	1.5	45
Street	3	682.8	100	23	24	8.7	-	44
	3		111	23	25	7.8	-	45
	3		100	23	24	8.7	1.7	44
	3		110	23	25	7.8	1.9	45
Average				101.47	4.27	23.25		
S.D.				61.11	2.23	37.22		

[1]: Variance after trend is 100%. If a Trend is flexible it accounts for part of the total variance and the numbers in this column are >100%.

[2]: Total variance - Variance explained by Trend = Variance after Trend = 100% by definition.

In Table 30 four alternative Kalman-filter results are listed for each station.

- The first alternative allows time-variability for the variables Temperature and WindClass. The Duration of Precipitation is not allowed to vary.
- The second alternative also allows variability for the Trend. The initial variance, which is the variance before trend, is more than 100%. The contributions to the variance explained by the variables refers to the variance after trend as 100 %. The reasoning behind this: the variables are the most interesting part of the model as they account for the day-to-day fluctuations in PM10 levels, while the Trend does not. By reporting this information as is done here, the importance of the variables in the model is conveyed best. The flexibility of the Trend allows the base level for fine particles to change with time. A justification for allowing such behaviour is that emissions change with time and season, in part, independent of meteorology.

- The third alternative has a non-flexible trend in combination with a fixed weekly pattern.
- The fourth alternative is similar to the third, with flexibility added to the trend. A flexible trend accounts for 5 to 10 per cent of initial variance by adjusting the base level of ambient fine particles.

Judging by the explained variances with a fixed trend in relation to a flexible trend, it seems that keeping the trend fixed does not in any substantial way decrease the power of the model. A model with a fixed trend will therefore be used as an example when presenting the results of Kalman-filtering graphically.

Time-variability for the variables Temperature and WindClass increased the power in the model. The gain as measured in percentage variance explained is 5-7 percent for WindClass and no less than around 15 to 20 percent for Temperature. This large gain for Temperature is in agreement with the seasonal dependence of PM10 on Temperature observed earlier. The total amount of variance explained rises to around 45% for all stations. This means that especially the modelling of the stations Background and Agriculture improves by using the Kalman-filter.

The weekly cycle as presented in §3.4.4 based on multiple linear regression calculations accounts for nearly the same amount of variance here (less than 2%). The patterns and contributions per day are essentially the same as those given previously based on multiple-linear regression.

The Port-Manteau and the Runs test indicate that, in general, the residuals do not behave like ‘white noise’ and that the number of runs in the data is less than expected by chance. This points to a systematic change in the residuals, such behaviour is, however, not apparent from a plot of the residuals against time (Figure 48). The NHI test shows that the residuals are not normally distributed or homoscedastic. But they are independent in most cases, according to this same NHI test. Since independence is the most important condition to meet, presentation of the results is justified.

As an example of the results of Kalman-filtering and how they can be interpreted in Figure 46 to Figure 51 the Town station is presented. The figures as they are presented here are, for all practical purposes, the same plots as automatically generated by the KALFIMAC software.

Figure 46 shows the original measurements alongside the calculated trend and the final model. It is clear that the model is able to follow the fluctuations from day to day in levels of PM10 reasonably well. However it misses a part of the measurements that are below or above the normal bandwidth of PM10 (from about 25-55 $\mu g/m^3$), resulting in an underestimation of episodic PM10 levels.

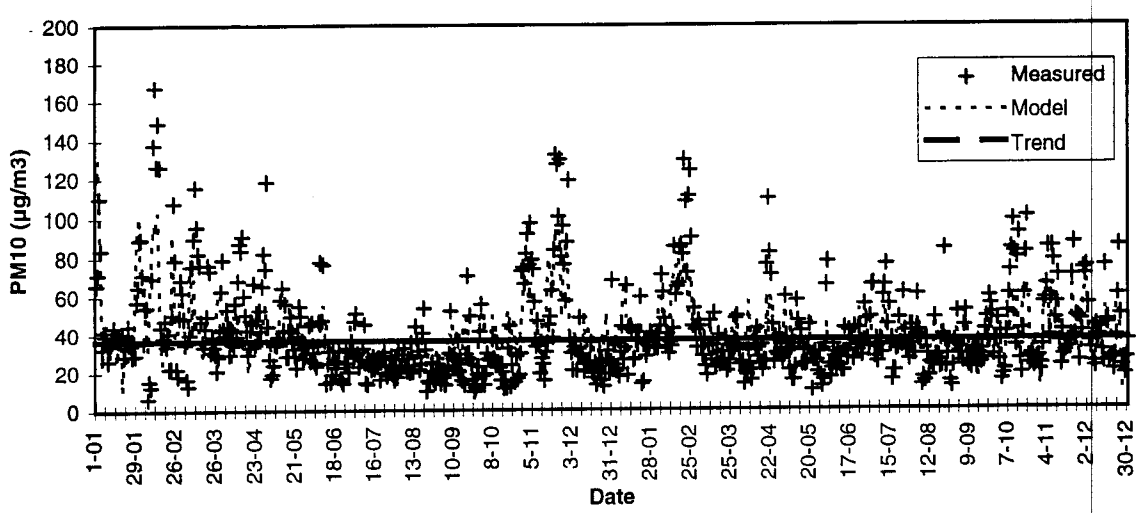

Figure 46: Kalman-filter model plot of PM10 (µg/m^3) for station Town.

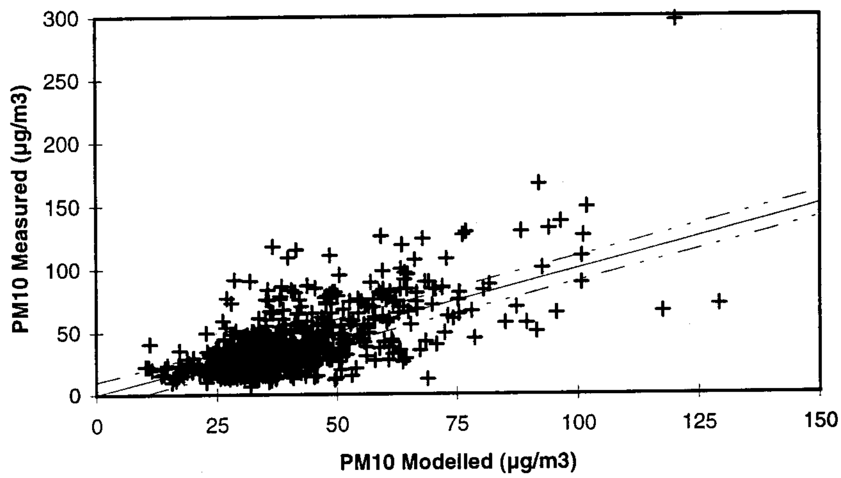

Figure 47: Scattergram of PM10 measured versus modelled by Kalman-filtering for station Town.

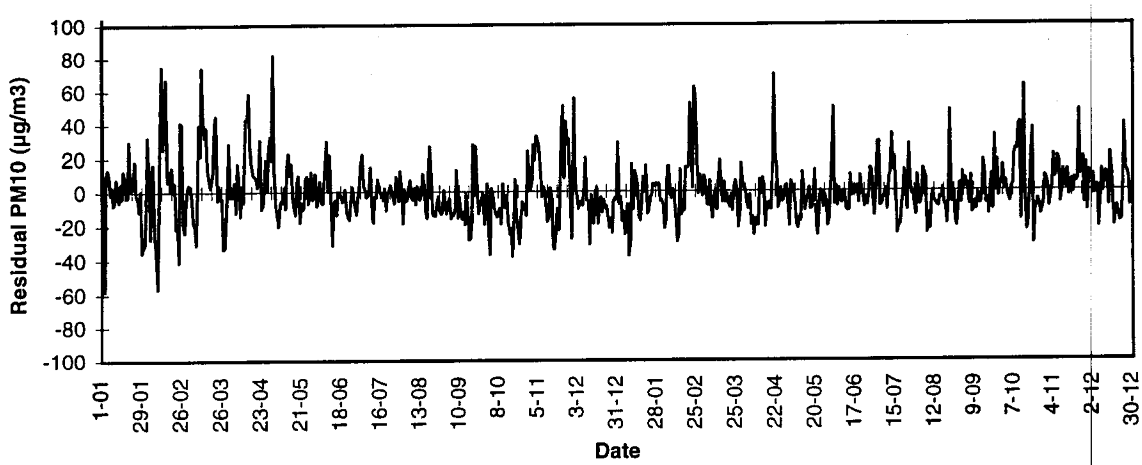

Figure 48: Kalman-filter residual plot of PM10 (µg/m3) for station Town.

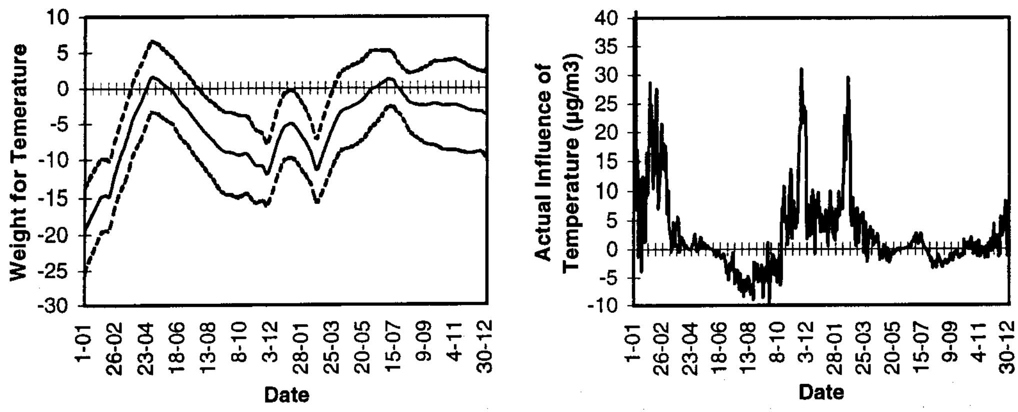

Figure 49: Kalman-filter weight and influence of Temperature on PM10 (µg/m³) for station Town.

