Ministry of
the Environment

National Environmental
Research Institute

The Danish Air Quality Monitoring Programme

NERI, Technical Report No. 60
1992

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The Danish Air Quality Monitoring Programme (LMP II)

Annual data report 1991

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Finn Palmgren Jensen
Kåre Kemp
Ole H. Manscher
Department of Emissions and Air Pollution

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Content

Preface

Monitoring Programmes

The goal of the first Danish Air Quality Monitoring Programme (Det landsdækkende Luftkvalitetsmåleprogram, LMP) was to give knowledge of the air quality in Danish urban areas (*Palmgren Jensen, F., 1987*). Within the programme the actual air quality was monitored at 30 monitoring stations in 7 Danish cities. In addition, methods for calculations of the air quality by meteorological air quality models were developed and applied in the Danish urban areas (*Berkowicz, R. et al., 1986, Løfstrøm, P and Rørdam Olesen, H., 1988*). The programme was initiated in 1982 and was finished by the end of 1986. Based on the results and the experiences from this programme a revised programme was started at the beginning of 1987 (LMP II), comprising monitoring in 6 urban areas only. The programme was revised again in 1991 and a new programme (LMP III) was started January 1992 in 3 Danish Urban areas.

Present Programme

The present programme (LMP II) was financed by the participating municipalities, Greater Copenhagen Council (after January 1st 1990, Greater Copenhagen Air Pollution Unit, HLU, which is a cooperation between the Municipalities of Copenhagen and Frederiksberg and the counties of Copenhagen, Frederiksborg and Roskilde), National Environmental Protection Agency and National Environmental Research Institute.

Annual Report 1991

This annual report of 1991 is a technical and a data report. A summarizing report for the period 1987-91 will be prepared separately.

The present report includes a description of the monitoring stations, the monitoring and analysis methods and a summary of all collected data.

In every chapter tables and graphs are given for all urban areas ranked after station numbers in the following order: Copenhagen, Fredericia, Esbjerg, Århus, Aalborg and Odense.

Summary

Previous programmes

In 1982 a Danish Air Quality Monitoring Programme was established as a cooperative effort between the authorities of the government, counties, municipalities and Greater Copenhagen Council. The programme was carried out in 7 Danish cities including Greater Copenhagen (*Palmgren Jensen and Kemp, 1986, 1987, 1988*). In addition to the monitoring, the programme included emission inventories and meteorological model calculations of sulphur dioxide (*Berkowicz et al., 1986*).

Present programme

The network was revised in 1987. Greater importance is now attached to monitoring of air pollutants from the traffic. The network includes monitoring of gasses and particulates. Many stations are located at kerb sides. Data from the monitoring stations are transmitted directly to NERI by public telephone lines.

Annual report 1991

The present report is summary of data from 1991. The report includes all collected data from all urban areas, reduced and presented in graphs and tables. Data from the previous years are reported by (*Palmgren Jensen, F. et al 1990 and 1991*).

Levels

The measurements showed that no Danish or EEC air quality limits values were exceeded at the monitoring sites. However, Danish, EEC and WHO guidelines for NO_2 were exceeded in the centre of Copenhagen.

Trends

During the monitoring programme the measurements showed slightly increasing concentrations of NO and NO_2 in most of the urban areas in Denmark. This was due to increasing traffic. In 1990 and 1991 the level of NO_2 was lower than the previous years, probably due to mild winters.

In the long term the emission of NO_x will be reduced due to limits on the emission from power plants and especially due to introduction of catalysts on all new cars from October 1990.

A very clear reduction in the concentration of lead in urban air was observed since the late seventies, i.e. a factor of 5 corresponding to the reduction of lead content in petrol from approx. 0.56 g lead/litre to approx. 0.1 g lead/litre.

In the same period the concentration of SO_2 was reduced a factor of 2 due energy saving, more district heating (with tall stacks instead of small chimneys), generally higher stacks, limits on sulphur content in fuel, increasing use of natural gas and more efficient combustion.

Sources

The local traffic was generally the most important air pollution source in Danish urban areas in the period 1982-91. However, other sources gave considerable contributions to the air pollution e.g. power plants, industries, house heating and distant sources in our neighbour countries. The contributions from different sources were demonstrated by analyses of the measured concentration in relation to meteorological data.

Future measurements

In the programme only traditional inorganic components were measured. From January 1992 the monitoring programme will include more components. The greatest emphasis will be laid on components describing the atmospheric processes to be included in air quality models and the toxic/carcinogenic substances, e.g. O_3 and organics.

Danish summary

LMP II 1991

Rapporten indeholder en oversigt over måleresultater fra det landsdækkende luftkvalitetsmåleprogram ajourført med data fra 1991.

Niveauer

Måleresultaterne for 1990 og 1991 viser, at niveauerne for de forureningskomponenter, der findes bindende grænseværdier for, i alle tilfælde ligger under grænseværdierne. Ud over de bindende grænseværdier har EF og WHO angivet vejledende grænseværdier for en række stoffer. Hvis disse vejledende grænseværdier overskrides anbefales det, at der tages forholdsregler så forholdene på længere sigt forbedres. Af de stoffer, som er omfattet af LMP målingerne er kun NO_2 koncentrationerne så store, at der er overskridelse af de vejledende værdier.

Udvikling

Siden 1982 har der været stigende NO_2 forurening i alle byer, i det væsentlige på grund af stigende trafik. Der var dog færre overskridelser i 1991 end i de foregående år, hvilket formentlig skyldes de meteorologiske forhold. Bl.a. for at nedsætte NO_2 koncentrationerne har man vedtaget, at alle nye benzindrevne biler skal udstyres med katalysatorer fra oktober 1990 og at der senest fra 2005 skal ske en reduktion af NO_x emissionen fra kraftværkerne. Selv om disse foranstaltninger på længere sigt vil virke i positiv retning, er der ikke udsigt til en øjeblikkelig forbedring.

Man kan konstatere en yderst gunstig udvikling af koncentrationen af bly i luften, idet den er faldet med ca. en faktor 5 i den tid LMP målingerne har været foretaget. Det er sket i takt med fjernelsen af bly i benzin fra ca. 0,56 g/l i begyndelsen af 1980erne til ca. 0.1 g/l. Faldet er fortsat i 1991, men på grund af det lave niveau er faldet mindre end tidligere. I løbet af de næste 10 år vil bly forsvinde fuldstændig fra benzin.

SO_2 koncentrationerne er i samme period faldet med en faktor to, Det skyldes bl.a. nedsættelsen af svovlindholdet i olieprodukter i 1986. Men også indførelsen af naturgas og bedre røgrensning har bidraget og kan ventes i de kommende år at bidrage til et fortsat gradvist fald i SO_2 niveauerne.

Fremtidige målinger

De stoffer, der er blevet målt i LMP programmerne, udgør kun en lille del af de toksiske stoffer, der findes i atmosfæren. De stoffer, der er valgt, har imidlertid sat os i stand til at give en vurdering af forureningskilderne. Den vil sammen med de meteorologisk baserede modeller, der er udviklet, gøre det muligt at beskrive udbredelsen af andre stoffer. Det er især organiske stoffer og ozon, man bør måle, bl.a. fordi kendskabet til oprindelse, forekomst og omdannelse er mangelfuldt. Dette vil i et vist omfang blive tilgo-

FAX til: Finn Palgren, FOLU

Danmarks Miljøundersøgelser
Forureningskilder og luftforurening
c/o Niels Bohr Institutet
Blegdamsvej 17
2100 København Ø
Telf.: 3532 5237, fax: 3142 1016

4. januar 1994

Finn Palgren
FOLU

Kære Finn,

Hermed de nye rettelser til LMP 91 rapporten. Jeg sender dem for en sikkerheds skyld gennem dig. Vil du give dem videre til Lene, som venter på dem?

Side 6, afsnittet mærket *Trends*. De tre første linier erstattes med:

> NO do show an increasing trend over the whole period 1982-1991, while no significant trend is observed for NO_2. In 1990

Side 8, afsnittet mærket *Udvikling*. De tre første linier erstattes med:

> I perioden 1982-1991 har vi ikke konstateret en signifikant ændring af NO_2 forureningen. Der var færre overskridelser af de vejledende grænseværdier i 1991 end i de foregående år, hvilket formentlig skyldes

Jeg medsender eksempler på de rettede sider.

Hilsen

Kåre

Summary

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deset i forbindelse med LMP III, som startede 1. januar 1992 i hovedstadområdet, Aalborg og Odense. De øvrige byer har ikke ønsket at deltage i LMP III.

1 Description of monitoring stations

Monitoring stations

The programme (LMP II) comprises monitoring at 13 stations. 3 are located in Copenhagen and 2 in each of the following cities: Aalborg, Århus, Esbjerg, Fredericia and Odense. The stations in Copenhagen are located in the City Centre. They are supplemented by the 5 stations of the Greater Copenhagen Air Pollution Unit (HLU). In the other cities one station are located in the city centre and the other in the urban areas (residential and industrial area). The exact locations are shown in table 1.1 and figures 1.1-6.

Filter samplers

All stations include a automatic filter sampler developed by NERI (*Kemp, 1985*). The sampler collects particles on disc filters (Millipore type RA) and SO_2 on KOH impregnated cellulose fibre filters (Whatmann 1). The filters are mounted in series in a filter holder. The exposed area has a diameter of 40 $mm^{\varnothing}$. Approximately 60 m^3 of air are sucked through the filters during 24 hours. 8 filter holders mounted at the sampler allow operation without inspection for a week (figure 1.7).

Monitors

All stations in this programme in Copenhagen and the centre stations in the other cities are equipped with monitors for NO and NO_x (WESTERBERG, 1983) based on chemiluminescence reaction between ozone and NO. The time constant of the monitors is 1 minute and ½ hour averages are calculated by a local computer. The data are transmitted every 24 hours or upon request to NERI via the public telephone network.

Stations for NO_x are also equipped with SO_2-monitors except at 1255, 1257 and 1258. Monitoring and data collection of SO_2 results are carried out by the same procedures as for the NO/NO_x. The SO_2-monitors are based on UV-fluorescence.

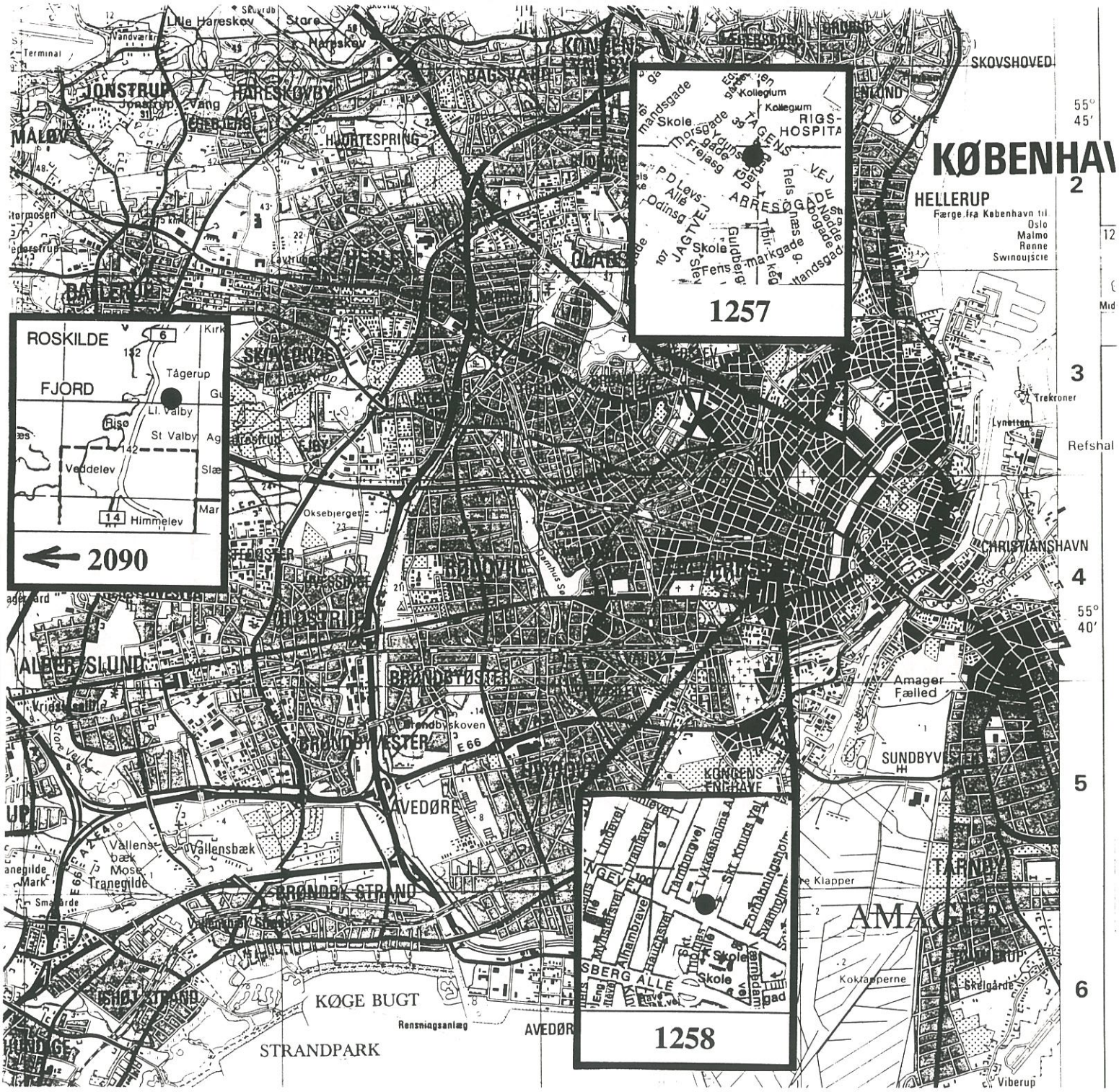
Operation and maintenance

The local laboratories are responsible for operation of the stations, i.e. inspection once a week and shift of filters. NERI perform a detailed control and calibration every 1-3 months after a fixed schedule (*Westerberg and Lund Thomsen, 1983, Westerberg, 1983*). Repairs are carried out by NERI.

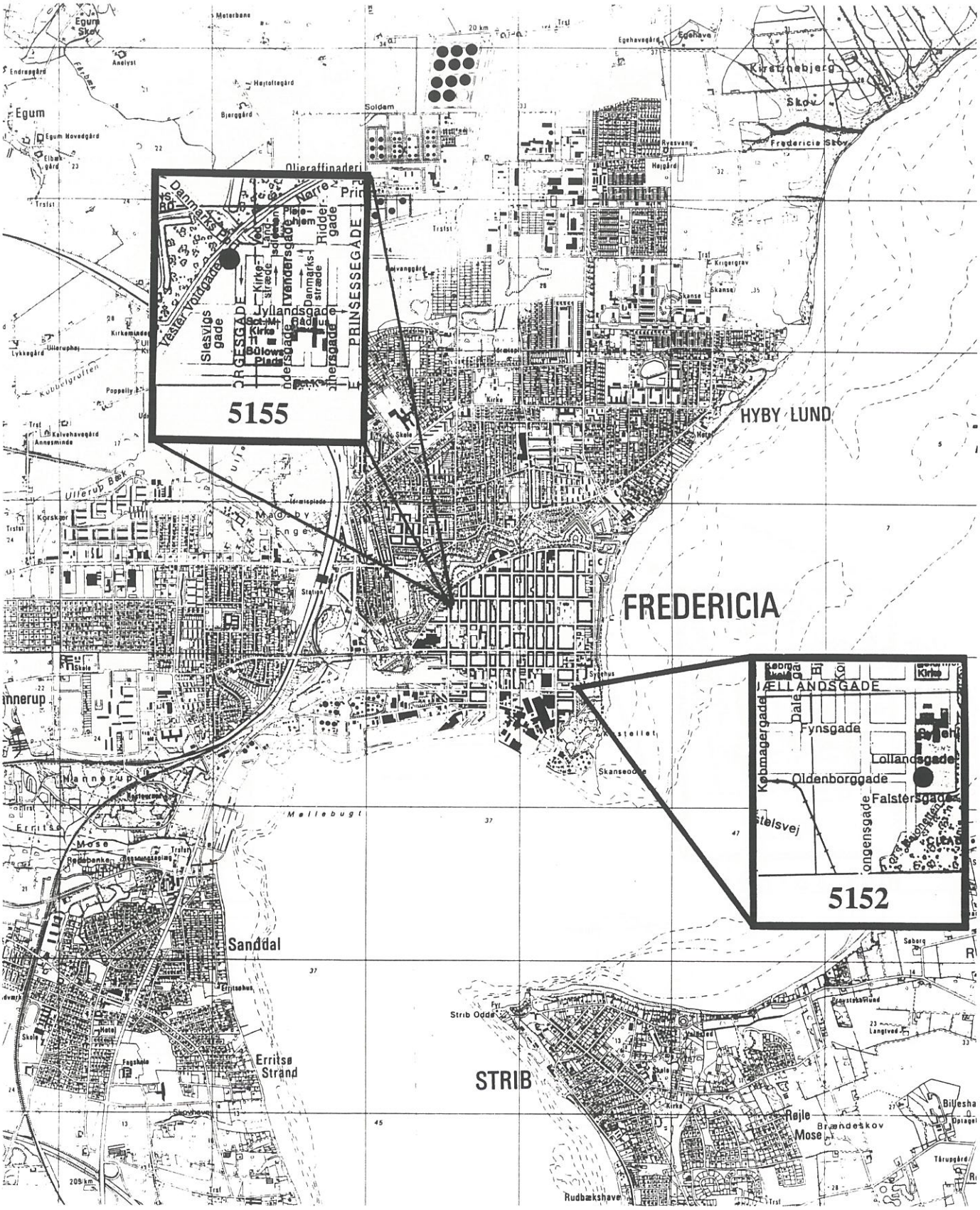
Table 1.1. Monitoring stations in the monitoring programme (LMP II). Traffic counts are based on information from the municipalities as number of vehicles per 24 hours in 1989 or 1990. For 8151 the number is the sum of the counts at Nyhavnsgade and Østre Allé.

Station number	City	Street	Start date	Traffic intensity (vehicles/24 h)	Monitoring programme *
1255	Copenhagen	Åboulevard	870912	50000	II
1256		Tiranglen	870915	37000	II
1257		Jagtvej	870919	22000	III
1258		Gl.Kongevej	910314	20000	II
2090		Ll. Valby	910601	-	III
5155	Fredericia	Danmarkseksport	820901	6000	III
5152		Sygehuset	820224	-	I
5655	Esbjerg	Skolegade	820525	6500	III
5652		Jagtvej	820224	6000	I
6151	Århus	Åboulevard	880423	13000	III
6152		Frederiks A.	820520	24000	I
8151	Ålborg	Limfjordsbro	820311	28500	III
8153		Nyhavnsgade	880311	22000	I
9155	Odense	Albanigade	820209	19500	III
9154		Ringvejen	820225	19500	I

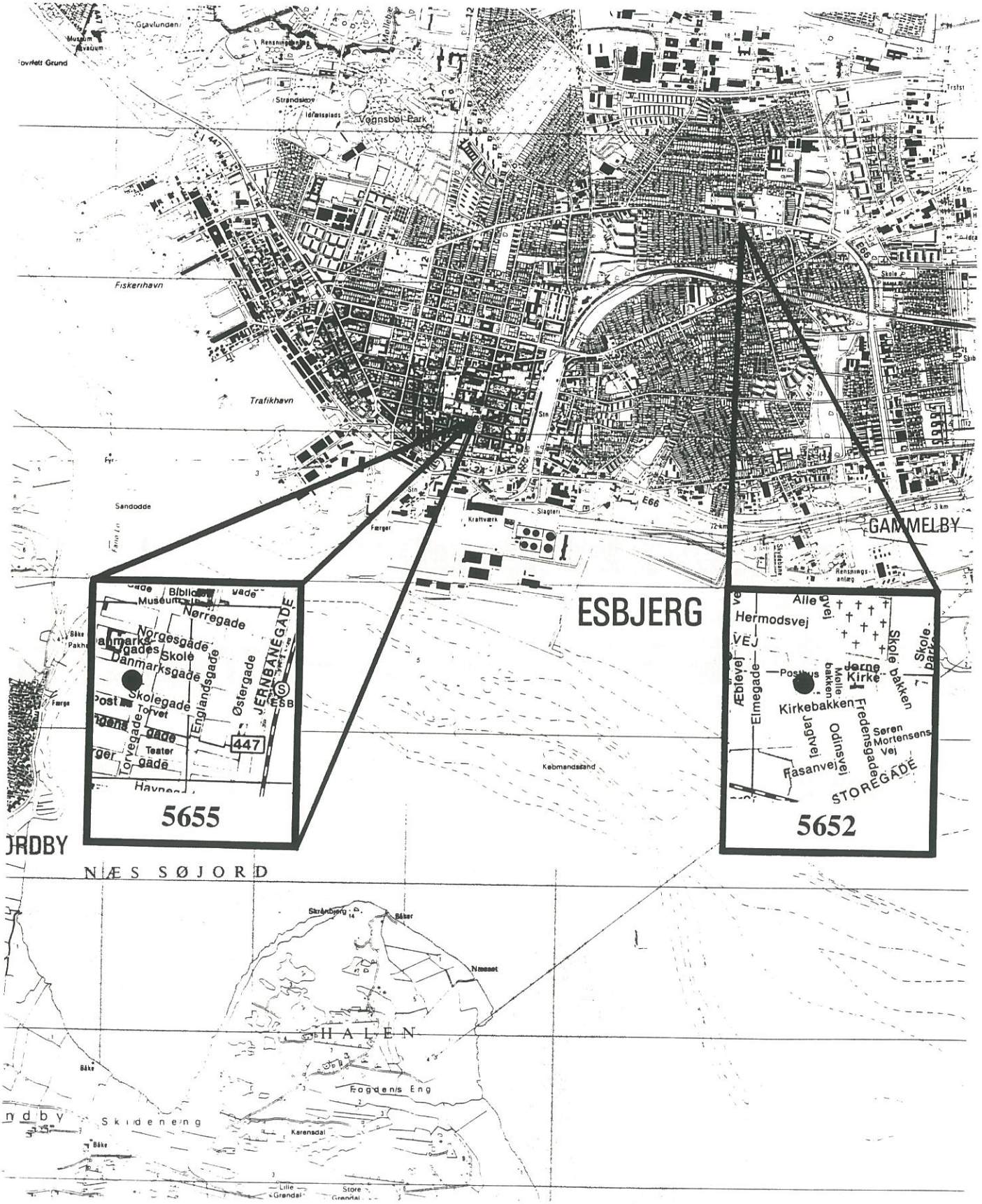
*) I : 24 hour measurements of SO_2, TSP and elements.
II : I + ½ hour measurements of NO of NO_2.
III: II + ½ hour measurements of SO_2



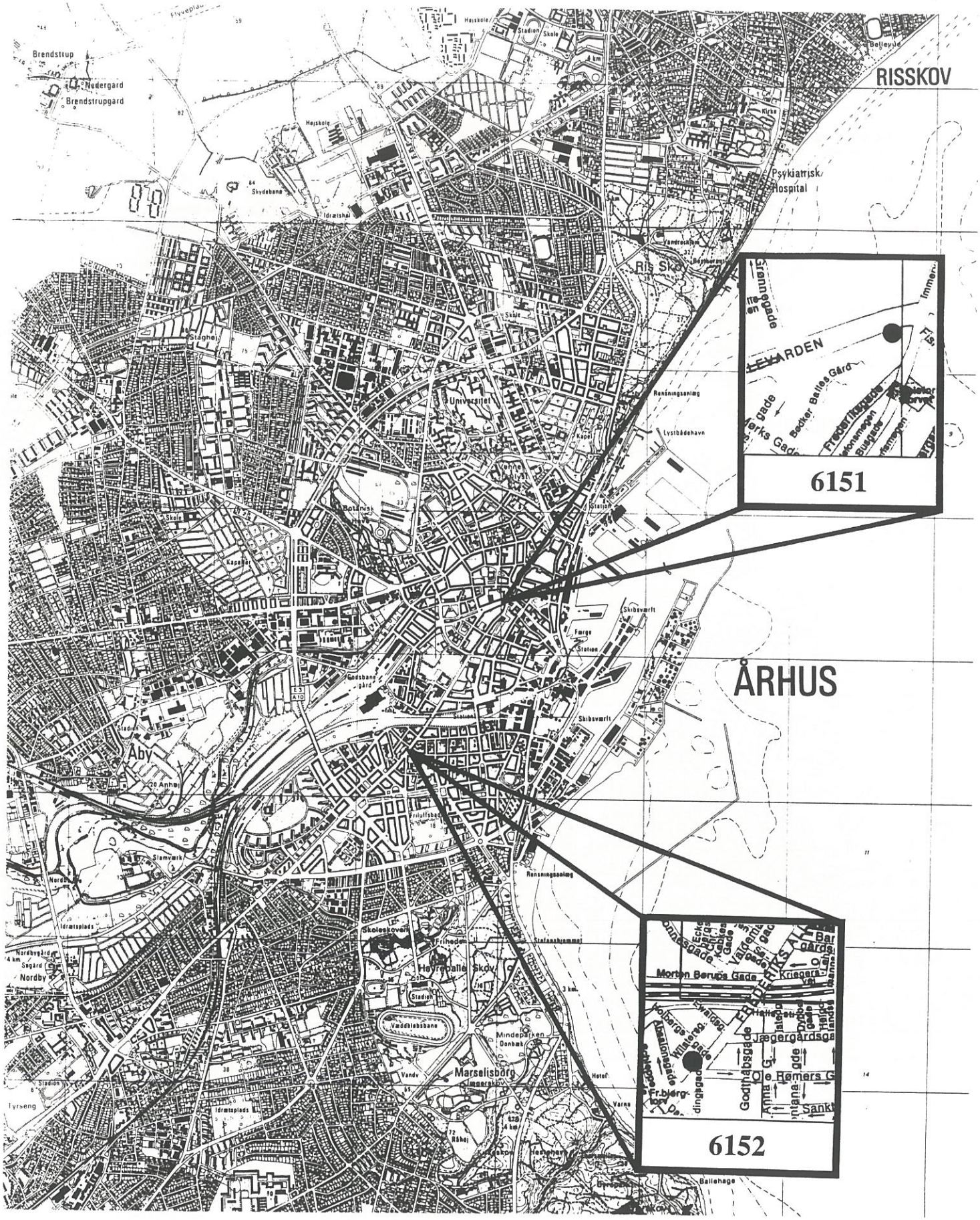
Figur 1.1 Station locations in Copenhagen.