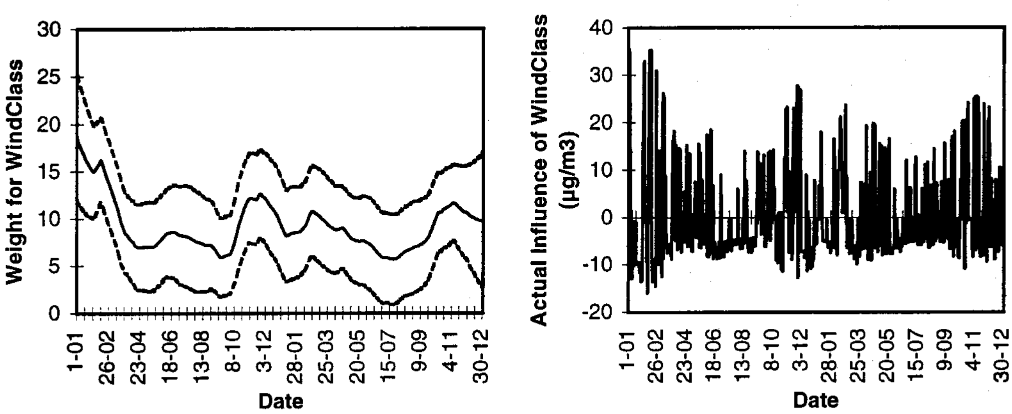

Figure 50: Kalman-filter weight and influence of WindClass on PM10 (µg/m³) for station Town.

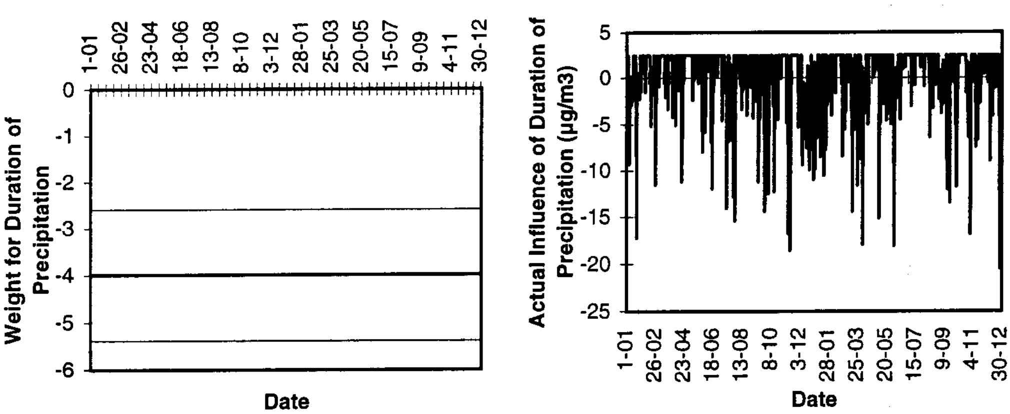

Figure 51: Kalman-filter weight and influence of Duration of Precipitation on PM10 (µg/m³) for station Town.

In Figure 47 the measured data are plotted against the calculated values. This scattergram of measured vs. model is not a standard KALFIMAC graph. From this plot it can be found that for low to normal levels of PM10 the model is accurate to approximately 10 μg/m^3, which is about 25% of the annual average for PM10. More than half of the data points are within this range around the indicated line y=x. In cases of high levels of PM10 the discrepancy between model and observation increases. The errors do not appear to be systematical as both over- and underestimation occurs. Similar information is contained in Figure 48, where the residuals are plotted against time. The majority of the residuals lies within the -10 to +10 μg/m^3 range, but with quite a few larger values as well. Evaluating this graph shows that the residuals do not increase or decrease considerably with time. The presence of a trend in the residuals as suggested by the Runs test is not confirmed by this graph. The reason for this is not known.

The next three figures (Figure 49 to Figure 51) consist of two plots side by side. The left plot shows the weight given to a variable against time, while the right plot shows the actual influence on the model result for that variable. In Figure 49, Temperature is shown. The weighting factors for Temperature change considerably with time. In winter they are large and negative then they go towards zero during spring and turn positive for a while during summer. During the autumn the weights turn to negative values again.

Weight and influence for wind direction i.e. WindClass are given in Figure 50. Here we see that during winter the weight given to WindClass is higher than in summer. As a result of this the amplitude of the actual influence is larger in winter and smaller in summer. Figure 51 shows the constant weight calculated for Duration of Precipitation. The influence for this variable is thus only influenced by different lengths of time when rain or other precipitation falls during the day, ranging from +2 μg/m^3 on days much dryer than average to -10 μg/m^3 on wet days. Even more negative on very wet days with a maximum value of around -20 μg/m^3.

In Table 31 the results of the Kalman-filtering are summarised. For Temperature the highest weights are given during the summer, the weights given are positive during this period. With increasing temperature levels of PM10 rise. The period of the year with such positive weights is much longer and has higher weights for the rural stations: Background and Agriculture. In winter Temperature is given more extreme, but negative, weights than in summer, a decrease in Temperature results in rising PM10 levels. The period with large negative weights is shortest for the rural stations. This period is more than 30 days longer at the Town and Street stations. The differences in for WindClass are smaller. For nearly the same number of days in winter a weight of more than 10 is given and the differences in the maximum weights given are also smaller. This indicates that the variability between the different types of stations is less for WindClass than it is for Temperature. The constant weight of duration of precipitation is similar for Background and Town. For Agriculture and Street it is stronger than for the previously mentioned stations. Please note that the number of days is totalled over the two-year study period.

Table 31: Summary of Kalman-filtering weights, number of days with extreme weights and extremes

Station	Temperature				WindClass		Duration of Precipitation
	W > 0 n (days)	Max. W	W < -10 n (days)	Min. W	W > 10 n (days)	Max. W	W
Background	324	4.5	86	-15.6	232	17.4	-3.6
Agriculture	367	12.0	79	-19.4	240	18.3	-6.3
Town	83	1.6	114	-19.5	238	18.7	-4.0
Street	103	2.6	133	-20.0	227	16.6	-5.5

Contributions of variables to normal and episodic PM10 levels during summer and winter are estimated based on the Kalman-filter results (Table 32) in relation to the annual average. The Wind direction is the variable that has the strongest influence on PM10 levels. Its contribution to episodic levels is rivalled by Temperature. The contribution of Duration of Precipitation to episodic levels is estimated to be small, since the Wind direction and Temperatures associated with episodes usually do not coincide with precipitation.

Table 32: Contributions of meteorological variables to normal and episodic PM10 levels (μg/m³) for winter and summer, based on Kalman-filtering

	Winter Average	Winter Episode	Summer Average	Summer Episode
Wind direction	-15 / +15	+30	-10 / +5	+10
Temperature	+0 / +10	+30	- 5 / +5	+10
Precipitation	- 5 / -10	+2	- 5 / -10	+2

4. Discussion and recommendations

In this section the findings are discussed and in many cases they lead to recommendations or suggestions for further research. First, topics relating to the measurement of PM10 and other air pollutants are discussed. Secondly, associations of meteorological variables and PM10 are dealt with. Thirdly, some observations based on analyses performed that may require further investigation are treated. Finally, recommendations are made that do not fit the previous categories.

4.1 Measurements

On the basis of the hourly values, the summer of 1994 shows a very distinct pattern of PM10 during the day, with high levels during the night and lower levels during the day. In the summer of 1993, this pattern was much less pronounced. It has been suggested that the observed pattern could be caused by an interaction of PM10 measurement with relative humidity. To investigate the suggested link with relative humidity the correlation between PM10 and relative humidity for July 1994 was calculated (Appendix F); the graph shown Figure 52 combines the courses of PM10 and relative humidity. The correlation between PM10 and relative humidity is not very strong with values for most stations in the range from about -0.10 to about 0.10. For most stations the association between PM10 and time of day is even weaker (Appendix F). The strongest correlation calculated is for relative humidity and time of day is -0.63. This in combination with the fitted splines in the graph, which show that the decline of PM10 starts before that of relative humidity, suggests that changes in relative humidity are not the cause of the observed pattern in the summer of 1994. However, Zhang *et al.* (1993) report a relation between water content of submicron aerosols, which is obviously related to weight, and relative humidity, when measuring at both ambient relative humidity and subsequently at several experimentally controlled relative humidities. In Pastuszka and Okada (1995) it is suggested that differences in hygroscopic properties of particles reflect differences in sources an ageing processes. Further investigation of relation between PM10 and relative humidity is recommended.

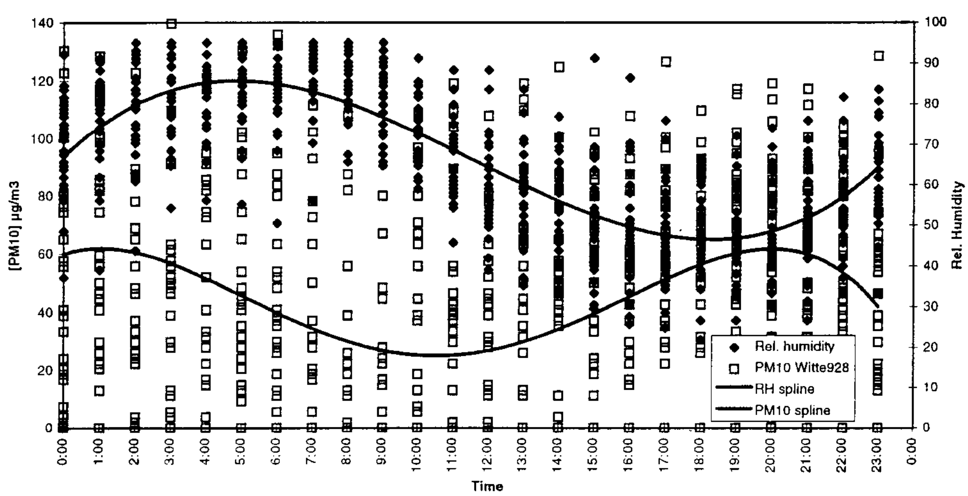

Figure 52: Courses of PM10 and relative humidity during the day in July 1994 on LML station Witteveen 928.

The chosen technique for measuring PM10 in the LML is a β-radiation monitor. A radiation source is used to measure the amount of dust collected on a filter tape. Because of the intrinsic randomness of radioactive decay, hourly measurements show a scatter of 10 $\mu g/m^3$ (van Elzakker *et al.*, 1992). As a result, single hourly measurement values are only of limited use. In this report the results presented are therefore based on many hourly values. For a detailed analysis of PM10 during a single episode, e.g. on the influence of wind speed, this variability poses a problem. A standardised way of handling this variability has to be decided upon.

The influence of wind speed on PM10 levels can best be studied on the basis of hourly values for episodes with high wind speeds. For low wind speeds, e.g. lower than 6 m/s where a strong scattering has been found using daily values, the relation, e.g. with wind direction, can be studied further using daily values as well. Another cause for high levels in combination with low wind speeds can be stagnation, which is likely to combine with a lower than usual height of the mixing layer.

It is suggested that possible differences in sampling efficiency between the PM10 measurements (β-radiation monitor) and measurements of secondary aerosol components (LVS filters) should be studied. This suggestion is in line with a more detailed study of the ratio of primary to secondary aerosol in PM10, suggested elsewhere in this report.

It is puzzling that for winter 1993, levels have been recorded at two street stations and two town stations that are well below all other stations, including rural ones. This is not seen in other parts of the same year nor is this noticeable for any period in 1994. Possible technical problems, e.g. a clogged filter, should be investigated further.

4.2 Associations

A possible artefact is the association between PM10 levels and Duration of Precipitation. This could be caused by the fact that precipitation, mainly rainfall, usually coincides with south-westerly winds, which is a relatively clean wind direction (Figure 53). When calculating correlations between PM10 and Duration of Precipitation, values are found ranging from -0.23 to -0.43 for the stations Background, Agriculture, Town and Street. These are weaker than the correlation between Duration of Precipitation and WindClass (-0.55). Stronger evidence is that a negative correlation is found even when the wind direction is confined to the sector around south-west (Table 33). But also for wind directions restricted to south-east, a negative correlation is found. This indicates that processes like prevention of resuspension or wash-out of particles have a real influence, albeit only small, on fine particle levels.

Table 33: Correlation coefficients between PM10, Wind Direction and Duration of Precipitation for south-westerly and south-easterly winds, including number of days, as calculated for the station Street

Correlation between Duration of Precipitation and	PM10 (µg/m3)	WindClass	No. of days
South-westerly wind	-0.3	0.05	231
South-easterly wind	-0.2	0.10	96

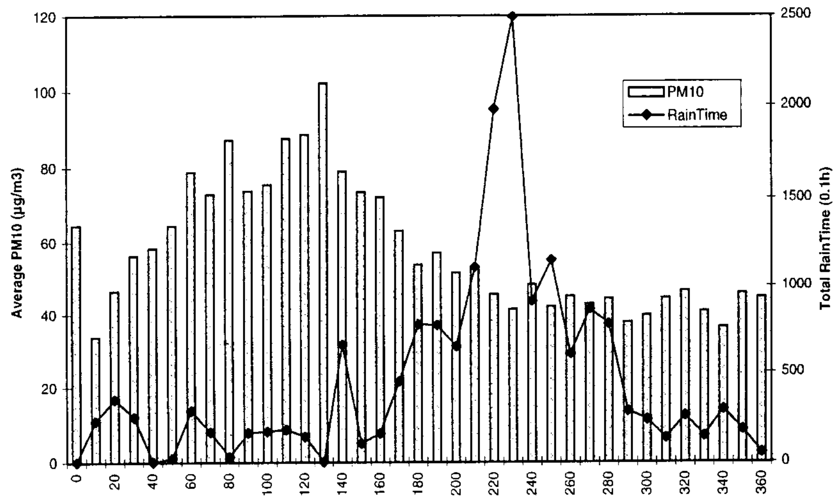

Figure 53: Average PM10 levels and total duration of precipitation per wind direction for 1993/1994 at the station Street.

For some wind directions the fine particle levels are more variable than for other directions. This is, however, not directly evident from the windroses. The most sensitive wind directions for this variability are south and north-east. What happens is that these are on the border of the wind sector that contributes most to high PM10 levels, the south-east. When winds blow from the south or the north-east, the air can come straight from clean areas like the North Sea and the Atlantic Ocean. But the same winds can also bring continental air to the Netherlands, at which time high levels of PM10 can occur, especially in winter. This means that large-scale weather patterns ('groβwetterlagen' or synoptic weather systems) determine PM10 levels.

Wind speed is a meteorological parameter that is of minor importance for PM10 under normal circumstances, as can be seen from the windroses for the LML stations with PM10 levels subdivided by wind speed. Figure 37, however, does suggest that some relation may exist. A difficulty is that at wind speeds below about 8 m/s the range of PM10 levels increases (with westerly winds likely to be responsible for lower levels of PM10 and easterly winds for higher levels; stagnation episodes can further confuse the picture and also mixing layer height may be involved). What the actual behaviour at very high wind speeds will be, cannot be made out from the present data set. Daily values with wind speed over 10 m/s are too scarce. In Bloemen et al. (1995) a distinct increase of fine particles is shown for wind speeds higher than 12 m/s based on hourly values of the episode in February 1994.

From the correlations found between the LML stations in 1993 and in 1994 for PM10, it is observed that high correlations are found between town and street stations. This can be a side-effect. Many of these are geographically close together in the western parts of the country and are also influenced by local sources. The exploratory cross-correlation analysis, as suggested by Thurston and Kinney (1995) to study variable interrelationships, should also be used to reveal similar relations between sampling sites.

For the stations Background, Agriculture, Town and Street, data from two stations have been lumped together. One reason for this was to reduce the influence of very localised sources on the PM10 behaviour. Another reason was to reduce the number of gaps in the time-series. The Background station became a combination of LML stations in the north while its closest relative, Agriculture, is made up from LML stations in the south. This means that when comparing these stations, the large-scale gradient across the country with lower levels in the north should be taken into account. This large-scale gradient is a result of the geographical distribution of sources and of differences in travel distance as a function of aerodynamic diameter (Table 34).

Table 34: Estimated travel distance for dust particles based on aerodynamic diameter and deposition velocity (van Jaarsveld, 1995) assuming a mixing layer height of 1000 m and an average wind speed of 3.5 m/s based on meteorological data from KNMI station De Bilt.

Aerodynamic diameter (μm)	Deposition velocity (cm/s)	Travel dist. (km)
10	5	60
5	4	75
2.5	0.5	300
1	0.01	15 000

The way in which daily values for the wind direction and wind speed were calculated from hourly values in this study, by vectorial addition, is standard practice in meteorology. In air pollution research the two are usually averaged separately. To study whether this makes a difference, a comparison can be made between a windrose based on hourly values and one based on daily values. For this comparison, the LML station Utrecht-Erzeijstraat 639 is suggested as the best choice. This is based on its proximity to the KNMI station, De Bilt 260, selected for the meteorological data for the stations Background, Agriculture, Town and Street.

4.3 Analyses

In the summer of 1994 higher levels of PM10 where recorded than in the summer of 1993. In the windroses it can be seen that this increase had the strongest influence on north-westerly winds. An increased sea salt contribution, as the cause for this observation, could be the subject of further investigation.

Levels recorded at the site, Breukelen-snelweg, are lower than those at other street stations. This effect was found in the winter of 1994 but not in the summer nor the autumn and should be investigated further. The difference between Breukelen-snelweg, a motorway in rural surroundings, and other street stations could be the absence of urban sources at that location and different traffic patterns.

The analysis of all episodes using Kalman-filtering cannot fully explain the very high concentrations during the episode in February 1994. Further analysis of the ratio between primary and secondary aerosol during this episode showed that during the first 2 or 3 days, PM10 concentrations for the larger part (>90%) were determined by primary aerosol caused by windblown dust from the very high wind speed during these few days. Normally, around 60% of PM10 is primary aerosol, both under normal circumstances and during episodes. An analysis of the range of primary to secondary aerosol within PM10, similar to Figure 32, as a function of meteorological parameters, is recommended.

Using regression techniques in conjunction with a variable of an inherently circular nature like wind direction presents a problem. As most regression techniques are not equipped to deal with this kind of behaviour in a variable, some way has to be found that linearises the variable. As already pointed out in §2.1, several ways of achieving this have been tested. Many worked to some extent, but finally a decision was made to use the variable WindClass. However, this variable uses knowledge of PM10 levels that have occurred historically and as such is not independent of the PM10 levels it predicts. One more objection is that no clear relationship exists between the WindClass and the original wind direction. It is suggested that, in future, the wind direction be coded using two variables. One variable stands for the wind in the direction north to south (1 to -1) and the second for the direction east to west (-1 to 1). Advantages include the small number of variables needed while allowing good spatial resolution and, in contrast to WindClass, there is a clear and direct relationship with the original wind direction without the need for historical knowledge of fine particle levels. An approach similar to this is used in Somerville *et al.* (1996) when trying to estimate the wind direction of maximum air pollutant concentration.

The changing influence of temperature on PM10 during the year warrants a derived variable: absolute difference between actual temperature and annual average. This complies with an observed association between health effects and absolute difference between actual temperature and annual average (Schwartz, 1993).

4.4 Recommendations

A detailed look was taken at a possible relation between above normal levels of PM10 and the height of the mixing layer. Both a selection of summer days (n=65) and a selection of winter days (n=26) showed no evidence of a clear relationship with mixing-layer height. A more detailed study of this subject is suggested.

An observation on the make-up of Town and Street is concerned with shopping behaviour and laws. In the Netherlands shops are allowed to stay open until 21:00 one evening in the week. On this evening -either a Thursday or Friday- traffic intensity in town centres is likely to be higher than usual. By combining stations with different shopping evenings athe weekly pattern may be flattened. Such a combination has occurred for the station Street (Thursday in Utrecht and Friday in Eindhoven). A more detailed study of this subject can be performed.