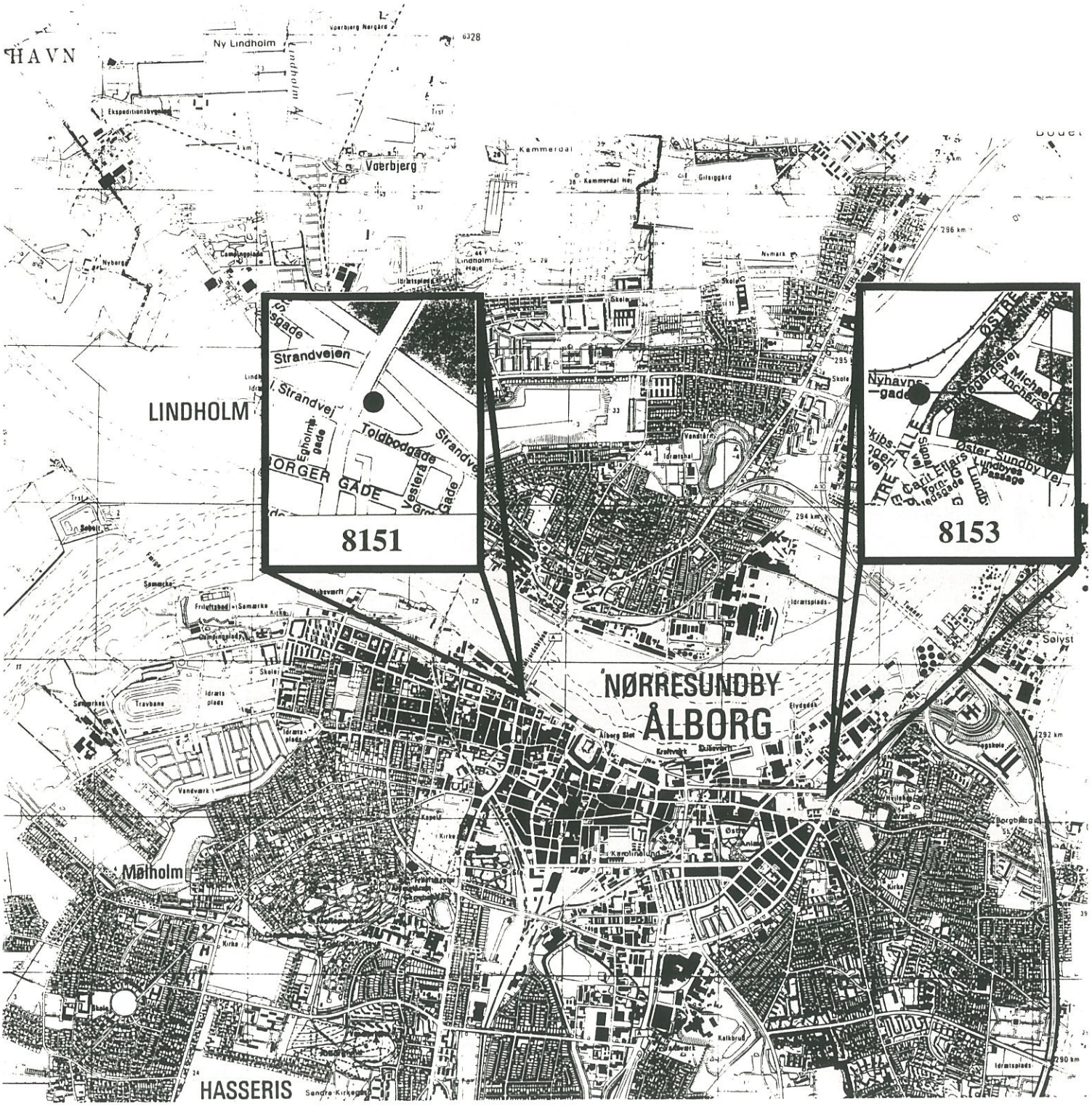
Figur 1.2 Station locations in Fredericia.



Figur 1.3 Station locations in Esbjerg.



Figur 1.4 Station locations in Århus.



Figur 1.5 Station locations in Aalborg.

2 Methods of analysis

NERI

Weighing of particle filters, analysis of SO_2 on KOH-impregnated filters and PIXE-analysis of elements in the TSP are performed at NERI.

TSP

The total amount of dust (TSP) on the exposed filters does normally vary between approximately 0,5 mg and 5 mg. The applied filters are membrane disc filters produced of a mixture of cellulose nitrate and cellulose acetate; the pore size is approximately 1,2 µm. Particles with an aerodynamic diameter down to 1/100 µm are collected with an efficiency of nearly 100% at the actual flow rate. The weight of one filter is approximately 100 mg, which is much more than the weight of the collected dust. A special procedure is applied for that purpose.

Weighing

Before mounting in the filter holder, the filters are stored in an air conditioned room at 23,0±0,5 °C and a relative humidity at 52±2% for at least 48 hours. After weighing they are mounted in the filter holders. After exposure they are dismounted and stored at least 7 days in the air conditioned room before weighing. The weighings are carried out in the air conditioned room. Control weighings are carried out for filters constantly stored in the air conditioned room (*Kemp, 1985*). The accuracy of the weighing is approximately ±50 µg corresponding to ± 1 µg/m^3 at the actual volume of air.

SO2 (impregnated filters)

KOH impregnated cellulose fibre filters are used for collection of SO_2. The filters are impregnated with a 2N solution of KOH including 5% glycerol. Under Danish weather conditions the collection efficiency is close to 100%. After exposure the filter are extracted in demineralized water and the analysis is performed by ionchromatography. By this method it is possible to determine concentrations down to 0.01 µg/m^3. The accuracy is better than 10%. (*Keiding and Hansen*, 1983).

Elements (PIXE)

The concentrations of elements are determined by Proton Induced X-ray Emission Spectroscopy (PIXE) (*Johansen and Campbell, 1988*). By the PIXE analysis the particle filters are exposed to protons at high energy (2.5 MeV). Protons eject electrons from the inner orbits and less bound electrons fill up the holes under emission of X-rays. The energy of the X-rays is characteristic for the elements from which they are emitted and the intensity is a measure of the amount of the element. By analysis of the X-ray spectrum it is possible simultaneously to determine all elements present above the detection limits. However, in practise it is not possible to determine elements with atomic number less than 13 (aluminum). By proton radiation in 2

minutes per filter detection limits down to 0.002 µg/m^3 are obtained. In most cases more than 15 elements are determined (see table 3.10-15).

Quality assurance

All analyses are performed in accordance with normal Good Laboratory Practise (GLP). Additionally, it is possible to control data by comparison of different results. A few methods are described below.

Sulphate

Sulphur in particulates, which is determined by PIXE, will mainly be sulphate formed by oxidation of SO_2 in the atmosphere. Because the reaction time is long compared to the transport time in Denmark, the concentration of sulphur in particulates will be nearly equal all over the country. If large differences are observed at one station compared to all other stations, an error at the station is likely. It is thus possible to detect errors and to date the start of the errors.

NO and lead

A comparison between the concentration of NO and lead at the same station is the basis for another control. The two pollutants are measured by two different and independent methods. At monitoring stations located near busy streets the main source of NO as well as lead is the traffic and by this a very close correlation between the concentration of the two pollutants will be expected (*Palmgren Jensen et al., 1990*). If this correlation does not exist, an error at one of the instruments is most likely.

SO_2

A very obvious control method is to compare simultaneous SO_2-measurements by impregnated filters and the UV-fluorescence monitors.

3 Annual averages, means and percentiles

Limit values

Limit values and guide lines for the air quality in Denmark are given in table 3.1. The limit values are based on the rules in EEC directives (*EEC*, 1980, 1982, 1985). In addition, recommended limit values from WHO are listed in the table.

Table 3.1. Limit values and guide lines (*) for air quality in Denmark. References: (1) (Miljøministeriet, 1986); (2) (EEC, 1982); (3) (Miljøministeriet, 1987); (4) (WHO, 1987).

Pollutant	Averaging time (hours)	Statistical parameter	Concentration (µg/-m^3)	Reference periode	Reference
SO_2	24	Median	80	1/4-31/3	(1)
		98-perc	250		
		median	130	1/10-31/3	
		mean	40-60(*)	one year	(4)
		max.	100-150(*)	-	
TSP	24	mean	150	1/4-31/12	(1)
		95-perc.	300		
Lead		mean	2	1/1-31/12	(2)
NO_2	1	98-perc.	200	1/1-31/12	(3)
		mediam	50(*)		
		98-perc.	135(*)		
		max.	400(*)	-	(4)
	24	max.	150(*)	-	

The data

The reported data are based on measurements from 1991 and earlier. Only values based on more than half of possible results within the given period are used. The yearly data in the tables and plots are calculated for calendar years. Data for tropical years (1. April - 31. March) for SO_2 and TSP are calculated for comparison with the limit values (Miljøministeriet, 1986). The difference between the two sets of results are almost negliglible. In addition to the tables most of the values are shown in histograms. The sequence of the columns follows the station numbers.

SO_2

The 50- and the 98-percentiles of the 24 hours measurements of SO_2 for the years 1987-91 are shown in the tables 3.2 and 3.3. In figure 3.1 the corresponding data are shown for 1991. The values are low

compared to the limit values, 10-20% of the limit values, 80 and 250 $\mu g/m^3$ respectively. Only small deviations between the stations are observed. Generally the levels in Copenhagen are higher than in the other cities.

Table 3.2. 50-percentiles of SO_2 measure in the years 1987-91 ($\mu g/m^3$). Winter is defined as the months October to March.

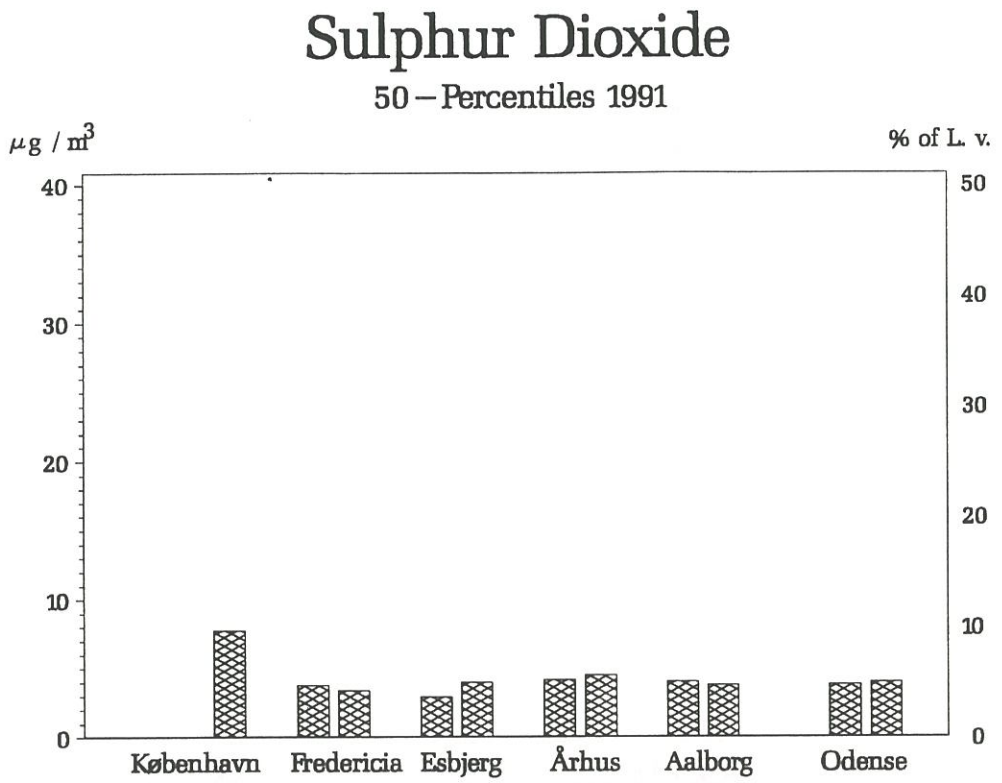
	Year					Winter			
City/station number	87	88	89	90	91	87/88	88/89	89/90	90/91
København/1255	-	17.4	16.5	12.9	-	19.5	19.6	18.7	-
København/1256	-	17.2	15.8	12.6	-	17.7	17.9	19.0	-
København/1257	-	20.6	19.7	16.8	15.6	20.6	22.2	23.0	19.4
København/1258	-	-	-	-	12.7*	-	-	-	16.2*
Ll. Valby/2090	-	-	-	-	4.5*	-	-	-	5.7*
Fredericia/5152	7.0	7.1	7.8	8.5	7.6	7.6	10.1	11.8	9.9
Fredericia/5155	-	9.5	7.5	7.9	6.8	12.7	9.9	9.8	8.9
Esbjerg/5652	-	4.1	4.9	5.6	5.9	-	5.8	8.8	8.3
Esbjerg/5655	-	7.8	8.3	7.9	8.1	-	9.0	10.1	10.6
Århus/6151	-	9.4	9.2	8.1	8.4	-	9.4	10.0	10.1
Århus/6152	-	11.5	10.8	9.8	9.0	-	11.7	12.2	10.4
Aalborg/8151	11.5	12.9	9.7	9.1	8.1	14.1	10.0	10.3	8.5
Aalborg/8153	11.6	10.8	9.3	8.2	7.6	12.5	10.7	9.3	7.8
Odense/9151	6.4	-	-	-	-	-	-	-	-
Odense/9154	7.9	9.0	8.3	6.8	7.7	10.7	7.3	10.5	9.0
Odense/9155	-	9.3	8.4	6.7	8.0	-	9.0	9.1	9.1
Limit values			80				130		

* Only part of the year.

Table 3.3. 98-percentiles of SO_2 measured in the years 1987-91 ($\mu g/m^3$).

City/station number	87	88	89	90	91
København/1255	-	57.5	66.5	51.3	-
København/1256	-	58.0	64.2	52.2	-
København/1257	-	62.1	63.2	56.9	59.0
København/1258	-	-	-	-	53.0*
Ll. Valby/2090	-	-	-	-	36.2*
Fredericia/5152	31.7	33.4	42.3	56.5	41.6
Fredericia/5155	-	51.1	48.1	43.4	41.0
Esbjerg/5652	-	40.3	48.0	119.6	57.7
Esbjerg/5655	-	29.7	40.4	60.9	43.4
Århus/6151	-	39.0	38.9	35.2	54.9
Århus/6152	-	40.7	34.5	35.6	29.6
Aalborg/8151	38.0	39.0	35.2	37.7	51.6
Aalborg/8153	35.1	34.9	28.6	38.0	42.0
Odense/9151	37.3	-	-	-	-
Odense/9154	37.1	48.1	41.8	46.7	45.1
Odense/9155	-	42.6	41.4	37.3	49.5
Limit value			250		

* Only part of the year.



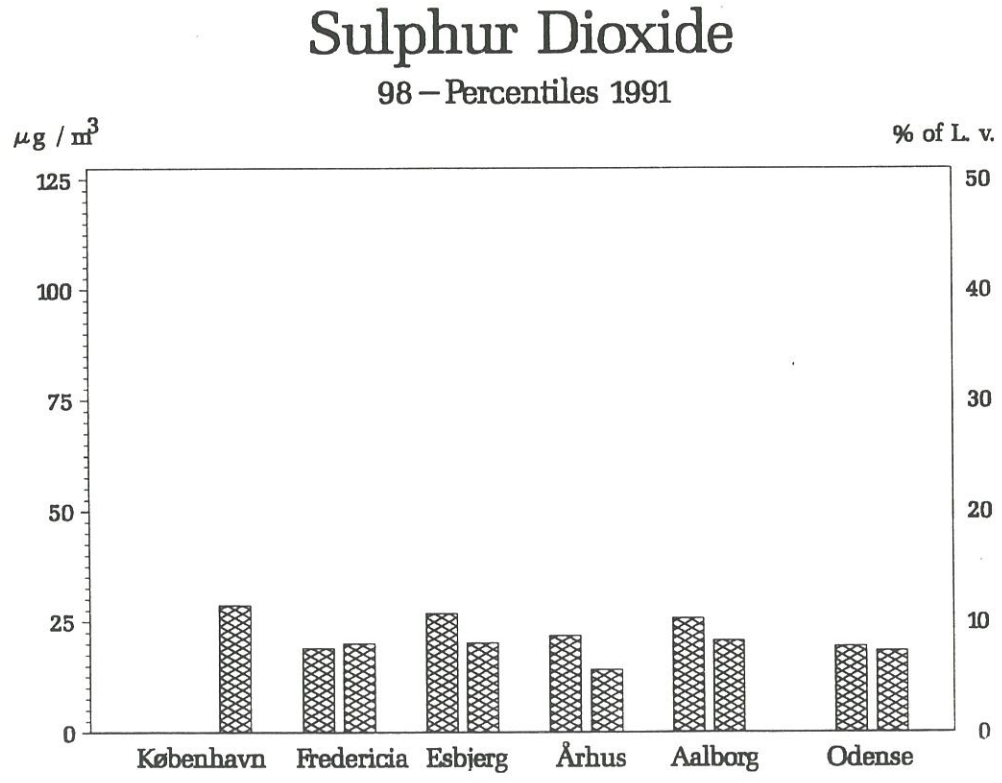


Figure 3.1 SO_2 50- and 98 percentiles in 1991 based on 24 hours measurements. The columns from left to right represent the stations 1257, 5152, 5155, 5652, 5655, 6151, 6152, 8151, 8153, 9154 and 9155. The right y-scale is relative to the limit value.

Particulates TSP

The annual averages and the 95-percentiles of the 24 hours measurements of TSP for the years 1987-91 are shown in the tables 3.4 and 3.5. In figure 3.2 the corresponding data are shown for 1991. The measurements are based on weighing of the filters before and after exposure. The levels are approximately 50% of the limit values 150 and 300 µg/m^3 respectively.

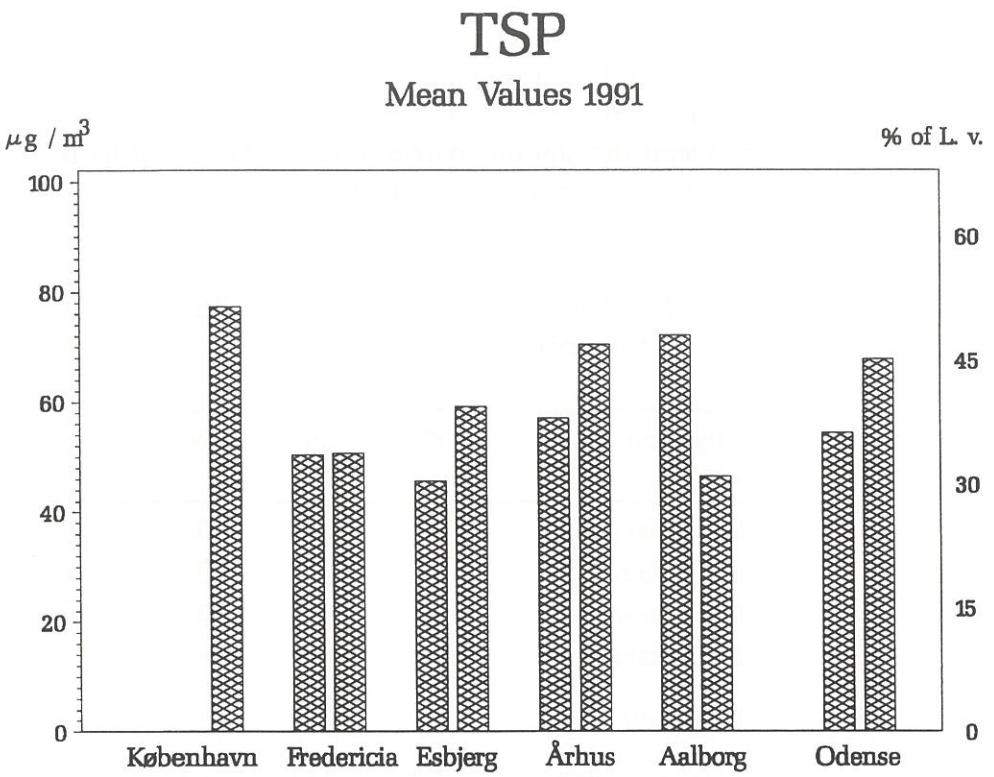
Table 3.4. Annual averages of TSP measured in the years 1987-91 (µg/m^3).

City/station number	87	88	89	90	91
København/1255	-	66.7	63.9	59.8	-
København/1256	-	77.6	74.7	64.4	-
København/1257	-	93.0	85.3	73.6	77.4
København/1258	-	-	-	-	62.6*
Ll. Valby/2090	-	-	-	-	-
Fredericia/5152	54.2	51.1	58.9	56.3	50.4
Fredericia/5155	-	66.8	56.2	55.1	50.8
Esbjerg/5652	-	41.2	45.4	46.0	45.8
Esbjerg/5655	-	60.2	59.0	59.5	59.2
Århus/6151	-	57.1	62.1	53.0	57.1
Århus/6152	-	66.0	71.2	63.0	70.4
Aalborg/8151	79.6	77.0	78.0	70.4	71.5
Aalborg/8153	54.9	47.4	47.7	44.9	46.6
Odense/9151	58.4	-	-	-	-
Odense/9154	66.3	56.6	57.6	54.4	53.9
Odense/9155	-	75.6	68.3	60.6	64.9
Limit value			150		

Table 3.5. 95-percentiles of TSP measured in the years 1987-91 ($\mu g/m^3$).