PM10 concentration differences are small(<20% across the Netherlands at a given moment) but the composition of PM10 is more varied due to local and regional emissions. Burton *et al.* (1996) report that around Philadelphia, PM2.5 dominates the measured PM10 levels and that this results in a diminished relation with local sources for PM10. For the fraction of PM10 >2.5μm, they report that concentrations vary spatially within Philadelphia. More detailed studies of the chemical composition and the size distribution of PM10, such as the CHEAP campaign, are called for. The need for detailed information on chemical composition and size distribution of sampled aerosol is also recognised in Cass (1995) and Phalen and McLellan (1995). Of interest in this respect are, for instance, polycyclic aromatic hydrocarbons and other hydrocarbons, especially volatile species. The monitoring efforts like these were already made during the CHEAP campaign - Characterization of Episodic Air Pollution in cities. Allen *et al.* (1996) found that about half of the PAHs with molecular weights up to 200 are associated with PM2.5 and that this proportion increases for larger PAHs. In the light of these findings the association of PAHs, partly volatile species, with PM2.5, can help to find a toxicological mechanism for the effects of fine particle air pollution.

Another chemical component of PM10 that may be important on a European scale is H^+ or 'strong acidity'. Strong acidity is considered in several publications (Thurston and Kinney, 1995; Lippmann and Ito, 1995) as a possible causal agent for the association of PM10 with adverse health effects.

It is recommended that organic/biogenic material be taken into account in the analyses of PM10 measurements because of the allergenic properties of such materials as pollen and fungal spores towards humans. Matthias-Maser and Jaenicke (1995) have found that, on average, 15% of the volume concentration of airborne particles is of a biological origin on a sampling site with both urban and rural influences. Especially urban and related sources were found to be related to an increased proportion of aerosol from a bacterial origin. Also Phalen and McLellan (1995) recommend the inclusion of biological particle categories in future research.

The addition to the LML of two PM10 sampling sites is suggested. By adding LML Zegveld 633 to the PM10 measurement stations, the influence of urban areas on regional and local concentrations can be studied better. Addition of LML Kollumerwaard 934 may help to clarify the importance of the south-north gradient across the county.

The Kalman-filter is a linear model, which assumes a normal distribution of the data. When, during episodes, processes combine in a non-linear way, the model is not suited to reproducing such behaviour. The model will also have difficulties in reproducing both episodic high PM10 levels (>100 $\mu g/m^3$) and low PM10 levels (<15 $\mu g/m^3$) from the tails of the distribution. Using the Kalman-filter for the study of PM10 levels in association with meteorological parameters is legitimate, keeping these limitations in mind. Much of the normally occurring daily fluctuations in PM10 levels can be explained using this technique. For the study of episodes with unusual concentrations, both high and low, a choice has to be made. One option is to use a different modelling technique; another option is to model a selection of the data to construct a model of episodic levels.

It may be of importance with high concentrations to distinguish between summer and winter episodes. During winter episodes, transport and limited dispersion may play a role. During summer episodes, formation of secondary aerosol can be important. Especially for summer episodes, it is recommended that the ratio between primary and secondary aerosol (mostly sulphates and nitrates) is investigated further. This may help us to understand the importance of the formation of secondary aerosol during summer episodes.

5. Conclusions

- At any moment in time, differences in PM10 between stations are small (<20%), indicating that PM10 levels in the Netherlands are influenced by large meteorological processes.
- The PM10 background concentration in the Netherlands, which occurs when the wind is blowing from the north-west, is about 10-15 μg/m^3.
- Long Range Transport from foreign source areas in the east and south may increase average concentrations in the Netherlands to about 40-50 μg/m^3.
- The results show a large-scale concentration pattern with a south-north and east-west gradient in the order of 10% of the yearly average (about 5 μg/m^3).
- A spatial analysis of meteorological variables showed a strong correlation between the KNMI stations. Stations furthest apart showed the lowest correlations. It is concluded that the use of a single meteorological station for the PM10 stations is sufficient when studying the general relationship between meteorology and fine particle levels. To minimise the influence of distance, the KNMI station De Bilt 260, located in the centre of the country, is recommended for this use.
- The broad band of daily variations in the concentration over the year, with not much difference between summer and winter can be largely explained by daily varying meteorological conditions i.e. wind direction, temperature and precipitation, as modelled by Kalman-filtering. Episodic PM10 concentrations (<60 μg/m^3) are not modelled well by Kalman-filtering using these parameters. This could be caused by abnormal meteorological circumstances that apply in these particular situations e.g. extreme low mixing-layer height, stagnation, strong easterly winds.
- Episodes occur when eastern winds concur with low temperatures (<0 °C)during winter. Besides increased emissions, non-linear processes correlated with temperature substantially increase the influence of the meteorological variables temperature and wind direction, as demonstrated by Kalman-filtering. This leads to PM10 concentrations that can be 4 to 5 times the yearly average.
- Temperature (which concurs with "no rain") may have a limited influence on PM10 concentrations during the summer months
- Based on this study an indicative table (Table 35) of relative differences in concentrations measured at different sites in the Netherlands was composed. The differences are the result of local and regional processes, as well as large-scale (meteorological) processes.
- Results from this study show that local PM10 emissions contribute markedly to local and regional PM10 concentrations.

Table 35: Relative contribution [1] of different sources to PM10 on different sites in the Netherlands

Source/Site	Background Coastal	Background Inland	Agricultural	Urban
Sea salt [2]	~10%	<5%		
Traffic/Urban [3]				~10%
Agricultural [4]			~10%	
Large-scale gradient (N-S) [5]	~10%	~10%	~10%	~10%

[1] ~10% equals 5-6 μg/m^3. [2] based on Table 15. [3] based on §3.2.3. [4] difference between Rural-high and Rural-average in Table 5 to Table 7. [5] difference between Rural-average and Rural-low in Table 5 to Table 7

Acknowledgements

We would like to thank the following people for their contributions: Frans van Alkemade for his help with the principal component analyses; Hans Visser for his good advice on the use of the Kalman-filter and the interpretation of the results; Henk Bloemen, Erik Noordijk, Hans Eerens and Hans van Jaarsveld for supplying ideas and suggestions during the research and the writing of the report; Ronald Smetsers and Henk Reinen for kindly supplying us with their data on ^{210}Pb.

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Appendices

Appendix A: Archived files and their contents.

Of all the files generated during this study the ones that are listed here have been archived (electronically) for the purpose of future reference. The files listed here are such that most of the steps performed during the study can be traced.

File name	Description of contents	Corrected for sampling efficiency
PM1093.rep	Hourly PM10 data as extracted from RIL+, 1993, ASCII	No
PM1093.slv	Hourly PM10 data for 31-12-1993 as extracted from RIL+, ASCII	No
PM1094.rep	Hourly PM10 data as extracted from RIL+, 1994, ASCII	No
PM1094.slv	Hourly PM10 data for 31-12-1994 as extracted from RIL+, ASCII	No
PM10HOUR.xls	Hourly PM10 data for 1993, plots per month	No
CELCHOUR.xls	Hourly Meteorological data for 1993 and 1994, Temperature	#
HUMIHOUR.xls	idem, Relative humidity	#
PASCHOUR.xls	idem, Atmospheric pressure	#
RAINHOUR.xls	idem, Amount of Precipitation	#
TIMEHOUR.xls	idem, Duration of Precipitation	#
SUNSHOUR.xls	idem, Insolation	#
WIDIHOUR.xls	idem, Wind direction	#
WIVEHOUR.xls	idem, Wind speed	#
MIXHT.dat	Hourly data file Mixing Layer Height (LIDAR)	#
WIND93.xls	Calculation daily average wind direction / speed 1993 Calculation for 1994 similarly in SPSS	#
PM10SPSS.xls	Calculation daily average PM10 for 1993 and 1994	No
NOV93.xls	Hourly value graphs for November 1993 (used in report)	Yes
PM10H24H.xls	24h course of PM10 during the day	No
PMRHHOUR.xls	Relation PM10 & rel. humidity summer / July 1994	Yes
REPPMRH.xls	Graph and Correlation matrix from PMRHHOUR for report	Yes
24HPATT.xls	24h pattern / courses during the day (used in report)	Yes
24HTABLS.xls	24h pattern / courses during the day tables with average PM10 concentrations for the period, used in report	Yes
PM10D.xls	Day values PM10 1993+1994	No
CELCD.xls	Day values Meteorological data 1993+1994, Temperature	#
HUMID.xls	idem, Relative humidity	#
PASCD.xls	idem, Atmospheric pressure	#
RAIND.xls	idem, Amount of Precipitation	#
TIMED.xls	idem, Duration of Precipitation	#
SUNSD.xls	idem, Insolation	#
WIND93D.xls	idem, Wind direction / speed 1993	#
WIND94D.xls	idem, Wind direction / speed 1994, N.B. direction miscoded	#
WIND94DS.xls	idem, as WIND94D but Wind direction corrected	#
WINDRO93.xls	Windroses 1993 (year)	Yes
WINDR293.xls	Windroses 1993 summer, winter	Yes
WINDRO94.xls	Windroses 1994 (year)	Yes
WINDRZ94.xls	Windroses 1994 summer	Yes
WINDRW94.xls	Windroses 1994 winter	Yes
WINDTAB.xls	Table Wind coding options for report	Yes
REPWINDR.xls	Windroses for report	Yes
WINDHODA.xls	Preliminary comparison of windrose based on hourly resp. day values for Utrec639	Yes
PM10MEAN.xls	Table Yearly / Summer / Winter average PM10 concentration (used in report)	Yes
PM10-93D.xls	Stations Background, Agriculture, Town, Street from LML stations, Correlation matrix LML stations, for 1993	Yes
PM10-94D.xls	as above for 1994	Yes