City/station number	87	88	89	90	91
København/1255	-	124	125	124	-
København/1256	-	141	150	124	-
København/1257	-	185	171	156	156
København/1258	-	-	-	-	113*
Ll. Valby/2090	-	-	-	-	-
Fredericia/5152	110	108	116	120	106
Fredericia/5155	-	154	109	115	104
Esbjerg/5652	-	78	86	92	88
Esbjerg/5655	-	110	106	116	110
Århus/6151	-	114	130	120	117
Århus/6152	-	132	144	124	126
Aalborg/8151	165	155	155	148	155
Aalborg/8153	116	92	95	100	92
Odense/9151	138	-	-	-	-
Odense/9154	140	125	117	119	104
Odense/9155	-	153	145	142	141
Limit values			300		

* only part of the year



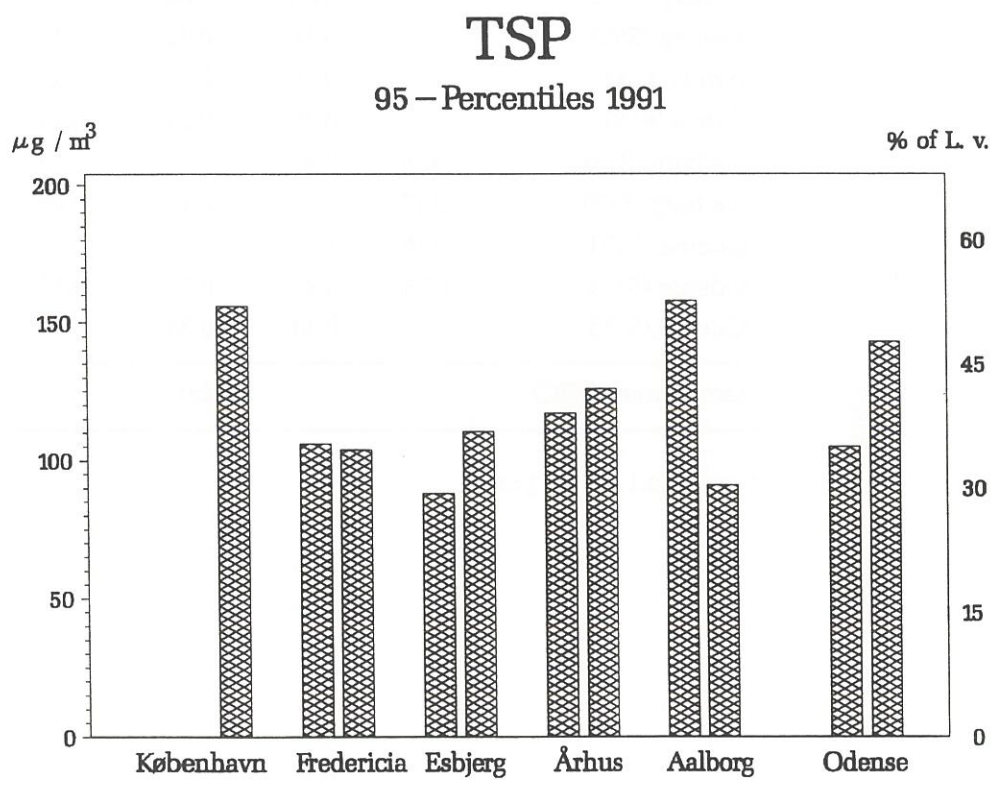


Figure 3.2 Annual averages and 95-percentiles based on 24 hours measurements of TSP in 1991. The columns represent the stations 1257, 5152, 5155, 5652, 5655, 6151, 6152, 8151, 8153, 9154 and 9155. The right y-scale is relative to the limit value.

Lead

The annual averages of the lead concentration in TSP based on PIXE-analysis for the years 1987-91 are shown in the table 3.6. In figure 3.3 the corresponding data are shown for 1991. The levels are much lower than the EEC limit value on 2 $\mu g/m^3$. The deviations between the stations correspond to the locations in relation to the streets and the traffic intensities at the streets.

Table 3.6. Annual averages of lead in TSP measured in the years 1987-91 ($\mu g/m^3$).

City/station number	87	88	89	90	91
København/1255	-	0.19	0.17	0.13	-
København/1256	-	0.34	0.31	0.21	-
København/1257	-	0.40	0.35	0.25	0.24
København/1258	-	-	-	-	0.28*
Ll. Valby/2090	-	-	-	-	0.03*
Fredericia/5152	0.05	0.04	0.04	0.04	0.03
Fredericia/5155	-	0.12	0.09	0.08	0.06
Esbjerg/5652	-	0.06	0.06	0.05	0.03
Esbjerg/5655	-	0.11	0.12	0.10	0.08
Århus/6151	-	0.18	0.17	0.12	0.11
Århus/6152	-	0.20	0.21	0.15	0.07
Aalborg/8151	0.38	0.37	0.34	0.29	0.26
Aalborg/8153	0.15	0.13	0.11	0.09	0.09
Odense/9151	0.16	-	-	-	-
Odense/9154	0.26	0.22	0.22	0.17	0.15
Odense/9155	-	0.30	0.24	0.15	0.17
Limit value (EEC)			2.0		

* only part of the year

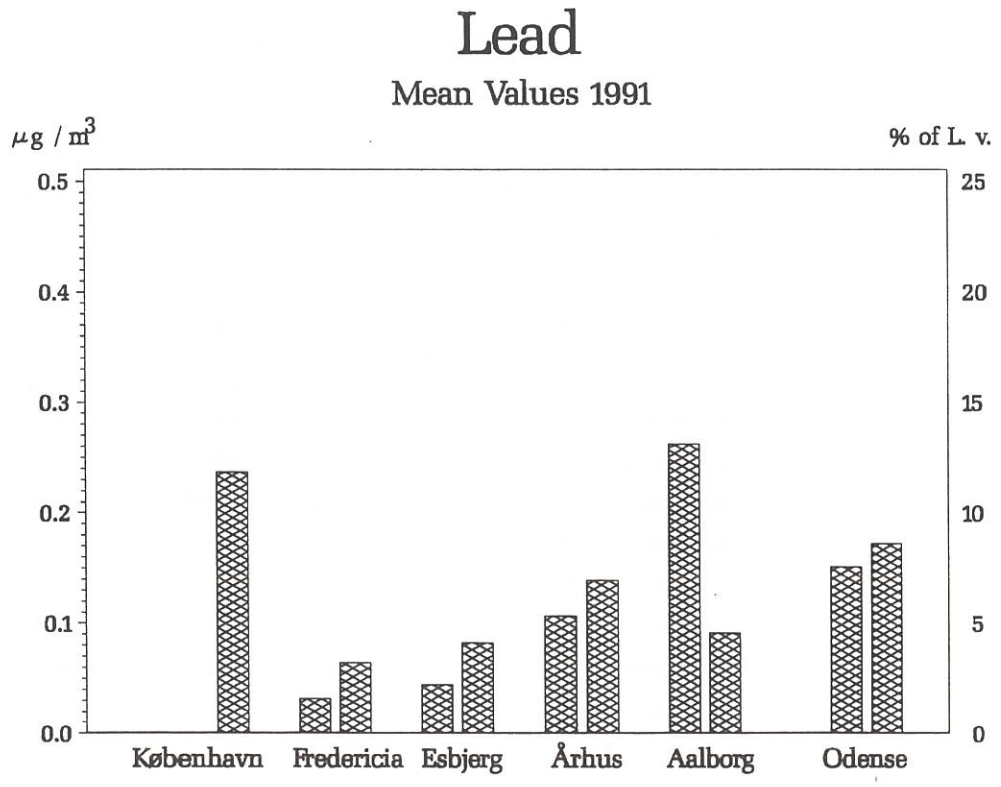


Figure 3.3 Annual averages for lead based on measurements in 1991. The columns represents from left to right the stations 1257, 5152, 5155, 5652, 5655, 6151, 6152, 8151, 8153, 9154 and 9155. The right y-scale is relative to the limit value.

Nitrogen oxides

The 1 hour mean values for NO and NO_2 are based on ½ hour measurements. The 50- and 98-percentiles of the 1 hour averages of NO and NO_2 for the years 1987-91 are shown in the tables 3.7 - 3.10. The corresponding data are shown for 1991 in figures 3.4 - 3.5. No Danish limit values exist for NO. For NO_2 the right axis is the percentage of the limit value. The guide lines are shown as horizontal lines in the figure. The limit value for the 98-percentile of NO_2 on 200 µg/m^3 was not exceeded at any station in 1991. The guide lines were not exceeded, but the levels were close to the guide line values in Copenhagen.

Table 3.7. The 50-percentiles for NO measured in the year 1987-91 (µg/m3).

City/station number	87	88	89	90	91
København/1255	-	37.4	38.0	34.0	-
København/1256	-	73.4	82.8	84.6	-
København/1257	-	75.8	76.9	68.5	78.4
København/1258	-	-	-	-	44.6*
Ll. Valby/2090	-	-	-	-	1.7
Fredericia/5155	-	9.9	7.6	10.3	7.1
Esbjerg/5655	-	27.4	33.1	30.6	35.3
Århus/6155	-	28.4	26.8	26.6	28.8
Aalborg/8151	55.7	63.2	66.8	64.7	65.5
Odense/9151	7.8	-	-	-	-
Odense/9155	-	33.8	31.1	24.2	35.8

Table 3.8. The 98-percentiles for NO measured in the years 1987-91 (µg/m3).

City/station number	87	88	89	90	91
København/1255	-	338.9	330.7	345.7	-
København/1256	-	434.7	442.5	420.6	-
København/1257	-	503.2	519.9	545.3	527.2
København/1258	-	-	-	-	240.3*
Ll. Valby/2090	-	-	-	-	45.3
Fredericia/5155	-	128.4	128.7	133.9	108.0
Esbjerg/5655	-	119.0	133.9	125.4	153.7
Århus/6155	-	306.0	287.5	269.7	277.9
Aalborg/8151	400.0	367.9	434.6	452.1	445.6
Odense/9151	118.7	-	-	-	-
Odense/9155	-	360.1	397.0	395.5	404.6

* only part of the year

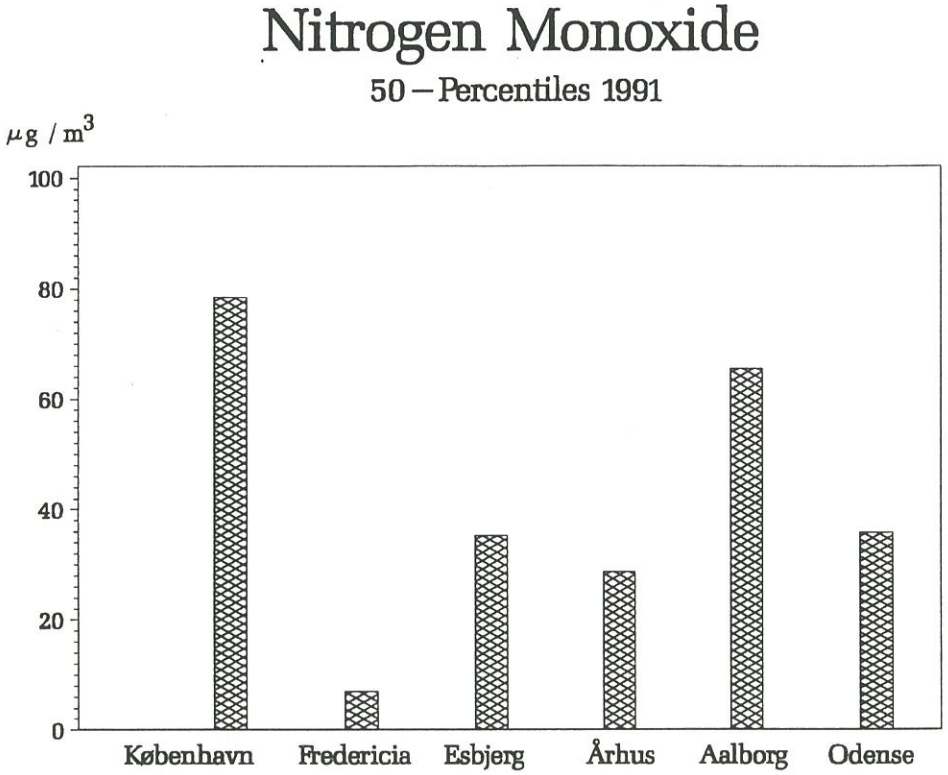
Table 3.9. The 50-percentiles for NO_2 measured in the years 1987-91 ($\mu g/m^3$).

City/station number	87	88	89	90	91
København/1255	-	48.1	49.3	45.3	-
København/1256	-	56.7	53.1	41.4	-
København/1257	-	56.0	57.4	51.5	52.4
København/1258	-	-	-	-	45.8*
Ll. Valby/2090	-	-	-	-	11.4
Fredericia/5155	-	24.2	25.4	22.4	23.4
Esbjerg/5655	-	32.1	37.6	35.3	39.5
Århus/6155	-	40.1	37.0	37.2	39.8
Aalborg/8151	35.2	45.9	40.3	34.7	39.0
Odense/9151	28.0	-	-	-	-
Odense/9155	-	42.4	36.2	33.1	40.2
Guide line			(50)		

Table 3.10. The 98-percentiles for NO_2 measured in the years 1987-91 ($\mu g/m^3$).

City/station number	87	88	89	90	91
København/1255	-	99.6	108.3	100.7	-
København/1256	-	151.4	113.0	104.9	-
København/1257	-	157.8	149.3	133.1	110.6
København/1258	-	-	-	-	90.3*
Ll. Valby/2090	-	-	-	-	55.2
Fredericia/5155	-	66.3	73.2	65.4	67.1
Esbjerg/5655	-	74.3	82.0	81.0	82.8
Århus/6155	-	98.2	93.7	96.8	96.3
Aalborg/8151	97.5	124.4	118.3	96.2	95.8
Odense/9151	87.5	-	-	-	-
Odense/9155	-	130.3	104.7	89.8	99.4
Limit value(Guide line)			200(135)		

* only part of the year



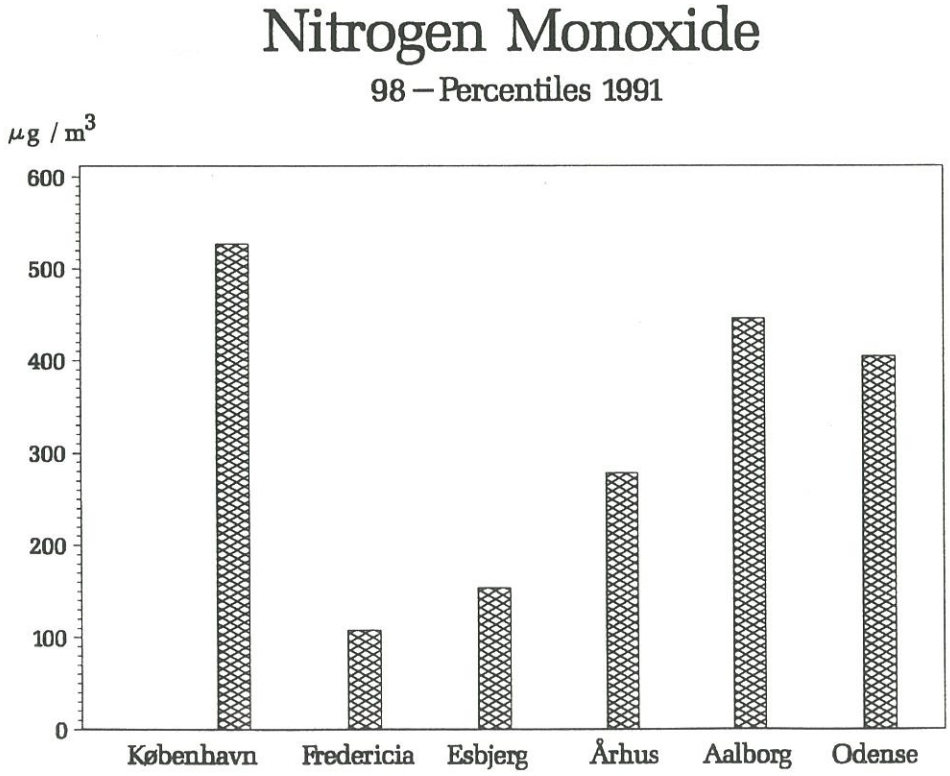


Figure 3.4 The 50- and 98-percentiles for nitrogen monoxide for 1991 based on 1 hour mean values. The columns from left to right represent the stations 1257, 5155, 5655, 6151, 8151 and 9155.

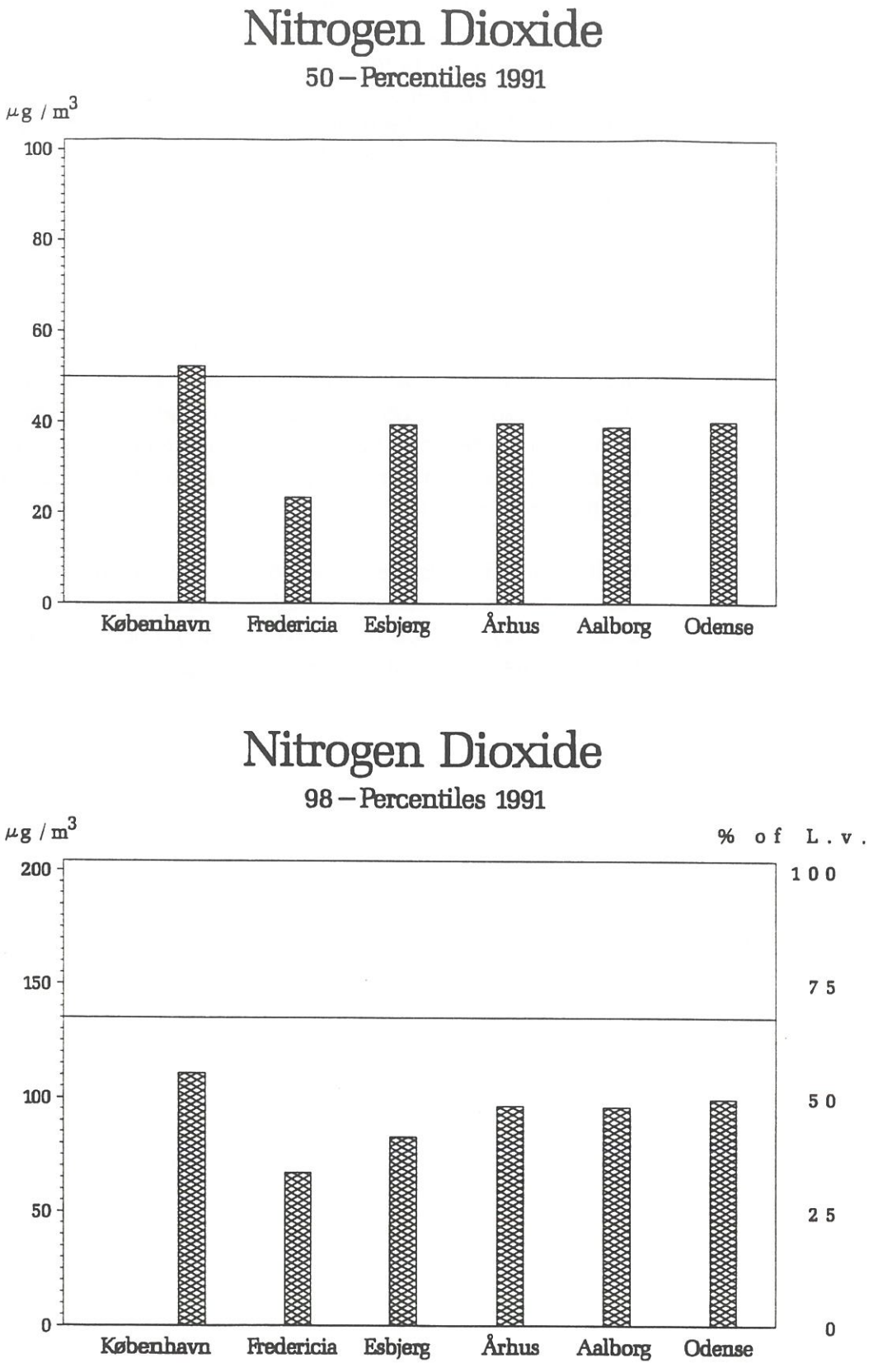


Figure 3.5 The 50- and 98-percentiles for nitrogen dioxide for 1991 based on 1 hour mean values. The columns from left to right represent the stations 1257, 5155, 5655, 6151, 8151 and 9155. The horizontal lines represent the guide values (cf. table 3.1). The right y-scale is relative to the limit value.

Annual means of 24 hour measurements

Annual averages in 1991 of all pollutants measured as 24 hours means are shown in the tables 3.11-16. For the elements the units are ng/m^3. The concentration of many elements are below the detection limit of the PIXE-method. If more than 10% of the measurements are below the detection limit the annual averages are calculated under the assumption of a logarithmic normal distribution fitted to the available data.

Indicator elements

Many elements can be used as indicators of specific source types, which contributes to the air pollution. Sulphur in TSP originates from oxidized SO_2 emitted in Europe and by this it represents long range transport. Vanadium and nickel originate primarily from heavy fuel oil. Silicon, calcium, titanium and iron originate from suspended soil dust. Most of the chlorine originates from the sea, i.e. Kattegat and the North Sea. However, some chlorine is resuspended from the streets due to salting of the kerb. Bromium and lead are emitted from vehicles.

WHO guidelines

WHO guideline values based on public health effects exist for vanadium, manganese and cadmium and they are 1 $\mu g/m^3$, 1 $\mu g/m^3$ and 10-20 ng/m^3 respectively (*WHO, 1987*). The measured values are for all elements at least one order of magnitude below these values.

Table 3.11 Annual averages for 1991 of SO_2, TSP and elements (SO_2 and TSP in µg/m³, the elements in ng/m³) in Copenhagen. N_{tot} is the total number of measurement results. NO is the total number of measurement results above the detection limits. * indicates annual averages calculated by assumption of a logarithmic normal distribution.

	1257			1258			2090		
	NO	Mean	SD	NO	Mean	SD	NO	Mean	SD
SO_2	344	18.2	13.0	275	15.4	9.3	190	6.2	6.4
Tsp	345	77.4	38.2	168	62.6	24.8			
Al	345	738	560	277	593.0	404	190	209.0	229.0
Si	344	1841	1512	277	1477.0	1114.0	171	420.0	583.0
S	345	2311	1527.0	277	2132.0	1387.0	190	1809.0	1484.0
Cl	324	2474	3574.0	251	1675.0	2054.0	155	989.0	4630.0
K	345	414	398.0	277	368.0	225.0	190	244.0	185.0
Ca	345	1627	1225.0	277	1313.0	856.0	190	258.0	341.0
Ti	344	100	70.0	277	82.0	49.1	163	17.2	18.2
V	292	12.2	6.8	228	10.4	7.2	162	6.5	4.4
Cr	292	6.0	3.7	225	3.8	2.6	36	1.0	3.3
Mn	345	25.1	17.3	277	20.3	12.6	185	7.0	6.9
Fe	345	1512	995.0	277	1065.0	581.0	190	190.0	198.0
Ni	337	6.1	3.3	276	6.2	3.6	188	2.7	2.1
Cu	345	55.9	33.9	277	32.9	16.4	186	3.7	3.5
Zn	345	127	81.0	277	102.0	59.0	190	29.4	27.2
Br	345	45.6	27.9	277	23.2	12.8	190	7.0	4.0
Sr	336	8.7	8.3	271	6.8	3.6	154	2.0	1.4
Mo	128	2.3	2.2*	85	1.7	1.1*	27	0.6	0.5
Cd	14	0.9	1.2*	9	0.6*	1.5*	9	1.1	0.7
Sn	44	3.3	3.5*	28	2.3*	2.5*	16	1.0	1.6
Sb	215	12.4	9.0*	134	7.8*	4.6*	17	1.5	2.6
Ba	308	57.0	44.8	246	38.2	51.3	15	3.5	4.2
Pb	345	237.0	139.0	277	121.0	65.0	190	24.8	22.4
Ntot	345			277			190		

Table 3.12 Annual averages for 1991 of SO_2, TSP and elements (SO_2 and TSP in $\mu g/m^3$, the elements in ng/m^3) in Fredericia. N_{tot} is the total number of measurement results. NO is the total number of measurement results above the detection limits. * indicates annual averages calculated by assumption of a logarithmic normal distribution.

	5155			5152		
	NO	Mean	SD	NO	Mean	SD
SO_2	358	10.2	10.0	342	10.1	9.9
Tsp	341	51.0	23.9	328	50.8	24.5
Al	358	436.0	424.0	343	315.0	310.0
Si	353	1021.0	1131.0	341	692.0	705.0
S	358	2058.0	1468.0	343	2105.0	1500.0
Cl	341	1890.0	2129.0	327	2237.0	2499.0
K	358	335.0	286.0	343	608.0	838.0
Ca	358	483.0	429.0	343	635.0	901.0
Ti	352	48.6	41.1	339	28.5	28.5
V	271	8.6	9.4*	287	11.5	11.4
Cr	205	2.5	2.7*	164	2.1	2.8
Mn	357	18.4	13.8	343	12.2	8.9
Fe	358	696.0	607.0	343	406.0	334.0
Ni	353	4.6	3.8	342	6.5	5.8
Cu	357	15.9	10.6	341	5.9	6.8
Zn	358	58.7	48.9	343	43.4	44.2
Br	358	13.7	7.7	343	9.6	4.3
Sr	342	5.0	5.7	325	17.5	55.7
Mo	91	1.2	0.9*	58	0.8	0.8*
Cd	16	1.1	0.9*	16	0.9	0.9*
Sn	40	2.0	5.6*	34	2.3	1.6*
Sb	70	4.2	2.8*	25	2.2	1.8*
Ba	198	16.5	14.6*	61	6.8	7.7*
Pb	358	64.0	42.5	342	31.2	24.9
Ntot	358			343		

Table 3.13 Annual averages for 1991 of SO_2, TSP and elements (SO_2 and TSP in $\mu g/m^3$, the elements in ng/m^3) in Esbjerg. N_{tot} is the total number of measurement results. NO is the total number of measurement results above the detection limits. * indicates annual averages calculated by assumption of a logarithmic normal distribution.

	5655			5652		
	NO	Mean	SD	NO	Mean	SD
SO_2	361	11.1	10.1	348	11.3	15.0
Tsp	360	59.2	24.9	346	45.8	21.2
Al	361	428.0	401.0	348	298.0	266.0
Si	355	993.0	1050.0	337	606.0	641.0
S	361	2246.0	1508.0	348	2046.0	1523.0
Cl	347	4288.0	4583.0	327	3289.0	3718.0
K	361	291.0	178.0	348	235.0	166.0
Ca	361	539.0	443.0	348	293.0	204.0
Ti	358	48.5	36.8	344	29.8	23.3
V	311	12.1	9.5	287	7.3	7.6
Cr	200	2.2	2.1	105	1.4	1.9
Mn	360	13.6	10.2	345	9.5	7.8
Fe	361	598.0	375.0	348	334.0	229.0
Ni	355	4.7	3.1	344	3.2	3.3
Cu	361	15.9	7.2	348	7.0	4.0
Zn	361	60.0	45.1	348	40.0	38.3
Br	361	20.5	9.3	347	12.9	7.6
Sr	344	5.5	4.2	322	3.8	2.0
Mo	92	1.3	0.9*	59	0.8	0.9*
Cd	12	0.7	1.1*	14	1.0	0.8*
Sn	26	1.9	1.8*	29	1.7	1.6*
Sb	76	4.4	2.7*	42	3.1	1.7*
Ba	247	23.4	28.7*	116	10.2	8.3*
Pb	361	82.4	41.9	348	44.2	30.4
Ntot	361			348		

Table 3.14 Annual averages for 1991 of SO_2, TSP and elements (SO_2 and TSP in µg/m³, the elements in ng/m³) in Århus. N_{tot} is the total number of measurement results. NO is the total number of measurement results above the detection limits. * indicates annual averages calculated by assumption of a logarithmic normal distribution.

	6151			6152		
	NO	Mean	SD	NO	Mean	SD
SO_2	336	11.7	11.2	293	11.0	6.9
Tsp	339	57.0	30.4	292	70.4	28.4
Al	339	481.0	465.0	294	660.0	444.0
Si	337	1109.0	1229.0	294	1662.0	1238.0
S	339	2030.0	1439.0	294	2202.0	1421.0
Cl	332	2218.0	2382.0	277	2314.0	2820.0
K	339	334.0	250.0	294	391.0	373.0
Ca	339	680.0	825.0	294	987.0	735.0
Ti	337	62.2	55.3	294	93.6	57.3
V	298	12.1	10.2	210	10.0	11.2
Cr	231	3.3	3.4	233	4.0	3.2
Mn	338	20.4	28.6	294	25.3	15.4
Fe	339	851.0	646.0	294	1306.0	772.0
Ni	339	7.0	4.6	290	6.5	5.2
Cu	339	26.4	17.8	294	37.8	7.8
Zn	339	67.2	53.2	294	89.3	48.7
Br	339	21.5	15.1	294	26.8	13.9
Sr	323	5.3	4.1	287	6.2	3.2
Mo	105	1.3	1.2*	97	1.8	1.7*
Cd	13	0.7	1.0*	9	0.6	2.0*
Sn	38	1.8	2.5	24	2.1	2.5*
Sb	130	6.1	4.8	150	8.5	5.5*
Ba	247	26.7	22.7	270	37.8	22.4
Pb	339	106.0	73.0	294	139.0	66.0
Ntot	339			294		

Table 3.15 Annual averages for 1991 of SO_2, TSP and elements (SO_2 and TSP in $\mu g/m^3$, the elements in ng/m^3) in Aalborg. N_{tot} is the total number of measurement results. NO is the total number of measurement results above the detection limits. * indicates annual averages calculated by assumption of a logarithmic normal distribution.

	8151			8153		
	NO	Mean	SD	NO	Mean	SD
SO_2	348	12.0	13.5	358	10.8	10.4
Tsp	351	71.8	39.0	358	46.5	22.0
Al	352	596.0	605.0	358	339.0	304.0
Si	352	1551.0	1887.0	355	702.0	714.0
S	353	2102.0	1500.0	358	1974.0	1404.0
Cl	345	3369.0	3477.0	358	2432.0	2664.0
K	353	390.0	525.0	336	290.0	208.0
Ca	353	1086.0	1218.0	358	612.0	587.0
Ti	351	69.7	65.1	358	37.8	31.6
V	261	7.1	6.9	355	7.3	4.4
Cr	263	3.9	4.6	313	2.0	2.1*
Mn	353	19.5	15.4	162	14.0	12.7
Fe	353	1145.0	876.0	357	498.0	441.0
Ni	348	4.4	3.1	358	3.7	2.4
Cu	353	38.8	24.5	356	13.4	12.7
Zn	353	103.0	91.0	358	50.6	60.5
Br	353	49.2	30.9	358	19.6	17.6
Sr	338	7.9	6.3	358	4.9	3.5
Mo	110	1.7	1.5*	341	1.2	0.9*
Cd	14	0.6	1.5*	100	0.5	0.8*
Sn	23	2.1	2.2*	6	1.1	1.6*
Sb	139	7.5	5.4*	19	3.3	2.7*
Ba	308	40.6	28.8	55	17.8	17.1*
Pb	353	263.0	163.0	205	90.8	89.8
Ntot	353			358		

Table 3.16 Annual averages for 1991 of SO_2, TSP and elements (SO_2 and TSP in $\mu g/m^3$, the elements in ng/m^3) in Odense. N_{tot} is the total number of measurement results. NO is the total number of measurement results above the detection limits. * indicates annual averages calculated by assumption of a logarithmic normal distribution.

	9155			9154		
	NO	Mean	SD	NO	Mean	SD
SO_2	356	11.0	10.4	333	9.9	9.5
Tsp	356	66.7	41.9	335	53.7	27.4
Al	357	641.0	678.0	335	440.0	403.0
Si	356	1630.0	2080.0	332	969.0	985.0
S	357	2175.0	1599.0	335	2123.0	1554.0
Cl	345	2731.0	6473.0	325	2016.0	3098.0
K	357	371.0	316.0	335	298.0	199.0
Ca	357	1085.0	1319.0	335	605.0	534.0
Ti	357	91.9	94.8	332	57.2	55.8
V	229	7.4	7.8*	253	7.3	7.2*
Cr	265	4.1	4.4*	233	2.9	2.9*
Mn	357	31.1	43.1	335	18.8	14.4
Fe	357	1127.0	1141.0	335	693.0	551.0
Ni	351	4.2	3.0	333	3.8	2.7
Cu	357	33.9	27.7	335	21.0	14.8
Zn	357	132.0	247.0	335	68.5	53.5
Br	356	32.6	28.2	334	27.6	21.0
Sr	334	6.5	5.4	317	4.6	3.1
Mo	128	1.7	2.1*	112	1.4	1.2*
Cd	10	0.6	0.8*	13	0.9	0.9*
Sn	54	2.6	3.6*	24	1.5	1.7*
Sb	149	8.4	7.6*	100	4.9	3.8*
Ba	275	36.6	40.4*	236	23.1	18.8*
Pb	357	168.0	143.0	335	149.0	108.8
Ntot	357			335		

4 The Trends

The data

The following time series are based on data from the period 1982-1991. The monthly averages, plotted as +, are calculated by the measured 24 hours values. smoothed curves representing moving averages, which illustrates a possible seasonal variations are drawn in the figures. In addition, 1. or 2. order polynomial regression curves are drawn, representing the concentration trends of the different pollutants at all stations. The regression curves are only drawn if more than 3 years of data are available.

Lead

A very clear decreasing trend is observed at all stations with several years of data available (figures 4.1-7). The decrease is closely correlated to the lead content in petrol. The apparent increase in Odense was due to movement of the monitoring site (9151 to 9155) to a street with more intense traffic.

TSP

Increases as well as decreases were observed for TSP at different stations (figure 4.8-14). The trend seem to be dependant on the local conditions, but no clear picture were observed and generally the level was unchanged.

SO_2

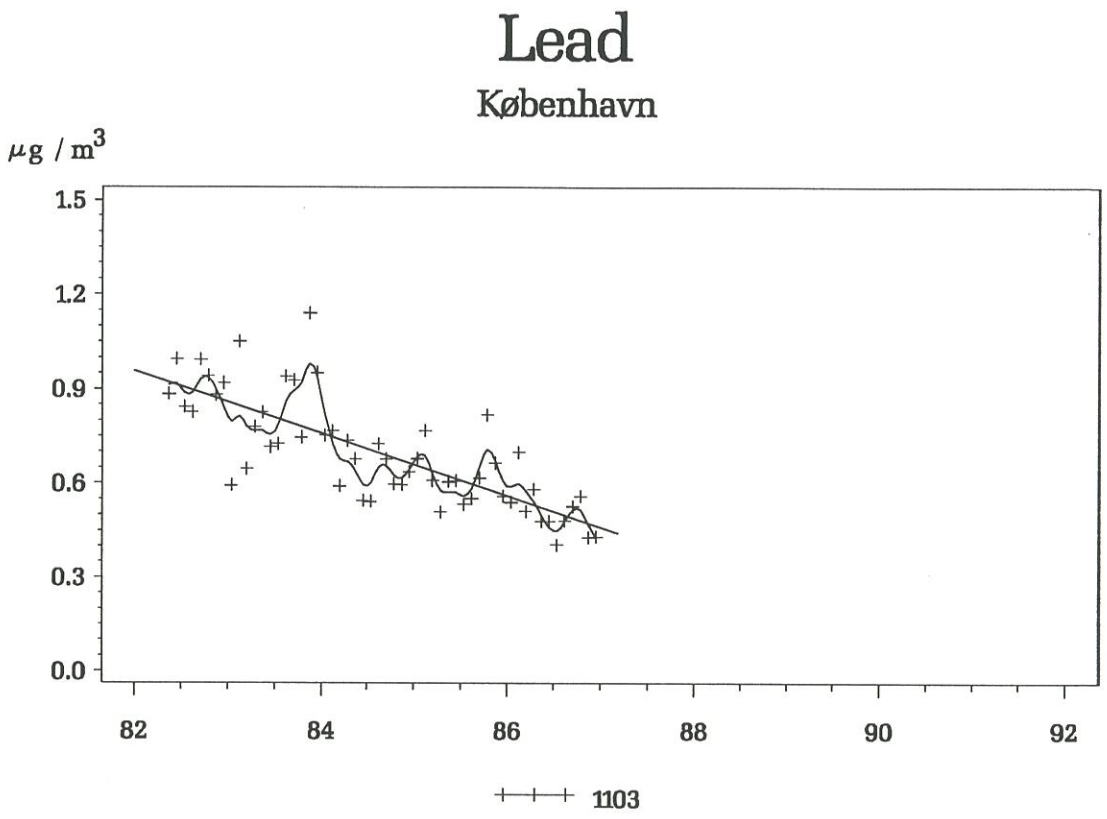
The SO_2 concentration showed very clear seasonal variations with higher levels in the winter seasons than in the summer seasons until 1988. The winters 1989-1990 were very mild and the seasonal variations were smaller. The SO_2 level was again a little higher in the winter 1991 at several stations (figure 4.15-21). The SO_2 concentration showed a clear decrease during 1986 when a reduction of sulphur content in oil products was introduced by law.

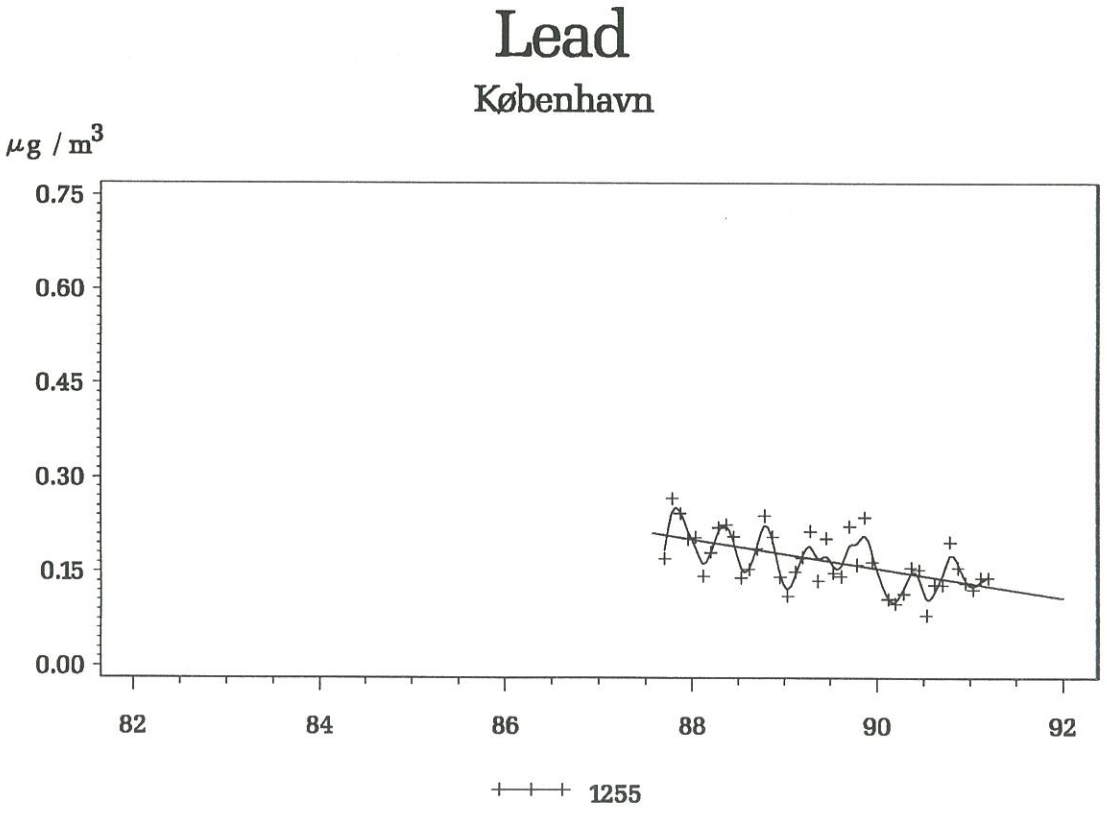
Sulphur (in TSP)

No clear trend was observed for sulphur (figure 4.22-28). Sulphur originates mainly from our neighbour countries to the east, south and west. It is important to observe that the trends of the sulphur concentration does not follow the decreasing trends of the SO_2 concentration. This indicates that our neighbour countries have not reduced the SO_2 emission correspondingly.

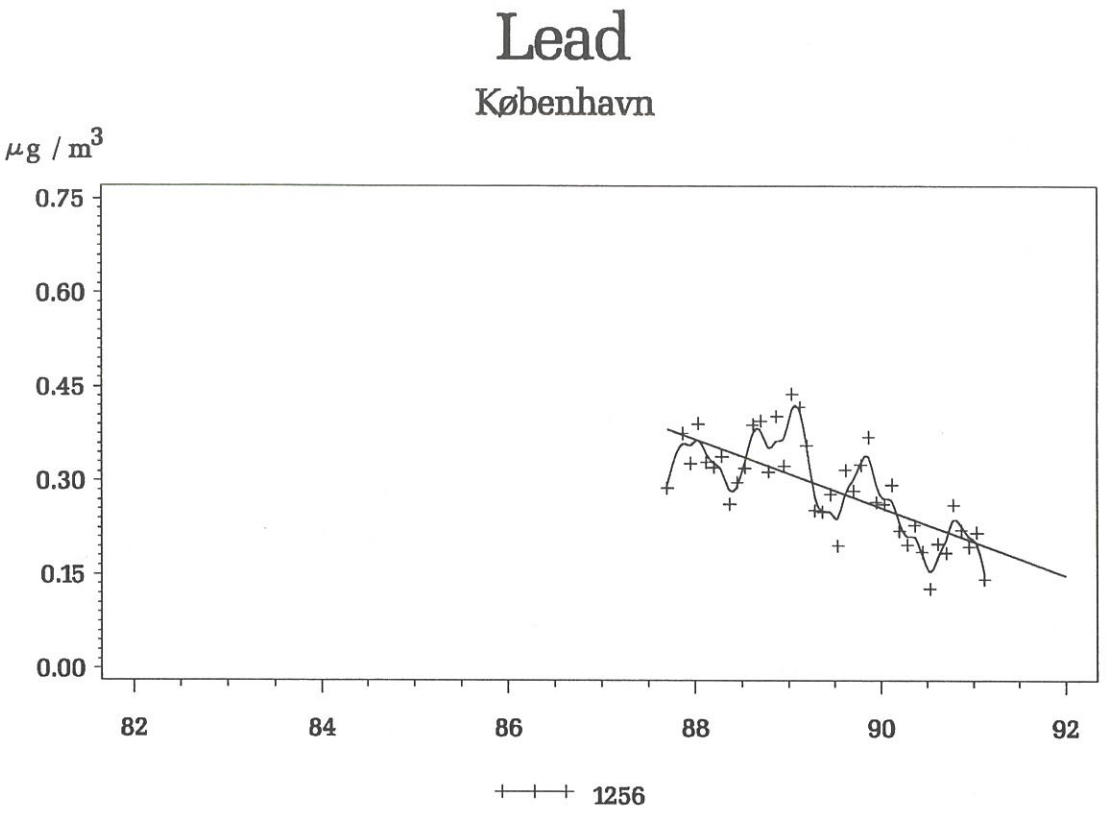
NO and NO_2

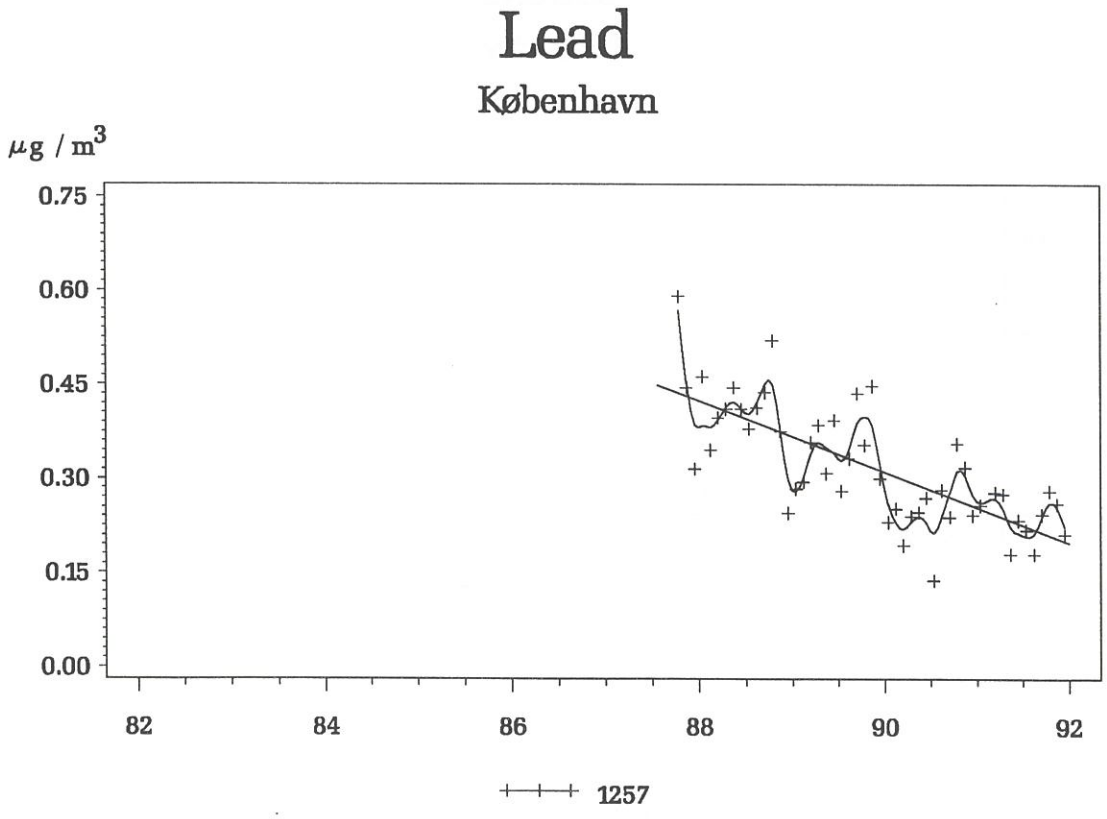
NO do show an increasing trend over the whole period 1982-91, while no significant trend is observed for NO_2 (figure 4.29-37). It is very difficult on the basic of these measurements alone to make definite conclusions. Longer time series and more detailed measurements of ozone and meteorology as initiated in LMP III combined with air quality models are needed.