File name	Description of contents	Corrected for sampling efficiency
BATSTABL.xls	Table descriptive statistics for stations B,A,T,S (used in report)	Yes
PM10REGR.xls	Regression lines and windroses for B,A,T,S	Yes
PM10REGB.xls	idem continued for additional regression lines	Yes
METEOCOR.xls	Correlation matrices for KNMI stations and Meteo parameters except wind directon and speed, 1993 and 1994	#
WINDSCOR.xls	Correlation matrices for wind direction and speed, 19943and 1994	#
REPCORRL.xls	Correlation matrices KNMI, Meteo, LML, 1993 and 1994 (used in report)	#
KALFI04.xls	Spreadsheet preparing KALFIMAC .dat input files	
PM9394F.dat	Kalman datafile 1993+1994, Background	Yes
PM9394C.dat	Kalman datafile 1993+1994, Agriculture	Yes
PM9394D.dat	Kalman datafile 1993+1994, Town	Yes
PM9394E.dat	Kalman datafile 1993+1994, Street	Yes
PM9394X1.opt	Kalman optionfile, mult.lin.reg., 3 exp. var. other option files number 2-5 are also multiple linear regressions but with other variables and/or a cycle	#
PM9394XA.opt	Kalman optionfile, filtering, 3 exp. var. other option files coded B-F are also filtering but with other variables and/or a a cycle and/or flexibility options.	#
pm9394xy.out	Kalman output files; output in three main tables, also contains data on %Variance explained per model component and results of statistical tests. x= C,D,E or F for Background, Agriculture or Street y= 0..1; A..F for corresponding option file	#
pm9394XY.all	Kalman output files; output in tabular format, suitable for creating graphs of the results. x and y coded as above.	#
KALFI05.xls	Linear regressions PM10 on Meteorological parameters (used in report)	Yes
KALFI05L.xls	idem	Yes
KALFI06.xls	idem	Yes
OVERVWF.doc	Document summarizing KALFIMAC.out results for Background	#
OVERVWC.doc	idem for Agriculture	#
OVERVWD.doc	idem Town	#
OVERVWE.doc	idem for Street	#
OVERVW.xls	Overview of KALFIMAC Multiple-linear regression results	#
PM9394DA.xls	Kalman-filtering output (.all) for Town, graphs used for report, also contains output data for PM9394XA.opt for the other three stations.	Yes
PM9394E2.xls	Kalfimac m.l.r. output (.all) for Street	Yes
PM9394FD.xls	Kalman-filtering output (.all) for Background	Yes
24HPA94.xls	Exploration of difference in weekly cycle between Rural-average, Rural-low, Rural-high, Town and Street	Yes
PM93DFIT.xls	Goodness of Fit testing PM10 day values for 1993	No
METEOFIT.xls	Goodness of Fit testing meteorological parameters KNMI De Bilt	#
REPCYCLE.xls	Weekly cycle data for B,A,T,S and graphs, used in report	Yes
REPMETAB.xls	Table descriptive statistics meteorological parameters KNMI De Bilt, used in report	#
NH3AGRIC.xls	Table with data on NH_3 in 1993 and 1994, used in report	Yes
HOUTAK.xls	idem for Houtakker, 1/1/93 - 28/2/95, not used	Yes
APELSPR.xls	idem for A'doorn-Stationsstraat, 1/2/93 - 31/1/95, used for Benzene correlation	Yes
DORDSPR.xls	idem for Dordrecht, 1/2/93 - 31/1/95, used for Benzene correlation	Yes
EINDSPR.xls	idem for Eindhoven-Genoveveln., 1/2/93 - 21/1/95, used for Benzene correlation	Yes
ROTSPR.xls	idem for Rotterdam-centrum, 1/2/93 - 31/1/95, used for Benzene	Yes

File name	Description of contents	Corrected for sampling efficiency
	correlation	
UTRSPR.xls	idem for Utrecht-Erzeijstr., 1/2/93 - 31/1/95, used for Benzene correlation	Yes
VREDESPR.xls	idem for Vredepeel, 1/2/93 - 31/1/95, used for ratio primary/secondary aerosol	Yes
WIERSPR.xls	idem for Wieringerwerf, 1/2/93 - 31/1/95, used for ratio primary/secondary aerosol	Yes
WITSPR.xls	idem for Witteveen, 1/2/93 - 31/1/95, used for ratio primary/secondary aerosol	Yes
ZILKSPR.xls	idem for De Zilk, 27/4/93 - 31/1/95, not used	Yes
SECAN.xls	Ratio primary to secondary aerosol in PM10	Yes
SECANT1.xls	idem final graph in black and white	Yes
jtwstati.xls	Data on stations e.g. name, no., location (KNMI & LML), used for calculating LML to KNMI twinning, used in report	#
CODATA	PCA datafile carbon monoxide	#
NO3DATA	PCA datafile nitrate	#
PM10DATA	PCA datafile PM10	No
RCO.pro	IDL Program file for CO PCA	#
RLEON.pro	IDL Program file for CO PCA	#
RLEON2.pro	IDL program file for CO PCA	#
RNO3.pro	IDL program file for NO3 PCA	#
RPM10.pro	IDL program file for PM10 PCA	#
RPM10INT.pro	IDL program file for PM10 PCA	#

= not relevant for these files.

Appendix B: Structure of the main datafiles/database.

APPNDXB.XLS PM10

	A	B	C	D	E	F	
1							
2	Component:	Witte928	Wieri538	Amste520	Breuk641	Utrec639	
3		928	538	520	641		639
4	Project	lml	lml	lml	lml	lml	
5	Unit	ug/m3	ug/m3	ug/m3	ug/m3	ug/m3	
6	Method	Witteveen r	Wieringerwerf r	Amsterdam-Noord t	Breukelen-snelweg s	Utrecht-Erzeijstraat s	
7	Hour\Station	Witte928	Wieri538	Amste520	Breuk641	Utrec639	
8	1/1/1993 1:00						
9	1/1/1993 2:00	105	110				
10	1/1/1993 3:00	125	103				
11	1/1/1993 4:00	180	132				402
12	1/1/1993 5:00	179	113				332
13	1/1/1993 6:00	192	130				406
14	1/1/1993 7:00	196	151				378
15	1/1/1993 8:00	155	162				288
16	1/1/1993 9:00	114	117				307
17	1/1/1993 10:00	101	102				333
18	1/1/1993 11:00	108	99				254
19	1/1/1993 12:00	116	113				
20	1/1/1993 13:00	116	120				
21	1/1/1993 14:00	116	152				125
22	1/1/1993 15:00	107	183				120
23	1/1/1993 16:00	85	155				132
24	1/1/1993 17:00	92	160				141
25	1/1/1993 18:00	80	199				155
26	1/1/1993 19:00	109	229				134
27	1/1/1993 20:00	99	227				117
28	1/1/1993 21:00	94	176				86
29	1/1/1993 22:00	83	137				69
30	1/1/1993 23:00	76	117				47
31	2/1/1993 0:00	52	98				41
32	2/1/1993 1:00						
33	2/1/1993 2:00	67	106				
34	2/1/1993 3:00	60	98				
35	2/1/1993 4:00	53	66				
36	2/1/1993 5:00	49	54				34
37	2/1/1993 6:00	56	39				45
38	2/1/1993 7:00	51	48				38
39	2/1/1993 8:00	49	38				49
40	2/1/1993 9:00	57	39				48
41							
42	File continues to the right with the other LML PM10 stations						
43	File continues down with the hourly values for the rest of the year						
44	N.B.one year of hourly values equals 8760 rows of data						
45	As a consequence only one year can be stored in a table like this on a Excel workbook tab						
46	Max . number of rows is 16384						

Appendix C: Examples of Kalfimac option files

Example of a KALFIMAC multiple linear regression option file: pm9394x1.opt.

```
1   PM10&Meteo 1993/94 T,WC15,RT fix stndrdzd
2   3 0 3
3   0.0 0.0 0.0 0.0
4   10
5   0
6   0
7   1
8   0 1
9   0 0
10  1 0.99
11  0
12  1 1 0 10 1
13  Concentration PM10 (µg/m3)
14  -4 10 7 2 8.0 1
15  Date
16  0 8 8 2 12.0 1
17  1.0
18  Temperature (0.1 oC)
19  WindClass
20  RainTime (0.1h)
21  0 8 8 2 12.0 1
22  0 8 8 2 12.0 1
23  730
24  0 1 1
25  '(10x,f10.0,f10.2,f10.2,30x,f10.2,50x,50x,30x,f10.2)'
26  1 2 3 5 4
```

Line 1 contains the title which is used on the KALFIMAC graphical output.
Line to define a Stochastic Level trend, a zero Cycle Lenght which is no cycle and 3 explanatory variables.
The row of zeros on line 3 is what makes this analysis a multiple-linear regression. It dictates that no elasticity is allowed for the Stochastic Level trend or the three variables included in the model.
The next four lines tell the program to use initiation period of 10 time-steps, no automatic optimization of elasticity settings (cf. Line 3), no Ln-transformation and to add a smoothing step after the initial filtering process.
The first number on line eight is a dummy, the second number instructs the program on the standardization to use. One (1) is the instruction for standardizing the explanatory variables.
Line 9 indicates that there will be no selection of explanatory variables, the second number indicates a forward or stepwise selection is as a result not functional in this case.
On the tenth line the presence of missing values is indicated and that their coded values is 0.99.
The next line is also about missing values. It tells the program that there are no ranges of missing values. If one or two of such ranges were present one or two lines would have to be added after this line each indicating the starting and ending timesteps of the ranges.
The twelfth line controls the output to file generated by KALFIMAC. The 10 on the fourth position is an instruction to calculate and present the autocorrelation function for up to ten time-steps.
The next lines that follow contain text to be used when generating plots and formatting options for the axes involved.
On Line 23 the number of records in the datafile is given: 730 days or two years in this case.
The twenty-fourth line is about prediction options. The initial zero indicates that no prediction of future values will be done.
Line 25 is the FORTRAN format defining the layout of the datafile. In combination with the final line in this option file, it governs the proper input of data into the program. This final line tells the program in which order to interpret the columns as read from the file. The 5 and 4 are out of sequence, indicating that the variables WindClass and RainTime are read from the file in reverse order but are used in their proper order.