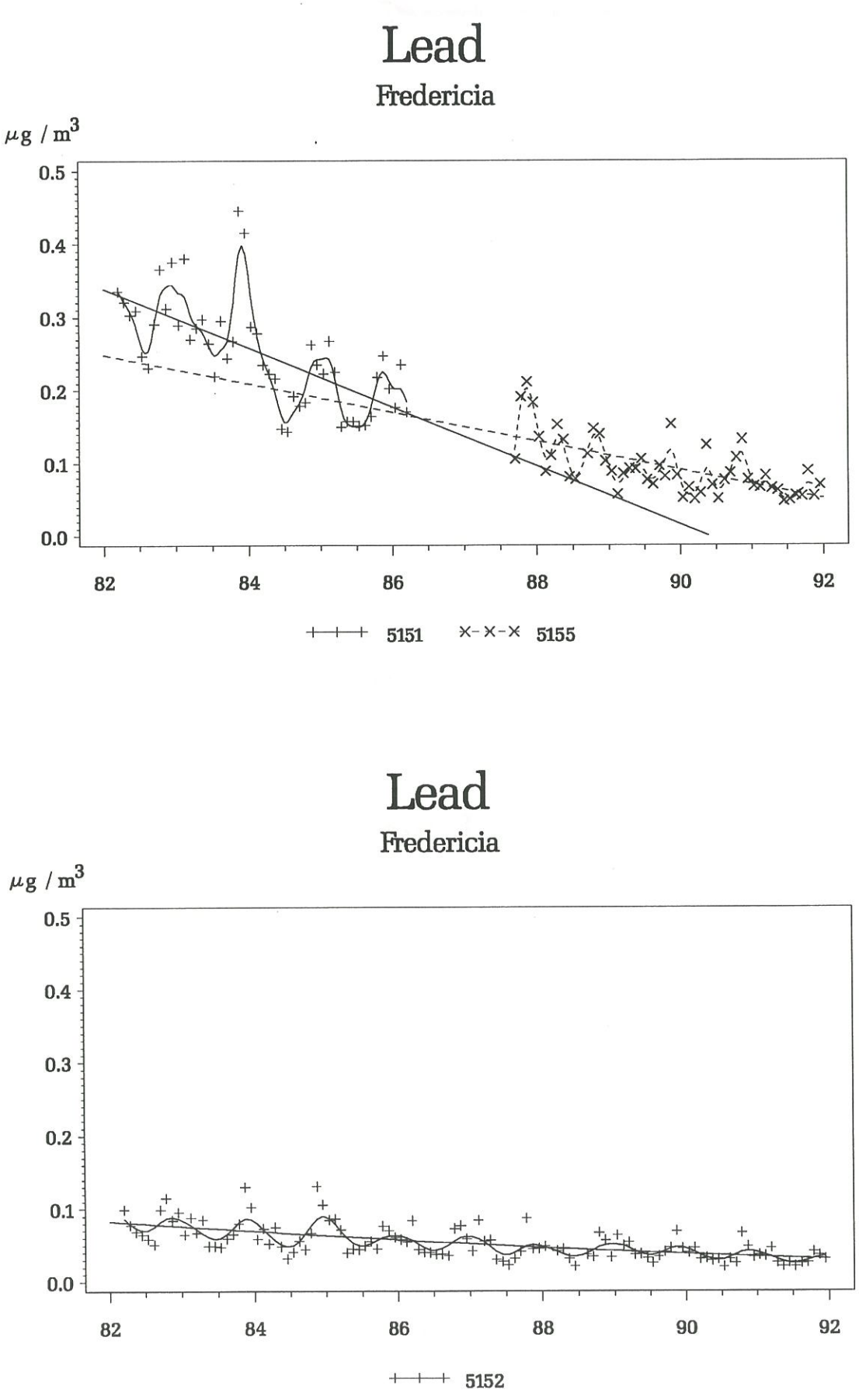


Figur 4.1. Time series for Lead (μg/m^3) in Copenhagen.

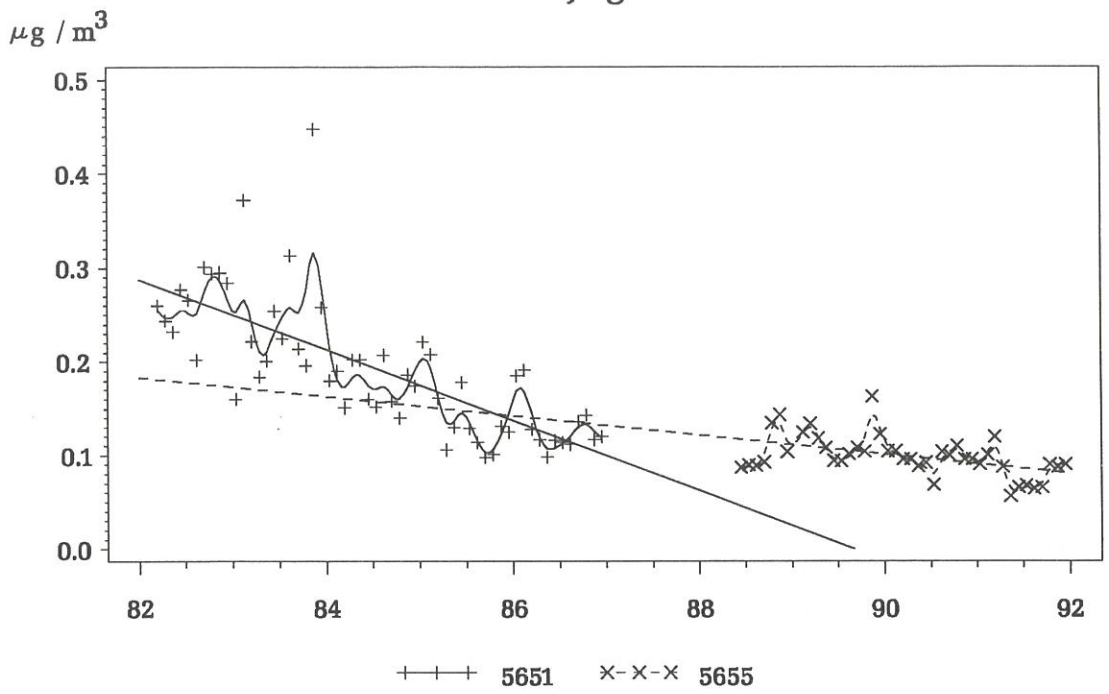


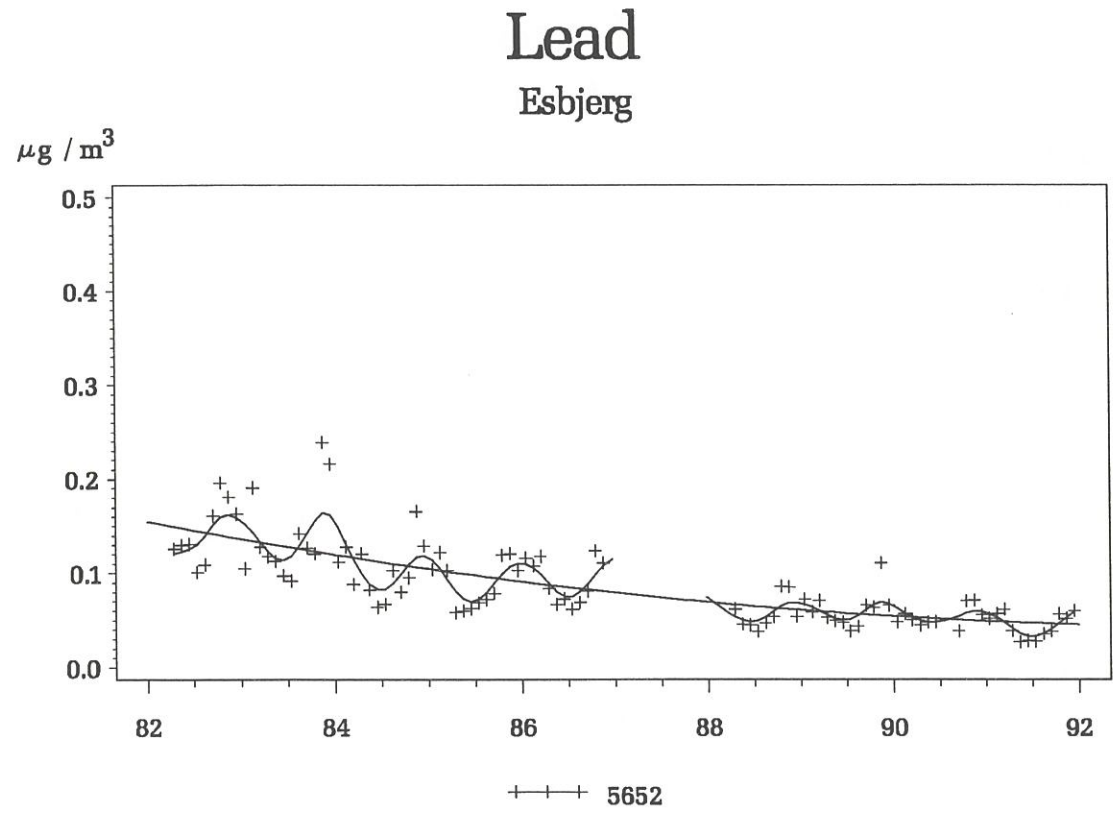


Figur 4.2. Time series for Lead (μg/m³) in Copenhagen.

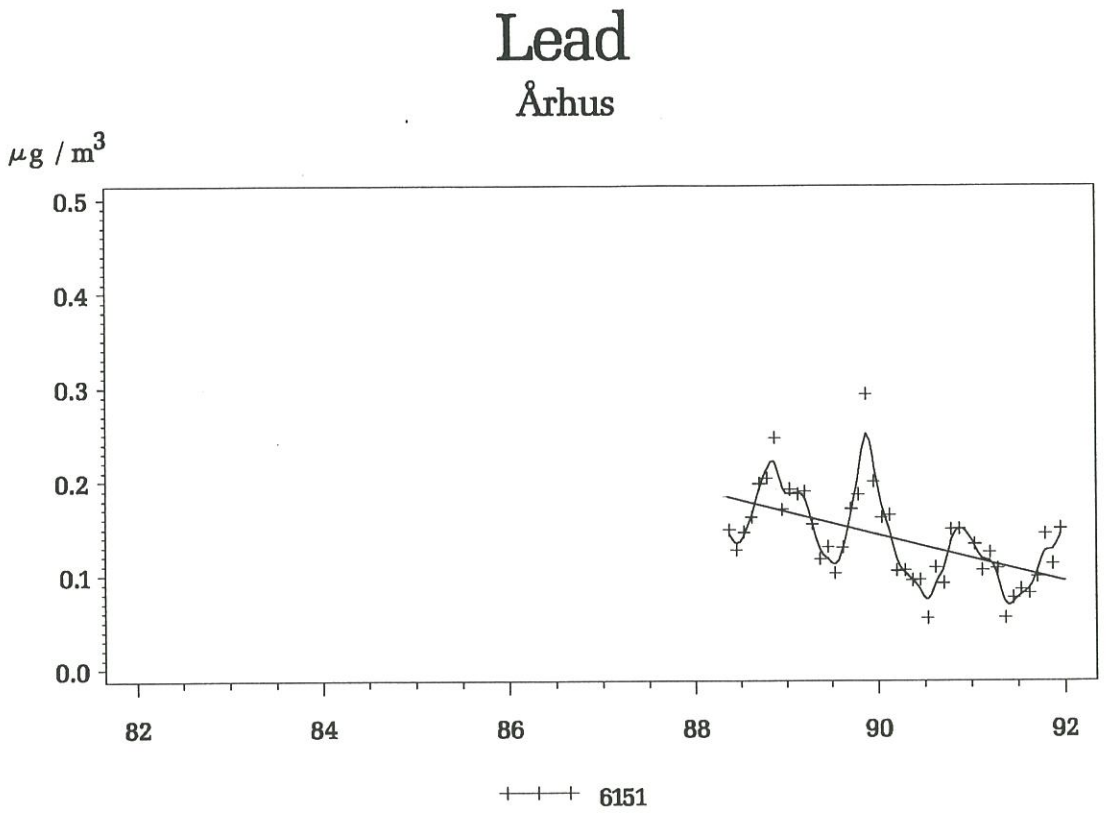


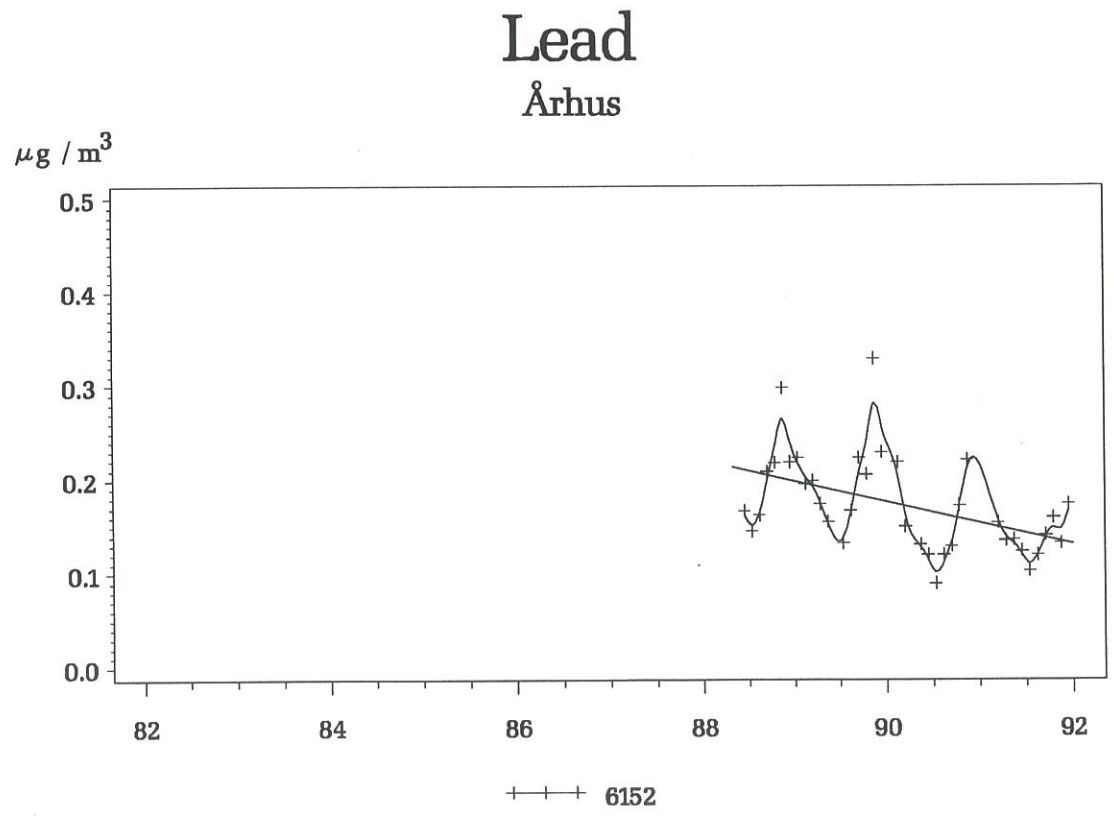
Figur 4.3. Time series for Lead (µg/m^3) in Fredericia.



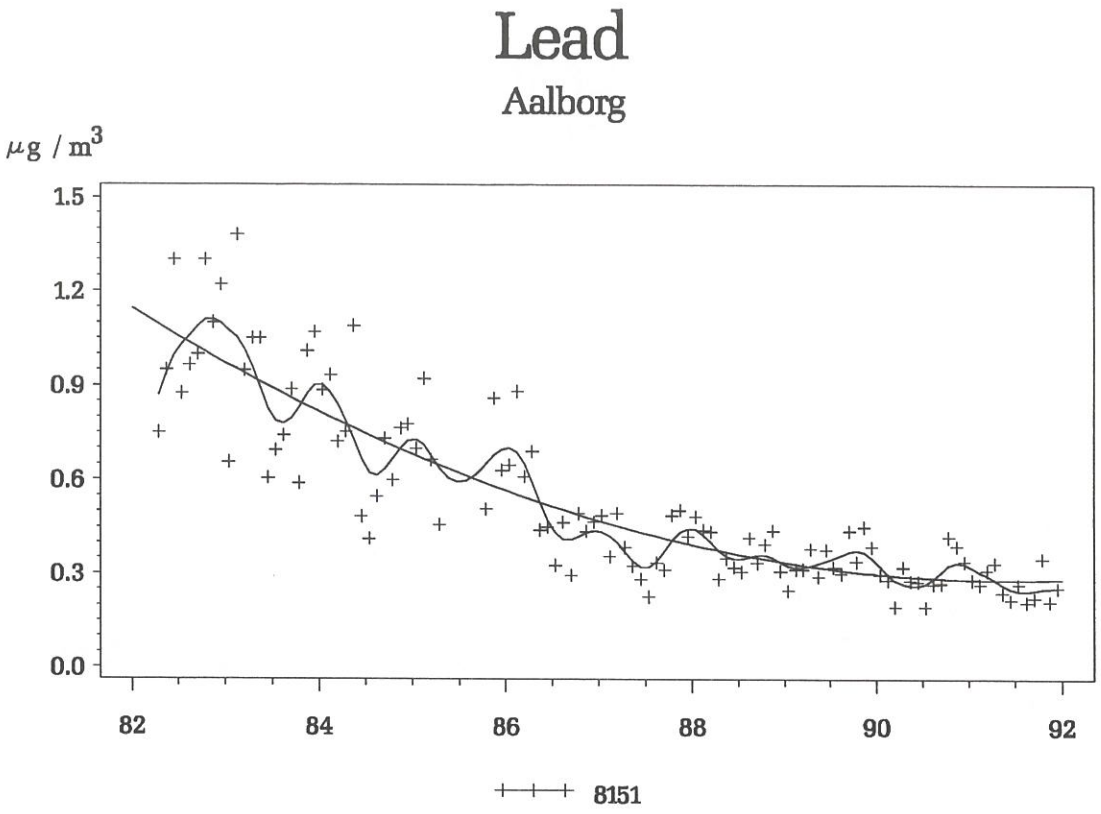


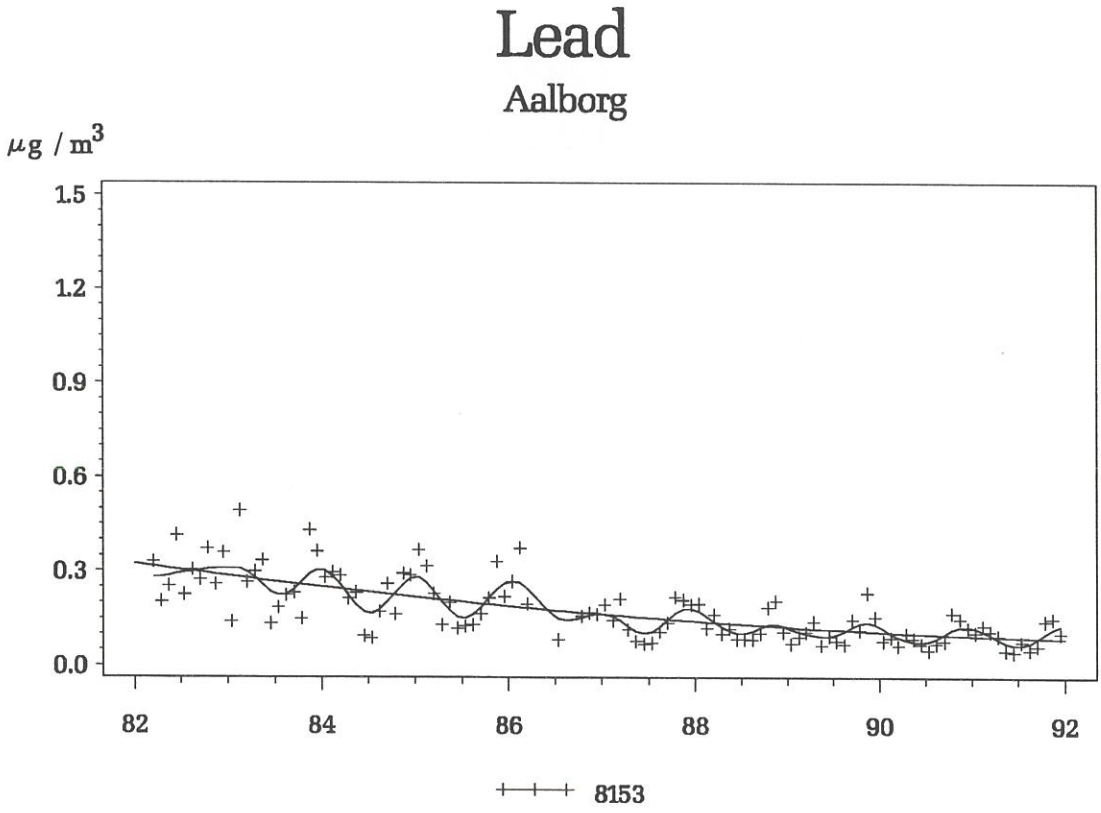
Figur 4.4. Time series for Lead (µg/m^3) in Esbjerg.



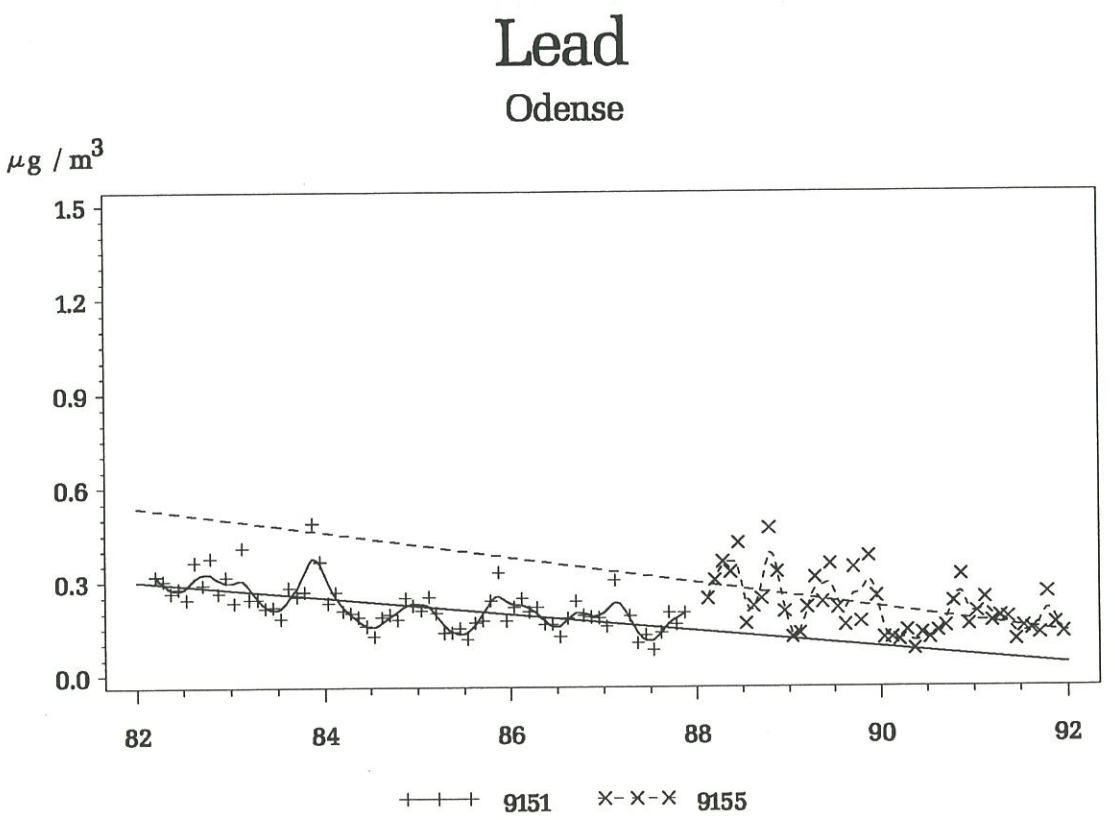


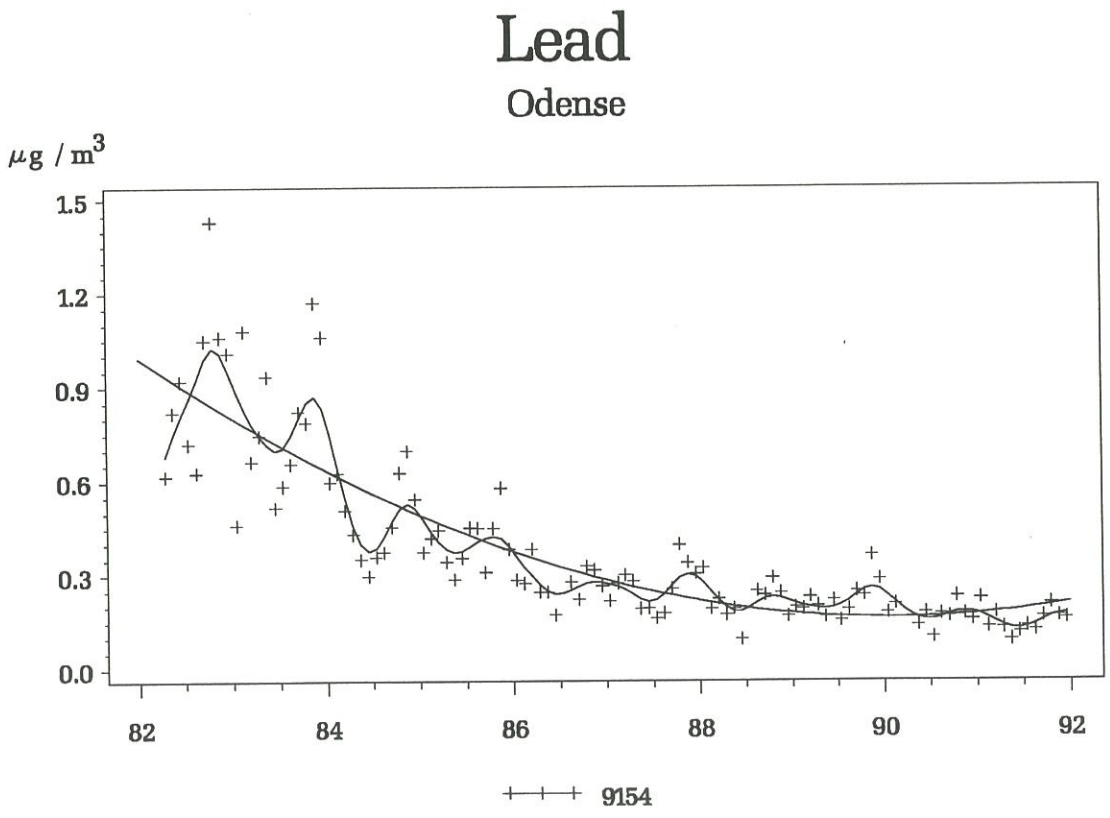
Figur 4.5. Time series for Lead (µg/m^3) in Århus.



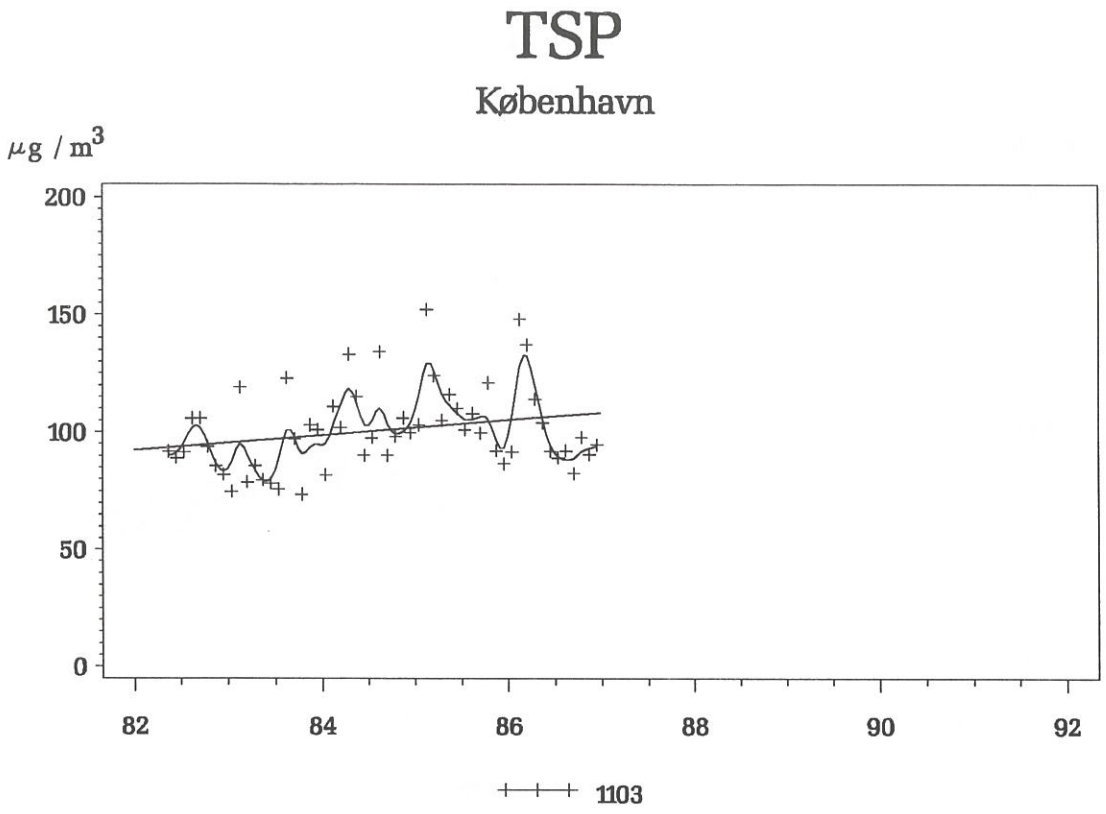


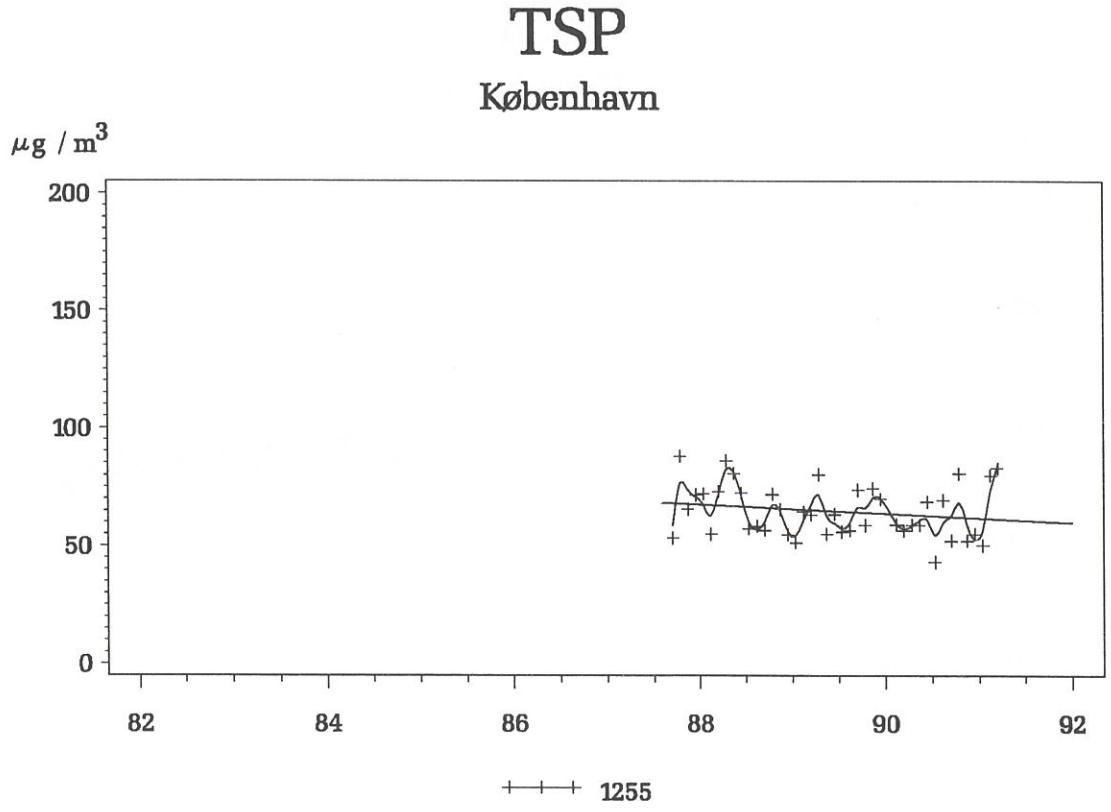
Figur 4.6. Time series for Lead (µg/m³) in Aalborg.



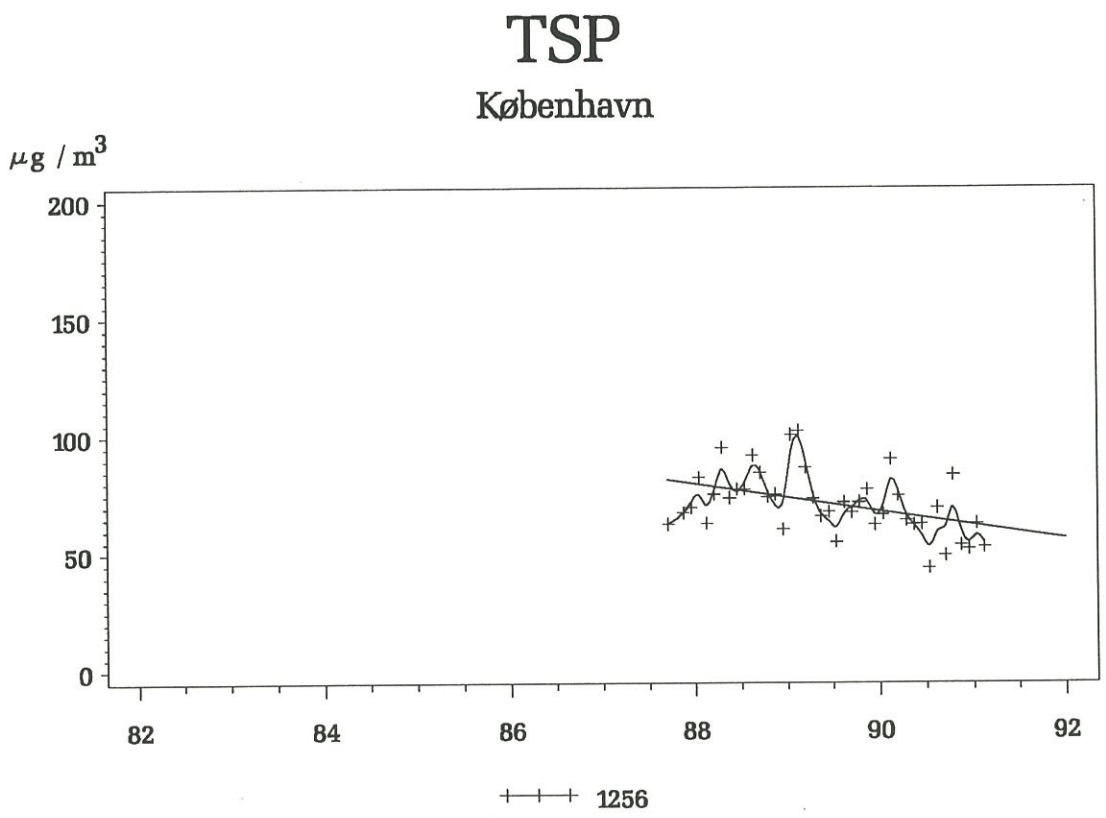


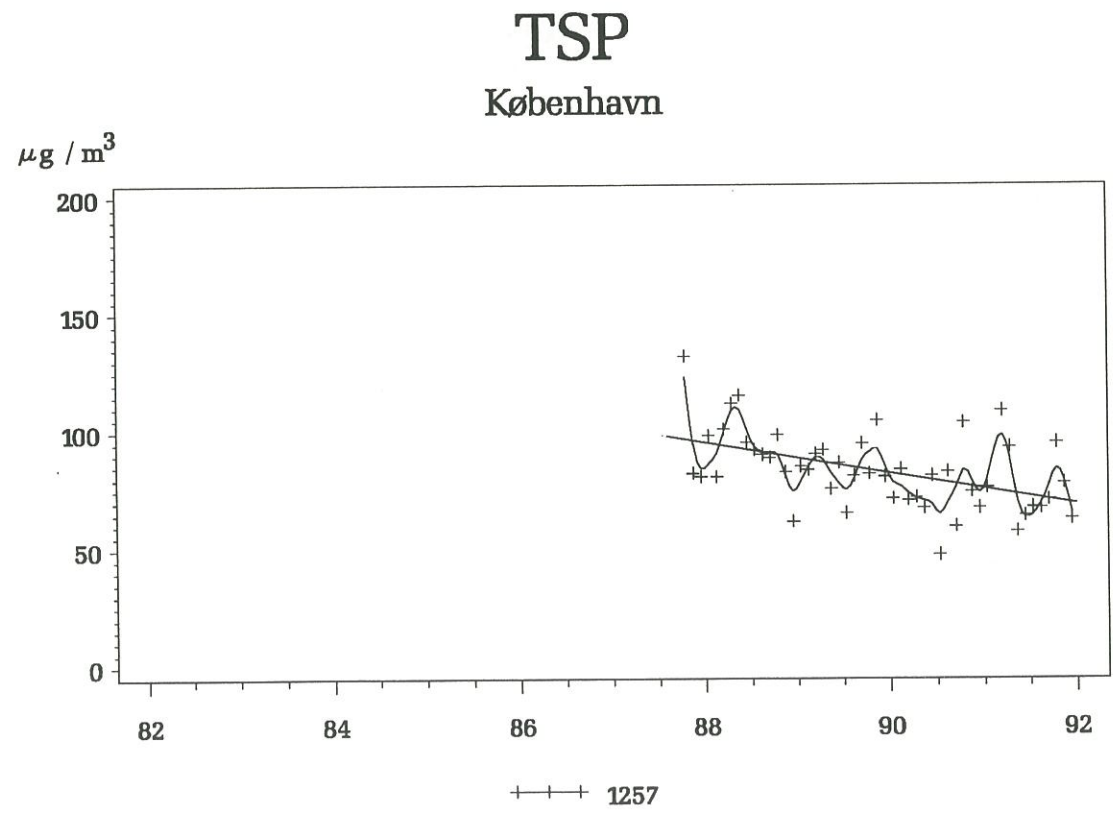
Figur 4.7. Time series for Lead (μg/m³) in Odense.



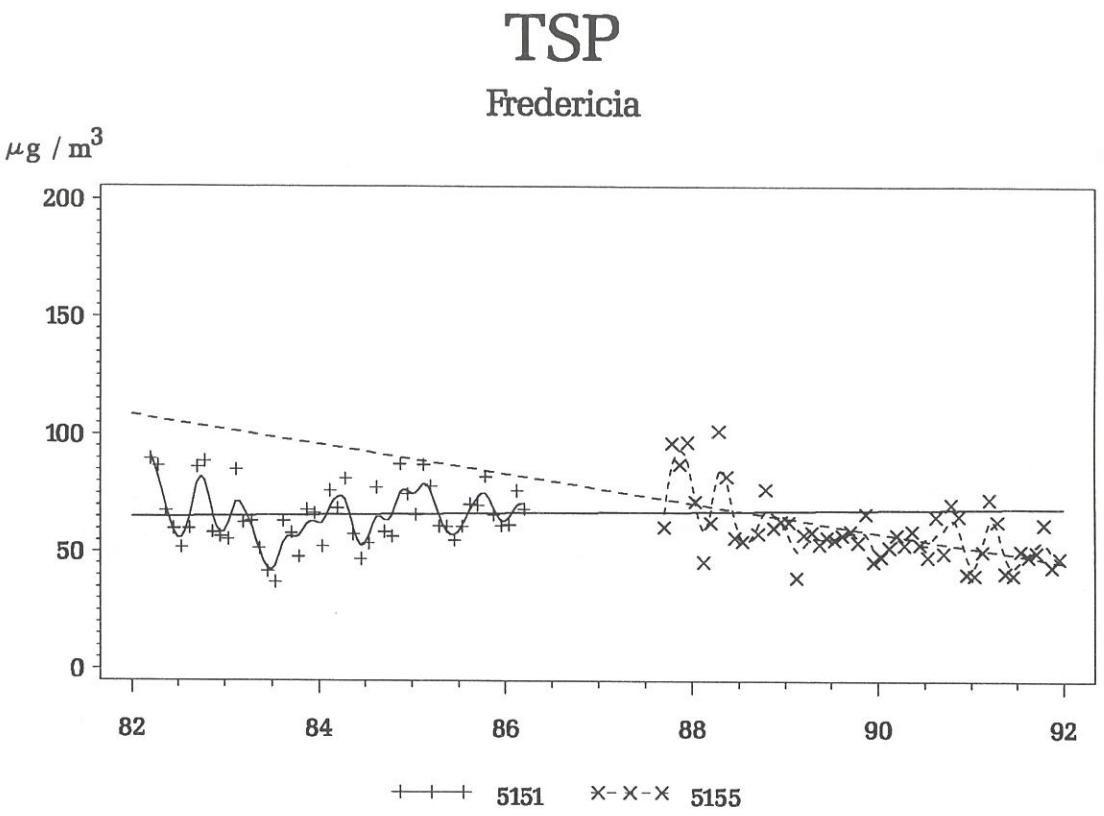


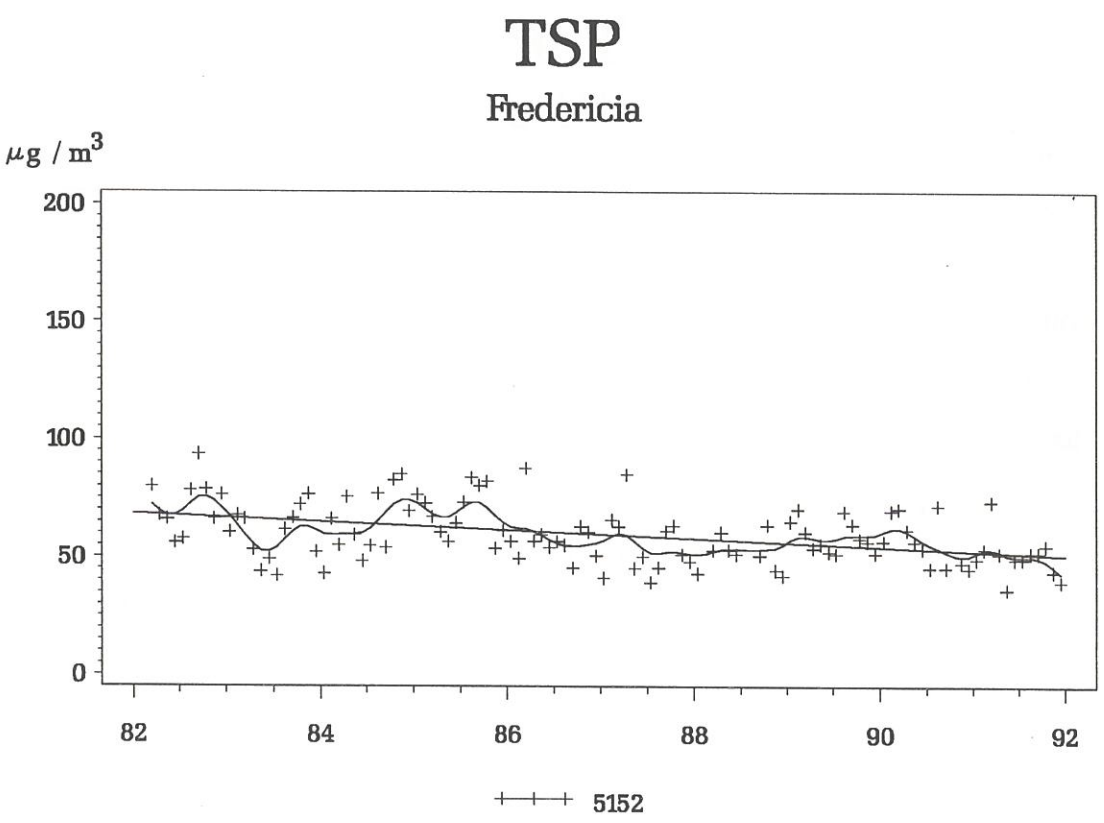
Figur 4.8. Time series for TSP (µg/m³) in Copenhagen.