Appendix C: Examples of Kalfimac option files

Example of a KALFIMAC filtering and smoothing option file: pm9394xd.opt.

```
1   PM10&Meteo 1993/94 T,WCl5,RT Cy=7 set stndrdzd
2   3 7 3
3   0.001 0.0 0.001 0.001 0.0
4   10
5   0
6   0
7   1
8   0 1
9   0 0
10  1 0.99
11  0
12  1 1 0 10 1
13  Concentration PM10 (µg/m3)
14  -4 10 7 2 8.0 1
15  Date
16  0 8 8 2 12.0 1
17  1.0
18  Temperature (0.1 oC)
19  WindCl5ss
20  RainTime (0.1h)
21  0 8 8 2 12.0 1
22  0 8 8 2 12.0 1
23  730
24  0 1 1
25  '(10x,f10.0,f10.2,f10.2,30x,f10.2,50x,50x,30x,f10.0)'
26  1 2 3 5 4
```

In discussing this option file most attention will be paid to the differences with the previous option file.
On line 2 the 7 adds the calculation of a 7 time-step cyclic pattern to the model.
On line 3 elasticity is added to the Stochastic Level trend, not to the cycle, but again added to Temperature and WindClass. No elasticity is allowed for RainTime.

Appendix D: Examples of Kalfimac data files

Appendix D: Example of a KALFIMAC data file

Date	DayNo	Bckgrnd	Temperatur	Linear	Linear-1	Linear-2	RainTime	MixHT	Rainfall	Humidity	Pressure	Insolation	Season	WindDirect	WindSpeed	WindRank	WindSector	WindClass	TempAlt	DegrDay	WindCl5ss
1/1/1993	1	172.47	-39.17	3.00	6.00	6.00	0.00	0.99	0.00	89.22	10353.83	396.00	2.00	120.00	15.00	8.00	1.00	4	120.00	156.90	8.00
2/1/1993	2	63.75	-35.67	3.00	3.00	6.00	0.00	0.99	0.00	73.25	10369.33	403.00	2.00	120.00	30.00	8.00	1.00	4	120.00	218.50	8.00
3/1/1993	3	86.02	-76.58	3.00	3.00	3.00	0.00	0.99	0.00	79.42	10354.17	371.00	2.00	100.00	25.00	9.00	1.00	4	120.00	235.63	8.00
4/1/1993	4	90.53	-52.00	5.00	3.00	3.00	0.00	0.99	0.00	76.83	10287.29	188.00	2.00	160.00	35.00	4.00	1.00	3	120.00	253.09	6.33
5/1/1993	5	81.45	13.29	5.00	5.00	3.00	91.00	0.99	86.00	83.83	10260.33	74.00	2.00	180.00	50.00	12.00	2.00	3	120.00	220.89	6.33
6/1/1993	6	23.54	78.88	7.00	5.00	5.00	110.00	0.99	137.00	97.88	10235.38	45.00	2.00	230.00	50.00	25.00	2.00	2	120.00	138.91	3.00
7/1/1993	7	25.53	45.17	7.00	7.00	5.00	22.00	0.99	10.00	98.67	10231.46	225.00	2.00	220.00	15.00	19.00	2.00	2	120.00	99.16	3.00
8/1/1993	8	31.95	56.17	8.00	7.00	7.00	30.00	0.99	28.00	82.88	10243.25	338.00	2.00	250.00	40.00	24.00	2.00	2	120.00	149.63	2.33
9/1/1993	9	34.06	66.13	6.00	8.00	7.00	35.00	0.99	32.00	91.25	10178.04	67.00	2.00	210.00	50.00	17.00	2.00	3	120.00	154.84	4.67
10/1/1993	10	26.20	106.25	6.00	6.00	8.00	51.00	0.99	25.00	87.88	10051.13	52.00	2.00	210.00	75.00	17.00	2.00	3	120.00	128.22	4.67
11/1/1993	11	22.61	100.50	7.00	6.00	6.00	22.00	0.99	35.00	82.29	9948.00	86.00	2.00	230.00	75.00	25.00	2.00	2	120.00	137.02	3.00
12/1/1993	12	23.39	59.88	7.00	7.00	6.00	47.00	0.99	34.00	76.25	10043.96	351.00	2.00	240.00	60.00	21.00	2.00	2	120.00	178.18	3.00
13/1/1993	13	28.19	70.54	6.00	7.00	7.00	39.00	0.99	23.00	80.63	10096.50	145.00	2.00	210.00	60.00	17.00	2.00	3	120.00	163.58	4.67
14/1/1993	14	28.80	65.83	7.00	6.00	7.00	0.00	0.99	0.00	74.63	10220.42	399.00	2.00	240.00	55.00	21.00	2.00	2	120.00	162.78	3.00
15/1/1993	15	31.08	85.38	6.00	7.00	6.00	4.00	0.99	2.00	81.96	10190.96	183.00	2.00	200.00	55.00	14.00	2.00	3	120.00	137.16	4.67
16/1/1993	16	36.11	115.38	7.00	6.00	7.00	0.00	0.99	0.00	80.88	10224.63	130.00	2.00	220.00	70.00	19.00	2.00	2	120.00	110.37	3.00
17/1/1993	17	36.08	93.13	7.00	7.00	6.00	7.00	0.99	5.00	77.00	10194.88	203.00	2.00	220.00	55.00	19.00	2.00	2	120.00	127.00	3.00

This is an example of the top few lines of a data file as used in this study. The PM10 data are taken from the combined station called Background and are placed in the column named Background. The meteorological variables are taken from the data for the KNMI station De Bilt. Please note that the first line with the column names should be deleted from the file before using it with KALFIMAC. The presence of this line is very useful when building the FORTRAN format on line 25 of the option files.

Appendix E: Correlationmatrices of meteorological variables per KNMI-station

Table E.1: Correlation matrices Temperature for 1993 and 1994

Correlationmatrix Temperature 1993

	De Kooy	Schiphol	De Bilt	Twente	Deelen	Rotterdam	Gilze-Rijen	Vlissingen	Eindhoven	Volkel	Maastricht
De Kooy	1.00	.99	.98	.97	.97	.98	.97	.98	.96	.97	.96
Schiphol	.99	1.00	1.00	.99	.99	1.00	.99	.98	.99	.99	.98
De Bilt	.98	1.00	1.00	.99	1.00	1.00	1.00	.98	.99	.99	.99
Twente	.97	.99	.99	1.00	1.00	.98	.99	.96	.99	.99	.98
Deelen	.97	.99	1.00	1.00	1.00	.99	.99	.97	.99	1.00	.99
Rotterdam	.98	1.00	1.00	.98	.99	1.00	.99	.99	.99	.99	.98
Gilze-Rijen	.97	.99	1.00	.99	.99	.99	1.00	.98	1.00	1.00	.99
Vlissingen	.98	.98	.98	.96	.97	.99	.98	1.00	.98	.97	.97
Eindhoven	.96	.99	.99	.99	.99	.99	1.00	.98	1.00	1.00	.99
Volkel	.97	.99	.99	.99	1.00	.99	1.00	.97	1.00	1.00	.99
Maastricht	.96	.98	.99	.98	.99	.98	.99	.97	.99	.99	1.00

Correlationmatrix Temperature 1994

	De Kooy	Schiphol	De Bilt	Twente	Deelen	Rotterdam	Gilze-Rijen	Vlissingen	Eindhoven	Volkel	Maastricht
De Kooy	1.00	.99	.98	.97	.98	.98	.97	.98	.97	.97	.96
Schiphol	.99	1.00	1.00	.98	.99	1.00	.99	.99	.99	.99	.98
De Bilt	.98	1.00	1.00	.99	1.00	.99	1.00	.98	.99	1.00	.98
Twente	.97	.98	.99	1.00	.99	.98	.99	.97	.99	.99	.98
Deelen	.98	.99	1.00	.99	1.00	.99	.99	.98	1.00	1.00	.99
Rotterdam	.98	1.00	.99	.98	.99	1.00	.99	.99	.99	.99	.98
Gilze-Rijen	.97	.99	1.00	.99	.99	.99	1.00	.98	1.00	1.00	.99
Vlissingen	.98	.99	.98	.97	.98	.99	.98	1.00	.98	.98	.97
Eindhoven	.97	.99	.99	.99	1.00	.99	1.00	.98	1.00	1.00	.99
Volkel	.97	.99	1.00	.99	1.00	.99	1.00	.98	1.00	1.00	.99
Maastricht	.96	.98	.98	.98	.99	.98	.99	.97	.99	.99	1.00

Table E.2: Correlation matrices Wind direction for 1993 and 1994

Correlationmatrix WindDirection 1993

	Valke210	De Ko235	Schip240	De Bi260	Deele275	Twent290	Vliss310	Rotte344	Gilze350	Eindh370	Volke375	Maast380
Valke210	1.00	.84	.88	.78	.67	.59	.71	.81	.77	.73	.70	.74
De Ko235	.84	1.00	.87	.74	.66	.61	.72	.73	.70	.65	.62	.68
Schip240	.88	.87	1.00	.82	.72	.67	.80	.77	.77	.68	.64	.70
De Bi260	.78	.74	.82	1.00	.85	.75	.78	.83	.86	.84	.80	.77
Deele275	.67	.66	.72	.85	1.00	.86	.72	.72	.75	.84	.87	.75
Twent290	.59	.61	.67	.75	.86	1.00	.68	.67	.65	.71	.78	.71
Vliss310	.71	.72	.80	.78	.72	.68	1.00	.80	.77	.74	.70	.73
Rotte344	.81	.73	.77	.83	.72	.67	.80	1.00	.83	.79	.77	.81
Gilze350	.77	.70	.77	.86	.75	.65	.77	.83	1.00	.86	.79	.79
Eindh370	.73	.65	.68	.84	.84	.71	.74	.79	.86	1.00	.87	.85
Volke375	.70	.62	.64	.80	.87	.78	.70	.77	.79	.87	1.00	.79
Maast380	.74	.68	.70	.77	.75	.71	.73	.81	.79	.85	.79	1.00
Cabau10034	.75	.69	.76	.85	.88	.75	.75	.76	.81	.78	.79	.71