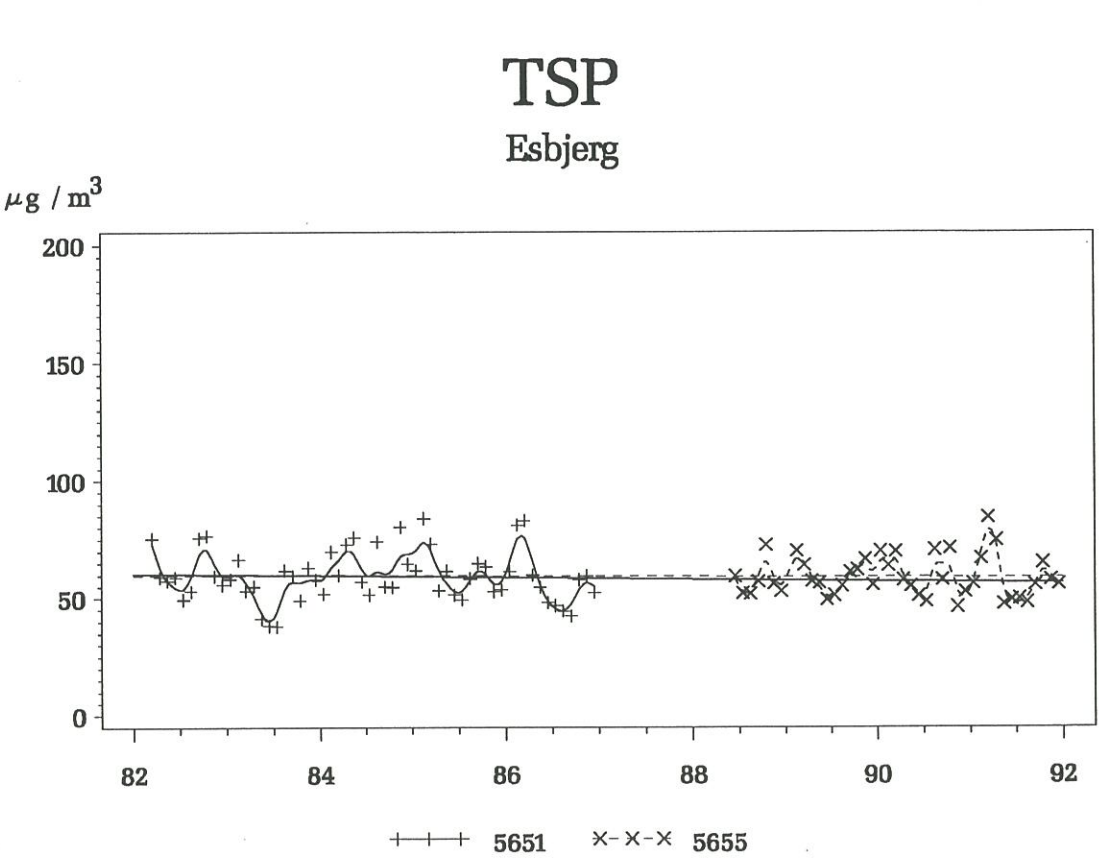


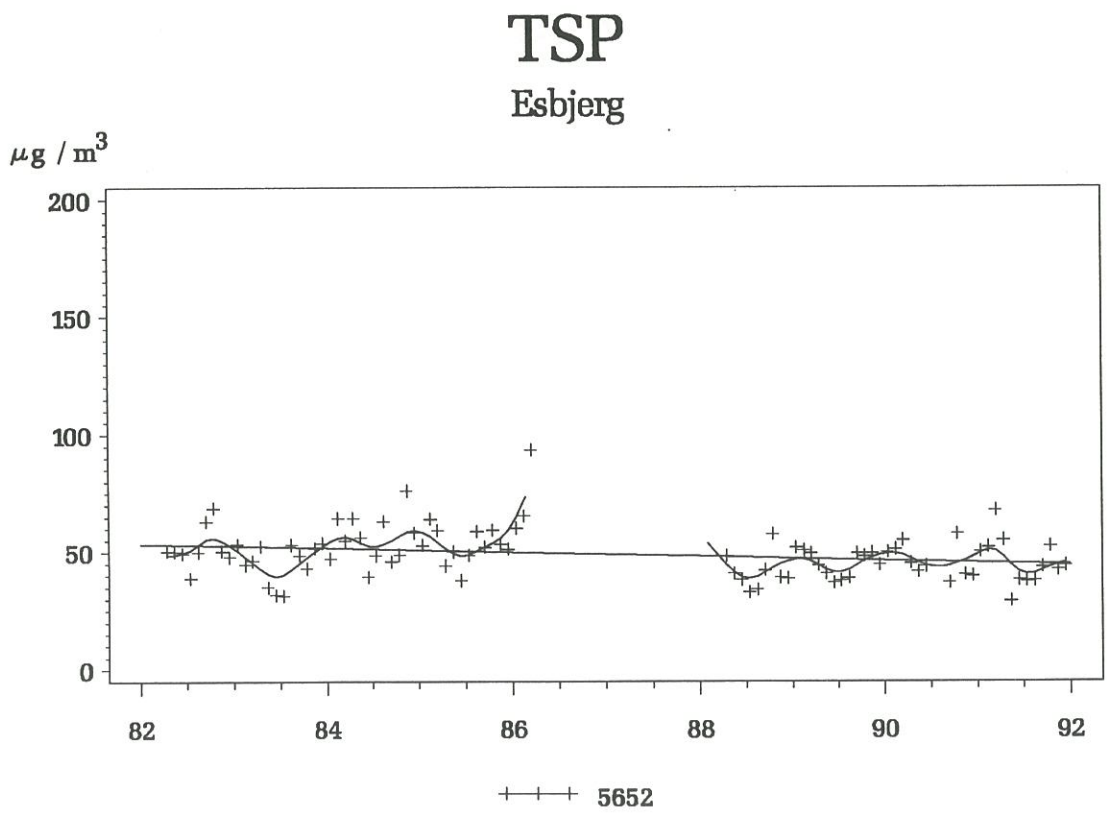
Figur 4.9. Time series for TSP (µg/m^3) in Copenhagen.



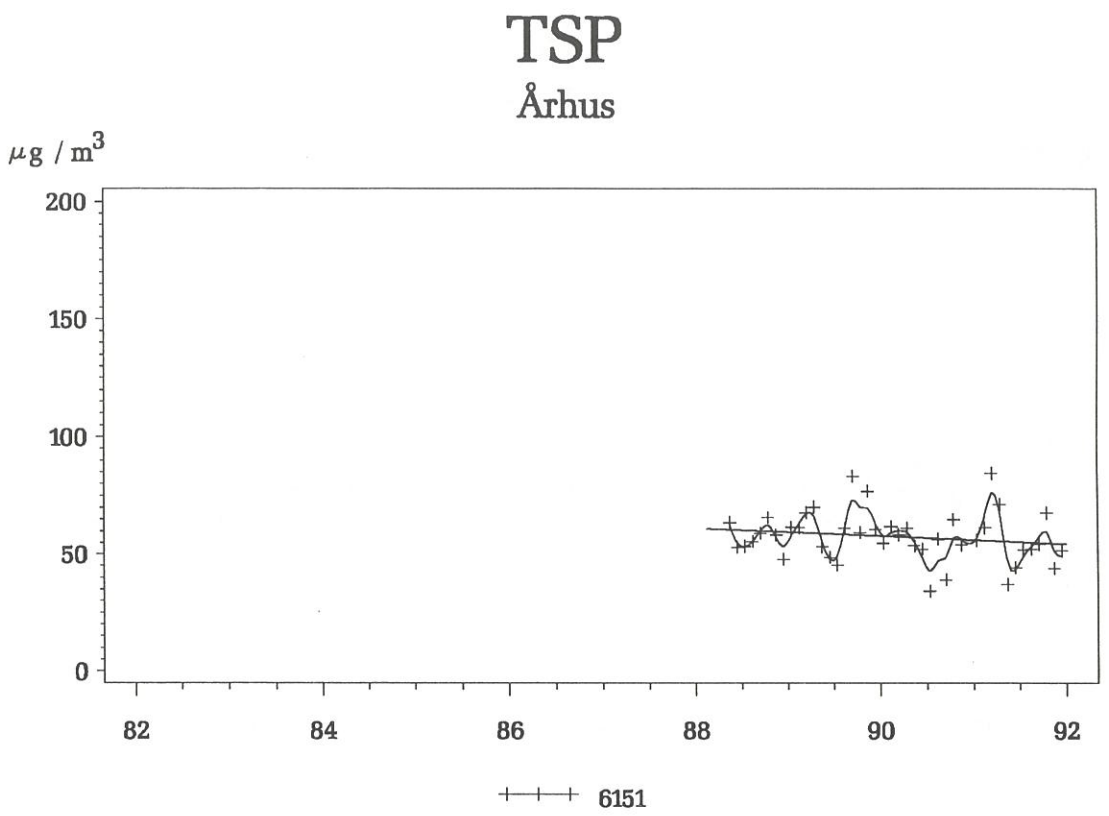


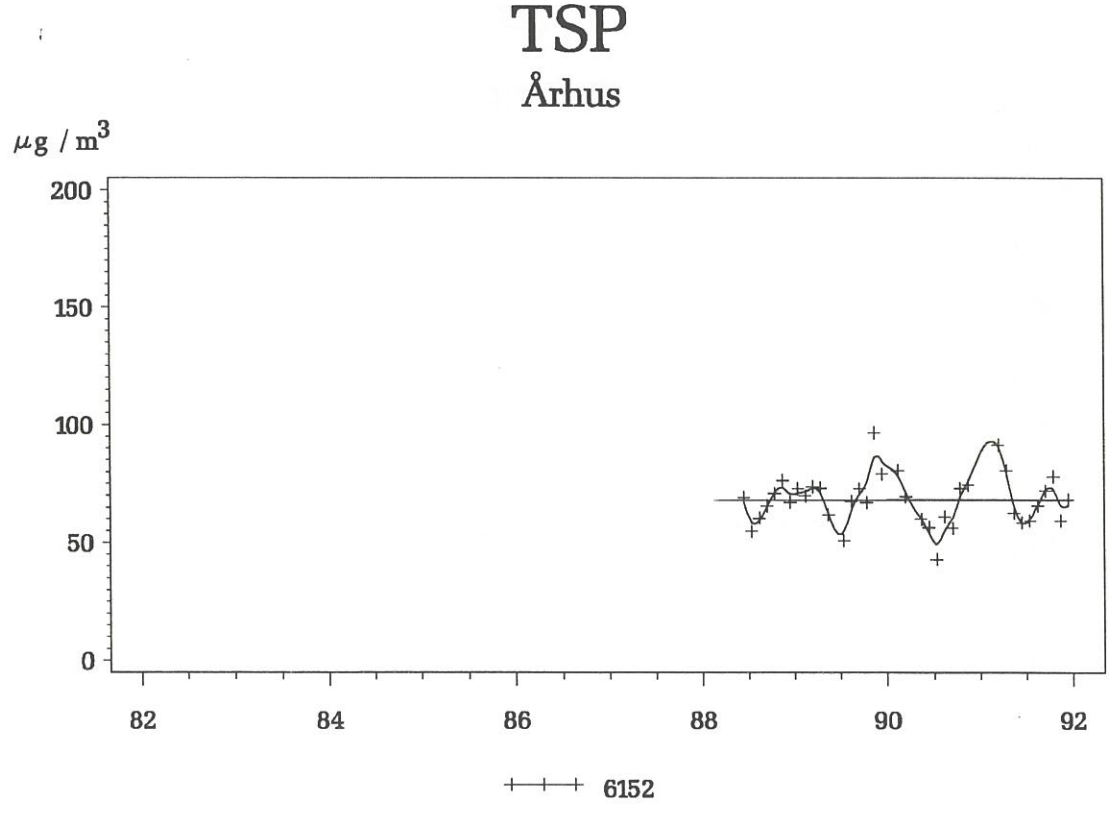
Figur 4.10. Time series for TSP (µg/m^3) in Fredericia.



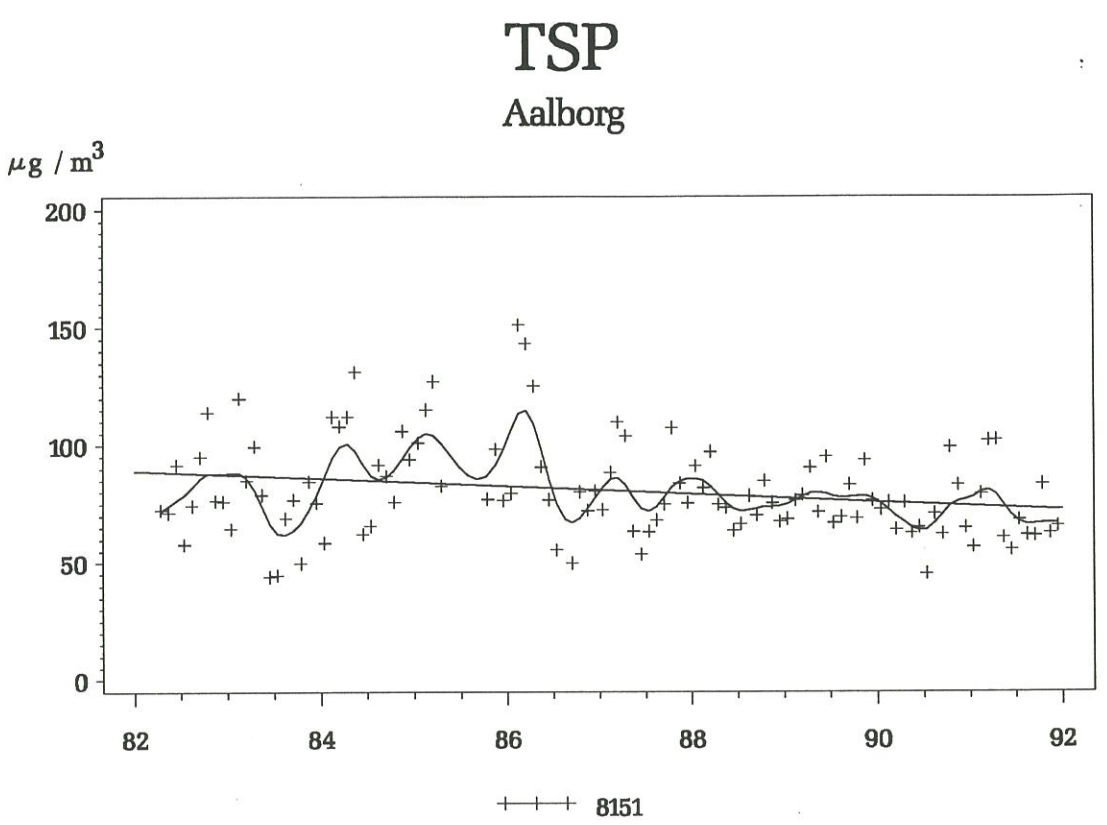


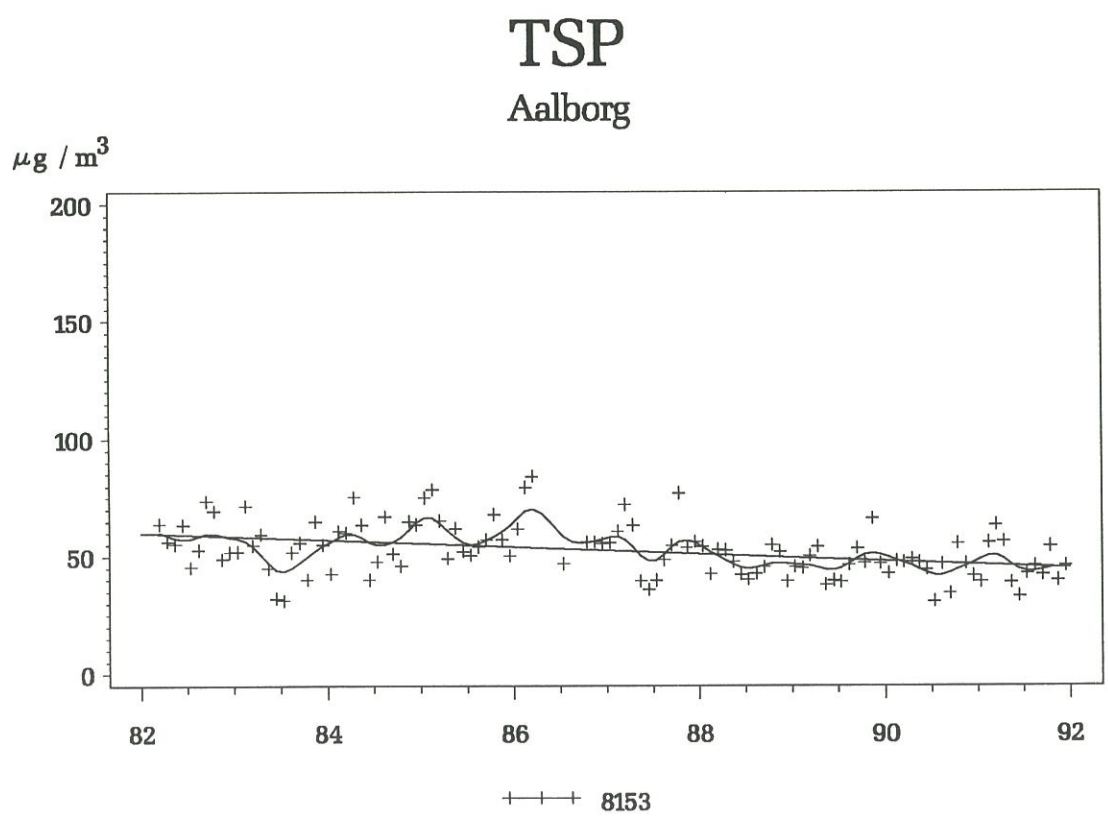
Figur 4.11. Time series for TSP (µg/m^3) in Esbjerg.



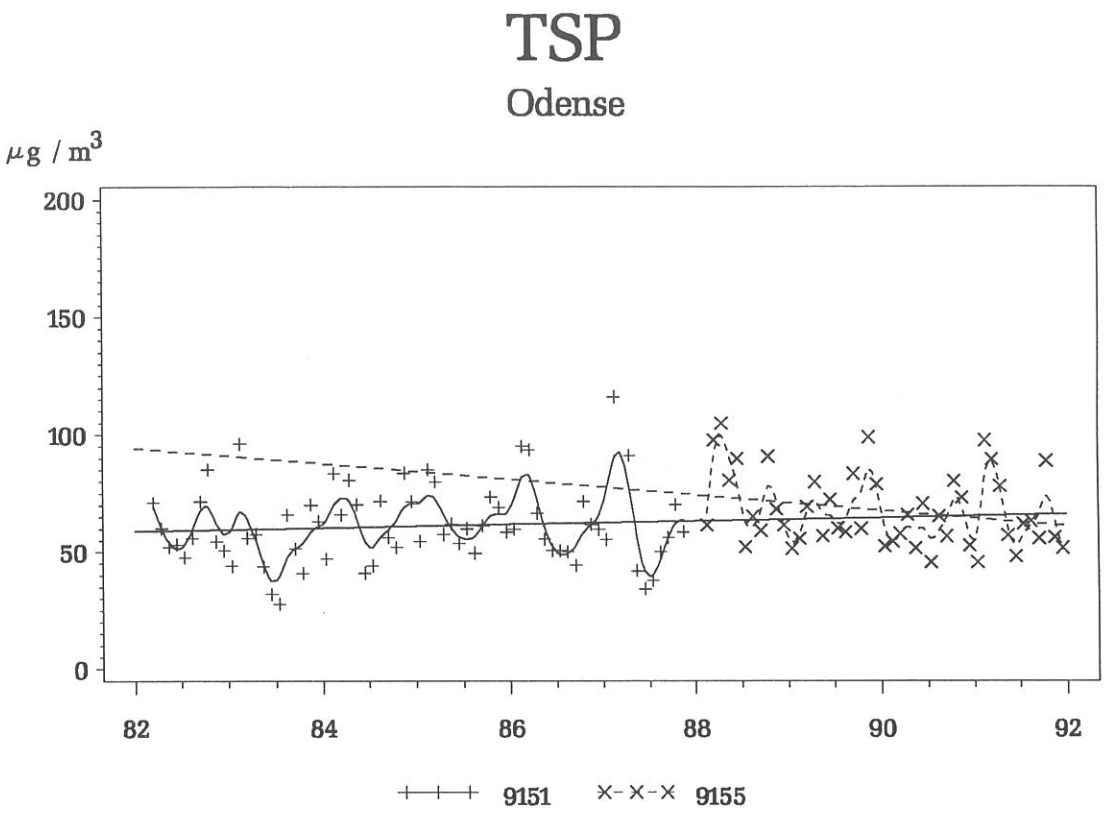


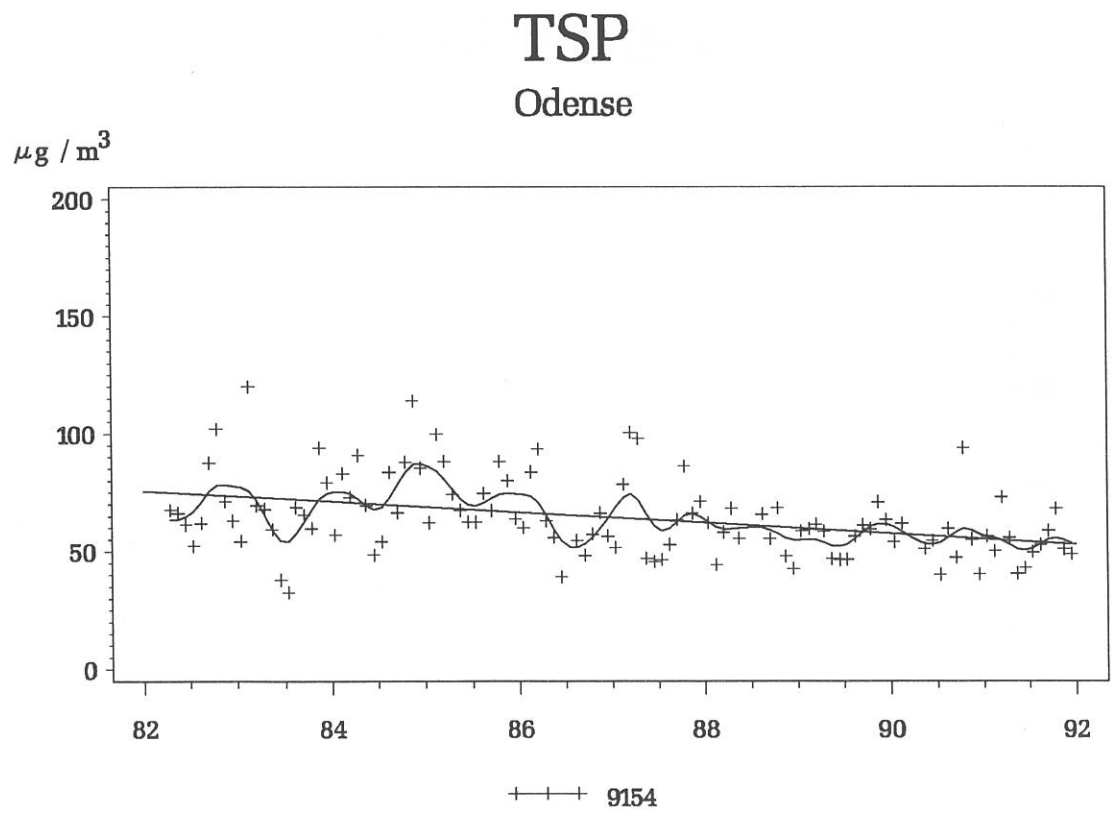
Figur 4.12. Time series for TSP (µg/m^3) in Århus.