Correlationmatrix WindDirection 1994

	Valke210	De Ko235	Schip240	De Bi260	Deele275	Twent290	Vliss310	Rotte344	Gilze350	Eindh370	Volke375	Maast380
Valke210	1.00	.81	.85	.89	.84	.79	.74	.96	.79	.77	.80	.71
De Ko235	.81	1.00	.91	.80	.79	.70	.79	.78	.75	.70	.73	.66
Schip240	.85	.91	1.00	.81	.77	.71	.80	.83	.81	.77	.78	.68
De Bi260	.89	.80	.81	1.00	.93	.77	.78	.86	.81	.80	.86	.74
Deele275	.84	.79	.77	.93	1.00	.78	.76	.81	.80	.78	.87	.74
Twent290	.79	.70	.71	.77	.78	1.00	.64	.77	.71	.71	.74	.71
Vliss310	.74	.79	.80	.78	.76	.64	1.00	.72	.81	.76	.72	.76
Rotte344	.96	.78	.83	.86	.81	.77	.72	1.00	.82	.81	.82	.74
Gilze350	.79	.75	.81	.81	.80	.71	.81	.82	1.00	.93	.84	.80
Eindh370	.77	.70	.77	.80	.78	.71	.76	.81	.93	1.00	.83	.85
Volke375	.80	.73	.78	.86	.87	.74	.72	.82	.84	.83	1.00	.80
Maast380	.71	.66	.68	.74	.74	.71	.76	.74	.80	.85	.80	1.00
Cabau10034	.89	.77	.82	.91	.89	.79	.79	.86	.90	.89	.82	.76

Table E.3: Correlation matrices Wind speed for 1993 and 1994

Correlationmatrix WindSpeed 1993

	Valke210	De Ko235	Schip240	De Bi260	Deele275	Twent290	Vliss310	Rotte344	Gilze350	Eindh370	Volke375	Maast380
Valke210	1.00	.89	.96	.92	.90	.87	.87	.96	.88	.89	.88	.79
De Ko235	.89	1.00	.92	.88	.86	.86	.79	.86	.79	.80	.83	.71
Schip240	.96	.92	1.00	.94	.93	.89	.87	.95	.90	.90	.91	.80
De Bi260	.92	.88	.94	1.00	.95	.89	.88	.91	.92	.92	.92	.83
Deele275	.90	.86	.93	.95	1.00	.93	.85	.90	.93	.93	.95	.84
Twent290	.87	.86	.89	.89	.93	1.00	.80	.88	.87	.89	.90	.82
Vliss310	.87	.79	.87	.88	.85	.80	1.00	.91	.88	.90	.87	.88
Rotte344	.96	.86	.95	.91	.90	.88	.91	1.00	.90	.92	.90	.85
Gilze350	.88	.79	.90	.92	.93	.87	.88	.90	1.00	.96	.93	.87
Eindh370	.89	.80	.90	.92	.93	.89	.90	.92	.96	1.00	.96	.93
Volke375	.88	.83	.91	.92	.95	.90	.87	.90	.93	.96	1.00	.90
Maast380	.79	.71	.80	.83	.84	.82	.88	.85	.87	.93	.90	1.00
Cabau10034	.94	.87	.96	.95	.94	.91	.91	.97	.93	.95	.94	.87

Correlationmatrix WindSpeed 1994

	Valke210	De Ko235	Schip240	De Bi260	Deele275	Twent290	Vliss310	Rotte344	Gilze350	Eindh370	Volke375	Maast380
Valke210	1.00	.89	.96	.92	.92	.91	.88	.96	.89	.91	.89	.85
De Ko235	.89	1.00	.92	.88	.88	.89	.80	.86	.81	.81	.84	.75
Schip240	.96	.92	1.00	.96	.95	.93	.89	.95	.91	.92	.92	.86
De Bi260	.92	.88	.96	1.00	.97	.92	.89	.92	.93	.93	.94	.86
Deele275	.92	.88	.95	.97	1.00	.95	.88	.92	.93	.94	.95	.88
Twent290	.91	.89	.93	.92	.95	1.00	.83	.90	.89	.91	.92	.86
Vliss310	.88	.80	.89	.89	.88	.83	1.00	.93	.90	.92	.90	.91
Rotte344	.96	.86	.95	.92	.92	.90	.93	1.00	.92	.94	.91	.89
Gilze350	.89	.81	.91	.93	.93	.89	.90	.92	1.00	.95	.93	.90
Eindh370	.91	.81	.92	.93	.94	.91	.92	.94	.95	1.00	.97	.95
Volke375	.89	.84	.92	.94	.95	.92	.90	.91	.93	.97	1.00	.94
Maast380	.85	.75	.86	.86	.88	.86	.91	.89	.90	.95	.94	1.00
Cabau10034	.94	.87	.96	.96	.95	.92	.92	.96	.93	.95	.95	.90

Appendix E: Correlationmatrices of meteorological variables per KNMI-station

Table E.4: Correlation matrices Atmospheric Pressure 1993 and 1994

Correlationmatrix Atmospheric Pressure 1993

	De Kooy	Schiphol	De Bilt	Twente	Deelen	Rotterdam	Gilze-Rijen	Vlissingen	Eindhoven	Volkel	Maastricht
De Kooy	1.00	1.00	.99	.99	.99	.99	.98	.98	.98	.98	.96
Schiphol	1.00	1.00	1.00	.99	1.00	1.00	.99	.99	.99	.99	.98
De Bilt	.99	1.00	1.00	1.00	1.00	1.00	1.00	.99	1.00	1.00	.99
Twente	.99	.99	1.00	1.00	1.00	.99	.99	.98	.99	.99	.98
Deelen	.99	1.00	1.00	1.00	1.00	1.00	1.00	.99	1.00	1.00	.99
Rotterdam	.99	1.00	1.00	.99	1.00	1.00	1.00	1.00	1.00	1.00	.99
Gilze-Rijen	.98	.99	1.00	.99	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Vlissingen	.98	.99	.99	.98	.99	1.00	1.00	1.00	1.00	.99	.99
Eindhoven	.98	.99	1.00	.99	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Volkel	.98	.99	1.00	.99	1.00	1.00	1.00	.99	1.00	1.00	.99
Maastricht	.96	.98	.99	.98	.99	.99	1.00	.99	1.00	.99	1.00

Correlationmatrix Atmospheric Pressure 1994

	De Kooy	Schiphol	De Bilt	Twente	Deelen	Rotterdam	Gilze-Rijen	Vlissingen	Eindhoven	Volkel	Maastricht
De Kooy	1.00	1.00	.99	.99	.99	.99	.98	.97	.98	.98	.95
Schiphol	1.00	1.00	1.00	.99	1.00	1.00	.99	.99	.99	.99	.98
De Bilt	.99	1.00	1.00	.99	1.00	1.00	1.00	.99	1.00	1.00	.98
Twente	.99	.99	.99	1.00	1.00	.99	.99	.97	.99	.99	.97
Deelen	.99	1.00	1.00	1.00	1.00	1.00	1.00	.99	1.00	1.00	.98
Rotterdam	.99	1.00	1.00	.99	1.00	1.00	1.00	1.00	1.00	1.00	.99
Gilze-Rijen	.98	.99	1.00	.99	1.00	1.00	1.00	1.00	1.00	1.00	.99
Vlissingen	.97	.99	.99	.97	.99	1.00	1.00	1.00	.99	.99	.99
Eindhoven	.98	.99	1.00	.99	1.00	1.00	1.00	.99	1.00	1.00	1.00
Volkel	.98	.99	1.00	.99	1.00	1.00	1.00	.99	1.00	1.00	.99
Maastricht	.95	.98	.98	.97	.98	.99	.99	.99	1.00	.99	1.00

Table E.5: Correlation matrices Amount of Precipitation for 1993 and 1994

Correlationmatrix Amount of Precipitation 1993

	De Kooy	Schiphol	De Bilt	Twente	Deelen	Rotterdam	Gilze-Rijen	Vlissingen	Eindhoven	Volkel	Maastricht
De Kooy	1.00	.72	.59	.58	.59	.62	.47	.50	.45	.52	.37
Schiphol	.72	1.00	.78	.62	.74	.79	.68	.70	.63	.67	.49
De Bilt	.59	.78	1.00	.69	.83	.86	.73	.70	.71	.75	.55
Twente	.58	.62	.69	1.00	.80	.62	.56	.51	.66	.77	.70
Deelen	.59	.74	.83	.80	1.00	.76	.71	.58	.75	.87	.71
Rotterdam	.62	.79	.86	.62	.76	1.00	.78	.83	.74	.73	.52
Gilze-Rijen	.47	.68	.73	.56	.71	.78	1.00	.76	.86	.75	.57
Vlissingen	.50	.70	.70	.51	.58	.83	.76	1.00	.69	.60	.41
Eindhoven	.45	.63	.71	.66	.75	.74	.86	.69	1.00	.86	.72
Volkel	.52	.67	.75	.77	.87	.73	.75	.60	.86	1.00	.81
Maastricht	.37	.49	.55	.70	.71	.52	.57	.41	.72	.81	1.00

Correlationmatrix Amount of Precipitation 1994

	De Kooy	Schiphol	De Bilt	Twente	Deelen	Rotterdam	Gilze-Rijen	Vlissingen	Eindhoven	Volkel	Maastricht
De Kooy	1.00	.78	.58	.56	.53	.63	.48	.51	.47	.49	.43
Schiphol	.78	1.00	.74	.65	.65	.77	.66	.63	.65	.69	.58
De Bilt	.58	.74	1.00	.68	.75	.81	.77	.68	.72	.72	.61
Twente	.56	.65	.68	1.00	.79	.65	.71	.51	.70	.74	.55
Deelen	.53	.65	.75	.79	1.00	.69	.73	.59	.69	.75	.59
Rotterdam	.63	.77	.81	.65	.69	1.00	.79	.72	.72	.72	.67
Gilze-Rijen	.48	.66	.77	.71	.73	.79	1.00	.67	.87	.83	.68
Vlissingen	.51	.63	.68	.51	.59	.72	.67	1.00	.63	.59	.63
Eindhoven	.47	.65	.72	.70	.69	.72	.87	.63	1.00	.86	.74
Volkel	.49	.69	.72	.74	.75	.72	.83	.59	.86	1.00	.70
Maastricht	.43	.58	.61	.55	.59	.67	.68	.63	.74	.70	1.00

Table E.6: Correlation matrices Duration of Precipitation for 1993 and 1994

Correlationmatrix Duration of Precipitation 1993

	De Kooy	Schiphol	De Bilt	Twente	Deelen	Rotterdam	Gilze-Rijen	Vlissingen	Eindhoven	Volkel	Maastricht
De Kooy	1.00	.86	.79	.71	.75	.80	.70	.66	.65	.70	.53
Schiphol	.86	1.00	.91	.78	.87	.92	.86	.80	.82	.84	.69
De Bilt	.79	.91	1.00	.81	.91	.90	.89	.81	.81	.86	.70
Twente	.71	.78	.81	1.00	.91	.75	.78	.65	.80	.84	.72
Deelen	.75	.87	.91	.91	1.00	.84	.86	.75	.87	.91	.79
Rotterdam	.80	.92	.90	.75	.84	1.00	.89	.87	.83	.83	.70
Gilze-Rijen	.70	.86	.89	.78	.86	.89	1.00	.86	.92	.88	.81
Vlissingen	.66	.80	.81	.65	.75	.87	.86	1.00	.81	.78	.65
Eindhoven	.65	.82	.81	.80	.87	.83	.92	.81	1.00	.93	.89
Volkel	.70	.84	.86	.84	.91	.83	.88	.78	.93	1.00	.84
Maastricht	.53	.69	.70	.72	.79	.70	.81	.65	.89	.84	1.00

Correlationmatrix Duration of Precipitation 1994

	De Kooy	Schiphol	De Bilt	Twente	Deelen	Rotterdam	Gilze-Rijen	Vlissingen	Eindhoven	Volkel	Maastricht
De Kooy	1.00	.89	.81	.76	.78	.81	.71	.67	.70	.71	.61
Schiphol	.89	1.00	.91	.79	.85	.91	.81	.76	.79	.80	.68
De Bilt	.81	.91	1.00	.86	.91	.93	.85	.76	.84	.87	.73
Twente	.76	.79	.86	1.00	.93	.80	.82	.67	.82	.87	.76
Deelen	.78	.85	.91	.93	1.00	.87	.88	.74	.88	.92	.80
Rotterdam	.81	.91	.93	.80	.87	1.00	.88	.82	.85	.86	.75
Gilze-Rijen	.71	.81	.85	.82	.88	.88	1.00	.88	.95	.93	.88
Vlissingen	.67	.76	.76	.67	.74	.82	.88	1.00	.84	.80	.79
Eindhoven	.70	.79	.84	.82	.88	.85	.95	.84	1.00	.95	.91
Volkel	.71	.80	.87	.87	.92	.86	.93	.80	.95	1.00	.87
Maastricht	.61	.68	.73	.76	.80	.75	.88	.79	.91	.87	1.00

Appendix E: Correlationmatrices of meteorological variables per KNMI-station

Table E.7: Correlation matrices for Relative Humidity for 1993 and 1994

Correlationmatrix Relative Humidity 1993

	De Kooy	Schiphol	De Bilt	Twente	Deelen	Rotterdam	Gilze-Rijen	Vlissingen	Eindhoven	Volkel	Maastricht
De Kooy	1.00	.80	.74	.65	.66	.74	.70	.70	.64	.66	.59
Schiphol	.80	1.00	.94	.81	.89	.94	.89	.84	.87	.87	.80
De Bilt	.74	.94	1.00	.87	.93	.94	.94	.83	.91	.92	.83
Twente	.65	.81	.87	1.00	.93	.81	.85	.66	.86	.90	.83
Deelen	.66	.89	.93	.93	1.00	.88	.91	.75	.92	.94	.85
Rotterdam	.74	.94	.94	.81	.88	1.00	.94	.87	.89	.88	.81
Gilze-Rijen	.70	.89	.94	.85	.91	.94	1.00	.86	.96	.93	.88
Vlissingen	.70	.84	.83	.66	.75	.87	.86	1.00	.81	.78	.73
Eindhoven	.64	.87	.91	.86	.92	.89	.96	.81	1.00	.94	.91
Volkel	.66	.87	.92	.90	.94	.88	.93	.78	.94	1.00	.89
Maastricht	.59	.80	.83	.83	.85	.81	.88	.73	.91	.89	1.00

Correlationmatrix Relative Humidity 1994

	De Kooy	Schiphol	De Bilt	Twente	Deelen	Rotterdam	Gilze-Rijen	Vlissingen	Eindhoven	Volkel	Maastricht
De Kooy	1.00	.86	.76	.71	.73	.81	.73	.77	.68	.71	.63
Schiphol	.86	1.00	.95	.86	.90	.96	.90	.82	.87	.88	.78
De Bilt	.76	.95	1.00	.92	.95	.95	.94	.77	.93	.92	.83
Twente	.71	.86	.92	1.00	.96	.88	.91	.68	.91	.92	.87
Deelen	.73	.90	.95	.96	1.00	.92	.95	.75	.95	.95	.89
Rotterdam	.81	.96	.95	.88	.92	1.00	.93	.81	.91	.91	.82
Gilze-Rijen	.73	.90	.94	.91	.95	.93	1.00	.82	.98	.97	.90
Vlissingen	.77	.82	.77	.68	.75	.81	.82	1.00	.78	.80	.75
Eindhoven	.68	.87	.93	.91	.95	.91	.98	.78	1.00	.97	.92
Volkel	.71	.88	.92	.92	.95	.91	.97	.80	.97	1.00	.91
Maastricht	.63	.78	.83	.87	.89	.82	.90	.75	.92	.91	1.00

Table E.8: Correlation matrices Insolation for 1993 and 1994

Correlationmatrix Insolation 1993

	De Kooy	Schiphol	De Bilt	Twente	Deelen	Rotterdam	Gilze-Rijen	Vlissingen	Eindhoven	Volkel	Maastricht
De Kooy	1.00	.97	.94	.90	.92	.95	.93	.93	.91	.93	.87
Schiphol	.97	1.00	.97	.91	.95	.99	.96	.95	.94	.95	.90
De Bilt	.94	.97	1.00	.94	.98	.98	.97	.94	.96	.97	.92
Twente	.90	.91	.94	1.00	.97	.91	.93	.87	.94	.95	.91
Deelen	.92	.95	.98	.97	1.00	.95	.96	.90	.96	.97	.91
Rotterdam	.95	.99	.98	.91	.95	1.00	.97	.97	.95	.96	.91
Gilze-Rijen	.93	.96	.97	.93	.96	.97	1.00	.95	.99	.98	.94
Vlissingen	.93	.95	.94	.87	.90	.97	.95	1.00	.94	.93	.90
Eindhoven	.91	.94	.96	.94	.96	.95	.99	.94	1.00	.98	.96
Volkel	.93	.95	.97	.95	.97	.96	.98	.93	.98	1.00	.95
Maastricht	.87	.90	.92	.91	.91	.91	.94	.90	.96	.95	1.00

Correlationmatrix Insolation 1994

	De Kooy	Schiphol	De Bilt	Twente	Deelen	Rotterdam	Gilze-Rijen	Vlissingen	Eindhoven	Volkel	Maastricht
De Kooy	1.00	.95	.92	.88	.91	.93	.91	.92	.89	.89	.86
Schiphol	.95	1.00	.96	.90	.94	.98	.95	.94	.93	.93	.88
De Bilt	.92	.96	1.00	.94	.98	.97	.97	.92	.96	.96	.90
Twente	.88	.90	.94	1.00	.97	.90	.92	.85	.93	.95	.89
Deelen	.91	.94	.98	.97	1.00	.94	.96	.90	.96	.98	.92
Rotterdam	.93	.98	.97	.90	.94	1.00	.96	.96	.94	.94	.88
Gilze-Rijen	.91	.95	.97	.92	.96	.96	1.00	.94	.98	.98	.93
Vlissingen	.92	.94	.92	.85	.90	.96	.94	1.00	.92	.91	.88
Eindhoven	.89	.93	.96	.93	.96	.94	.98	.92	1.00	.98	.95
Volkel	.89	.93	.96	.95	.98	.94	.98	.91	.98	1.00	.94
Maastricht	.86	.88	.90	.89	.92	.88	.93	.88	.95	.94	1.00

Appendix F: Correlation matrix of PM10, relative humidity and hour for July 1994

Station name	Correlation coefficient PM10 : RH	RH : Hour
Witteveen	-0.00	-0.03
Wieringerwerf	-0.11	-0.02
Amsterdam-Noord	-0.10	0.04
De Zilk	-0.20	0.18
Breukelen-snelweg	0.10	-0.10
Utrecht-Erzeijstraat	-0.00	-0.07
Eibergen	0.12	-0.10
Apeldoorn-Stationsstraat	-0.13	0.14
Wageningen	0.09	-0.07
Den Haag-centrum	-0.13	-0.00
Vlaardingen-macro	-0.05	0.00
Rotterdam-centrum	-0.11	0.04
Westmaas	-0.11	-0.03
Dordrecht	-0.10	0.04
Houtakker	0.04	-0.07
Braakman	-0.09	-0.00
Eindhoven-Genovevelaan	-0.01	0.03
Vredepeel	0.13	-0.02
Wijnandsrade	-0.06	0.06
Hour	-0.63	