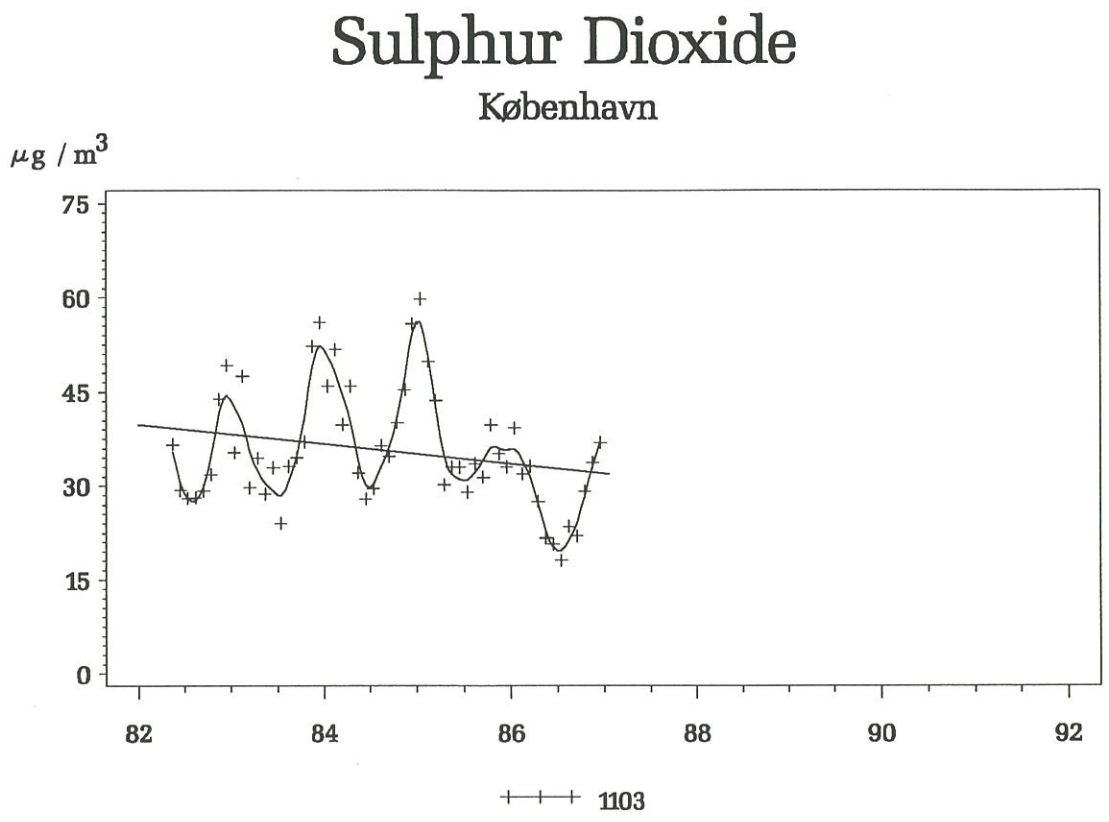


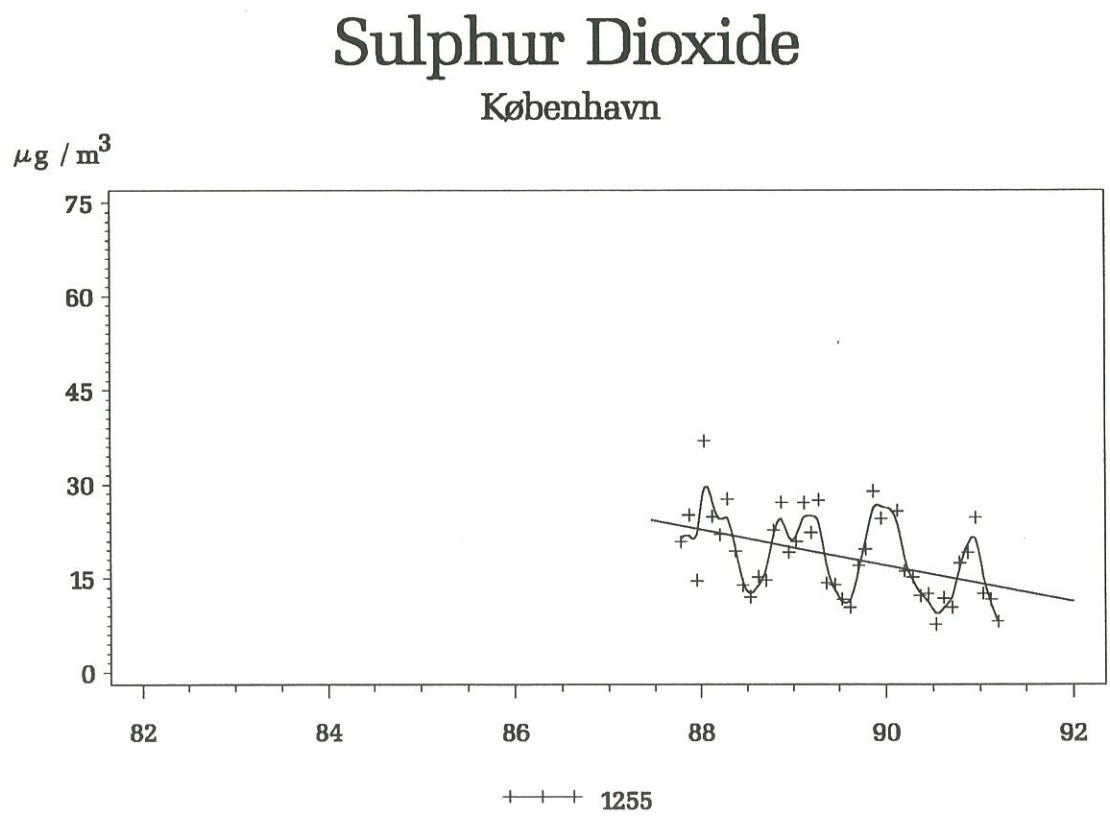
Figur 4.13. Time series for TSP (µg/m³) in Aalborg.



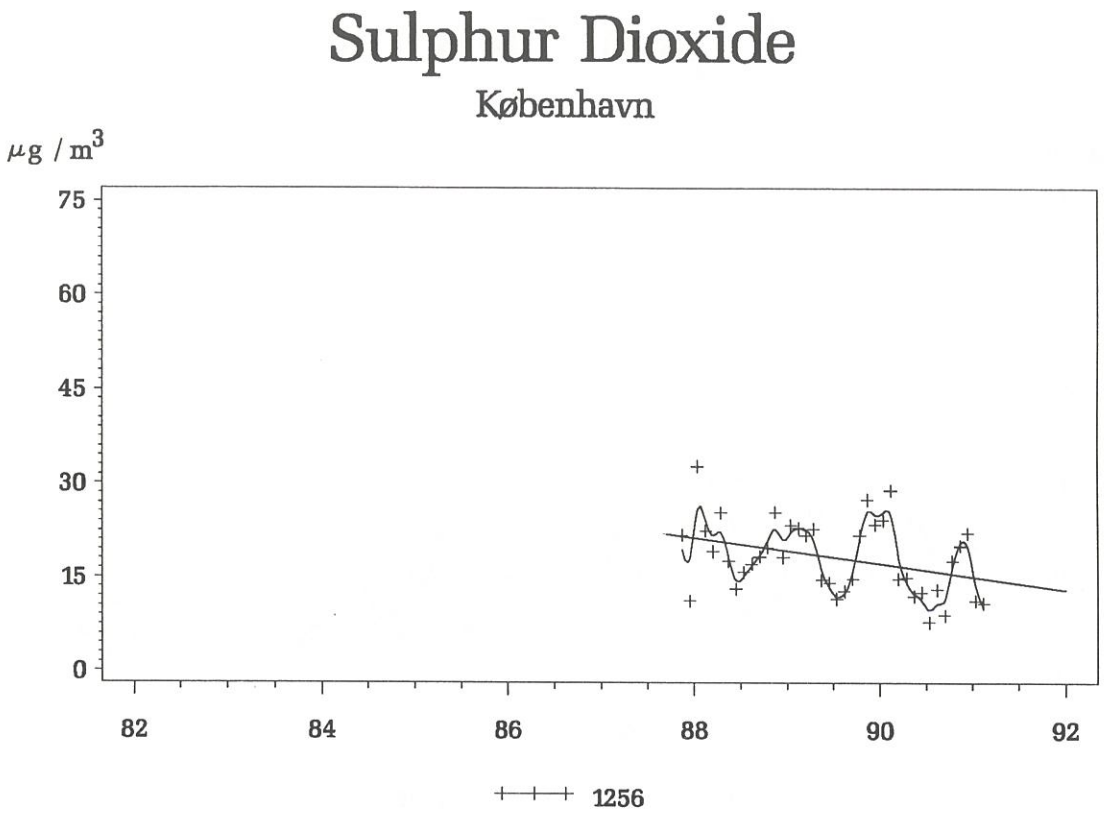


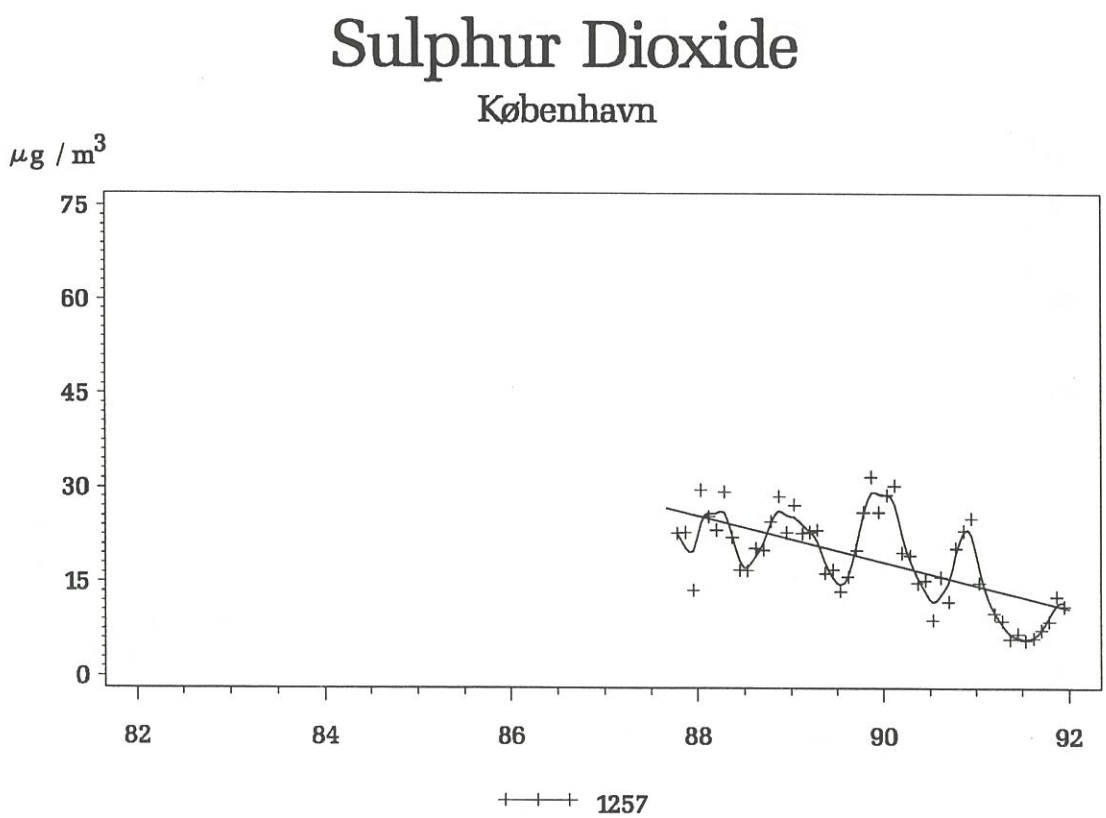
Figur 4.14. Time series for TSP (µg/m³) in Odense.



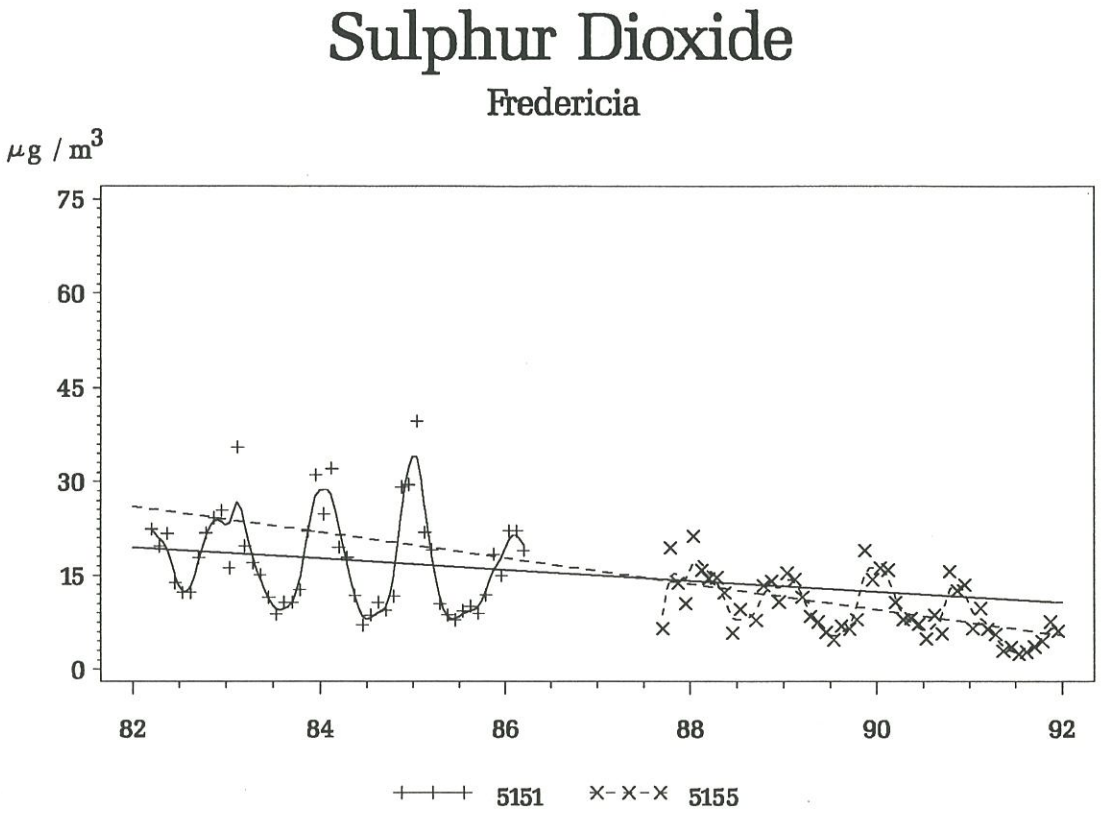


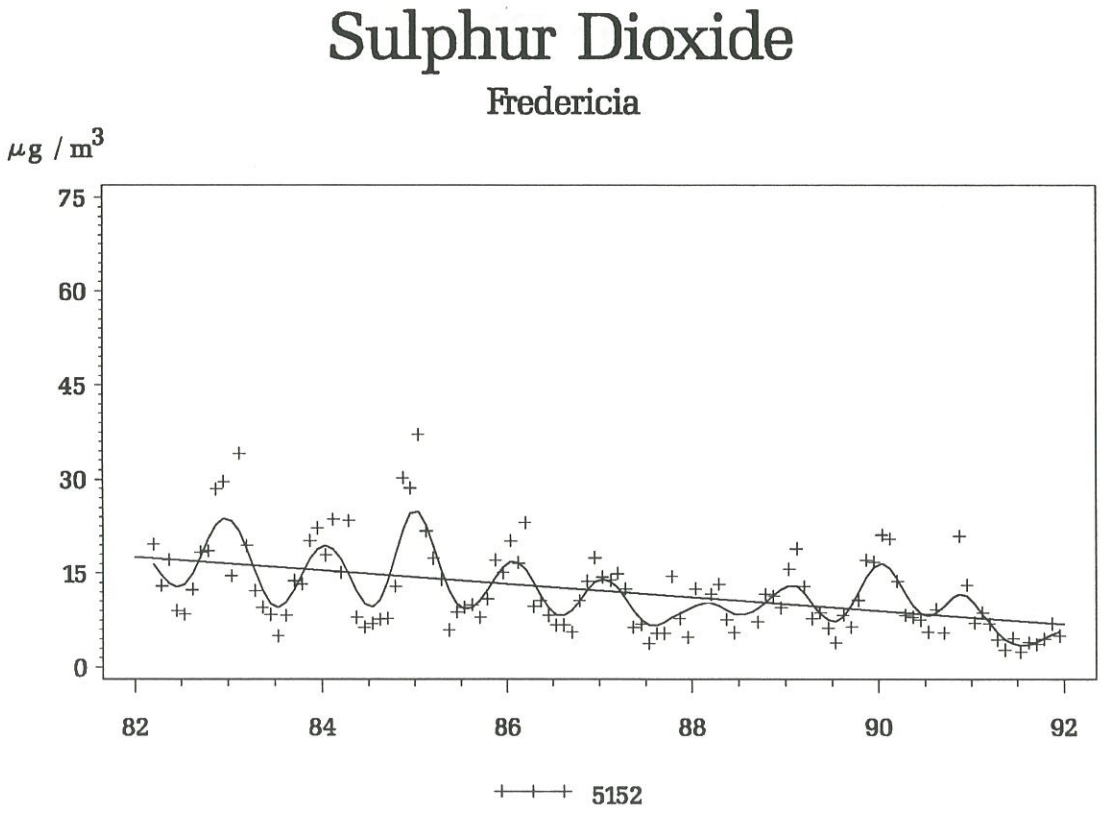
Figur 4.15. Time series for Sulphur Dioxide (µg/m^3) in Copenhagen.



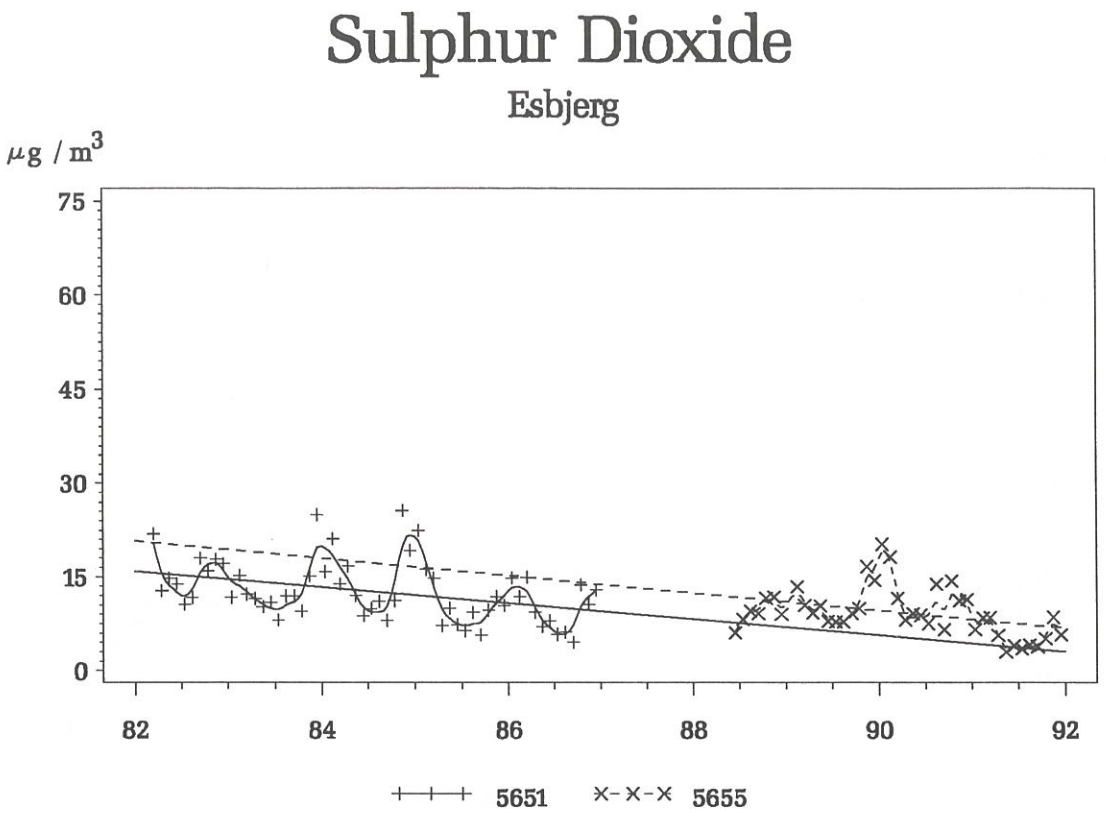


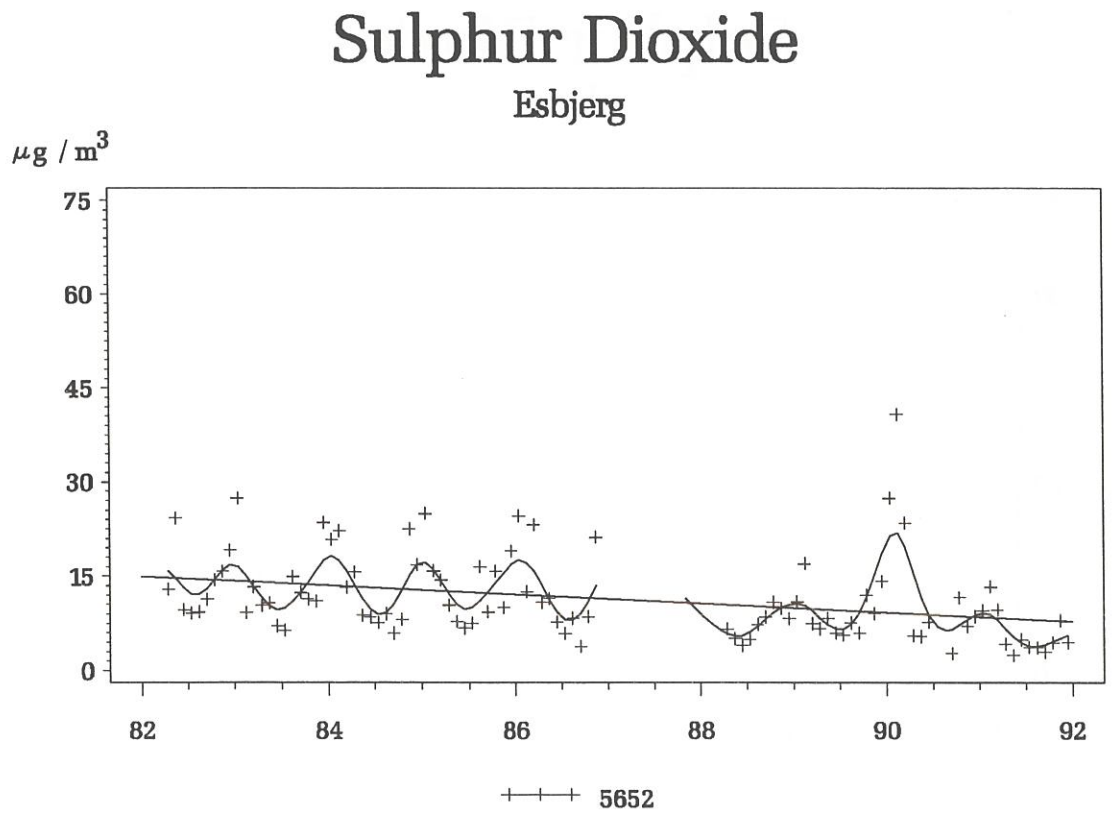
Figur 4.16. Time series for Sulphur Dioxide (μg/m³) in Copenhagen.



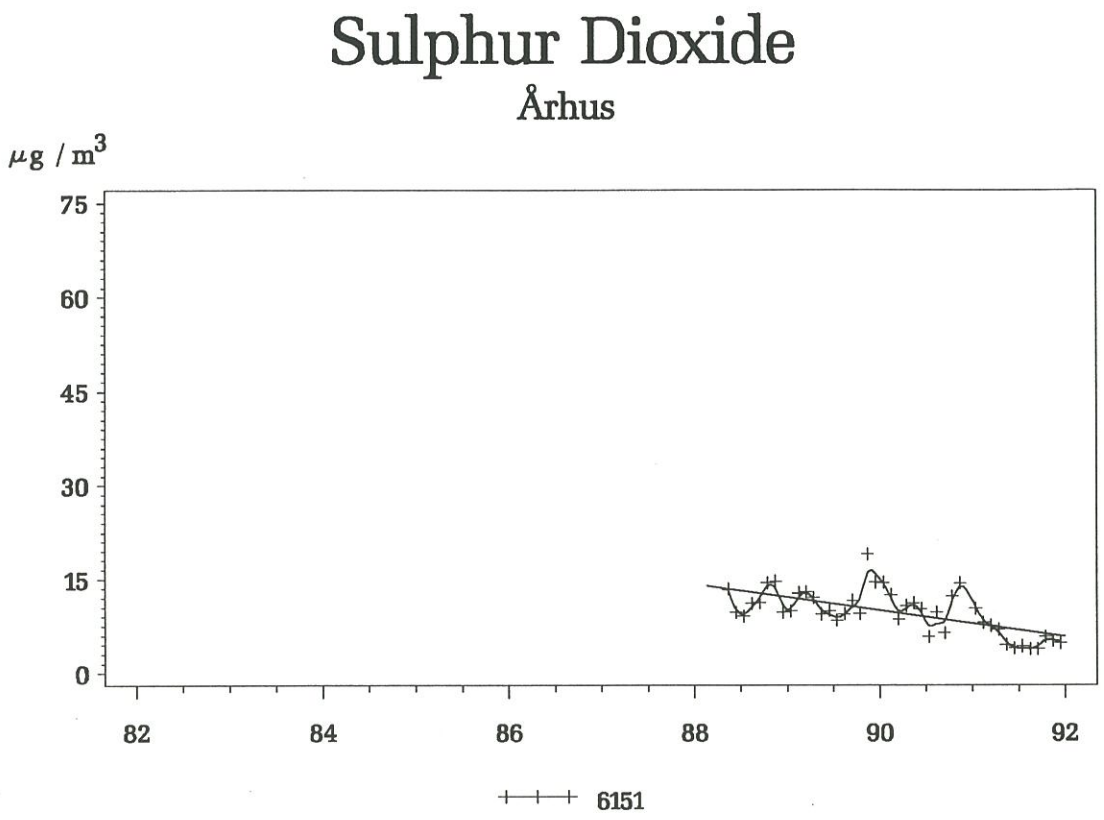


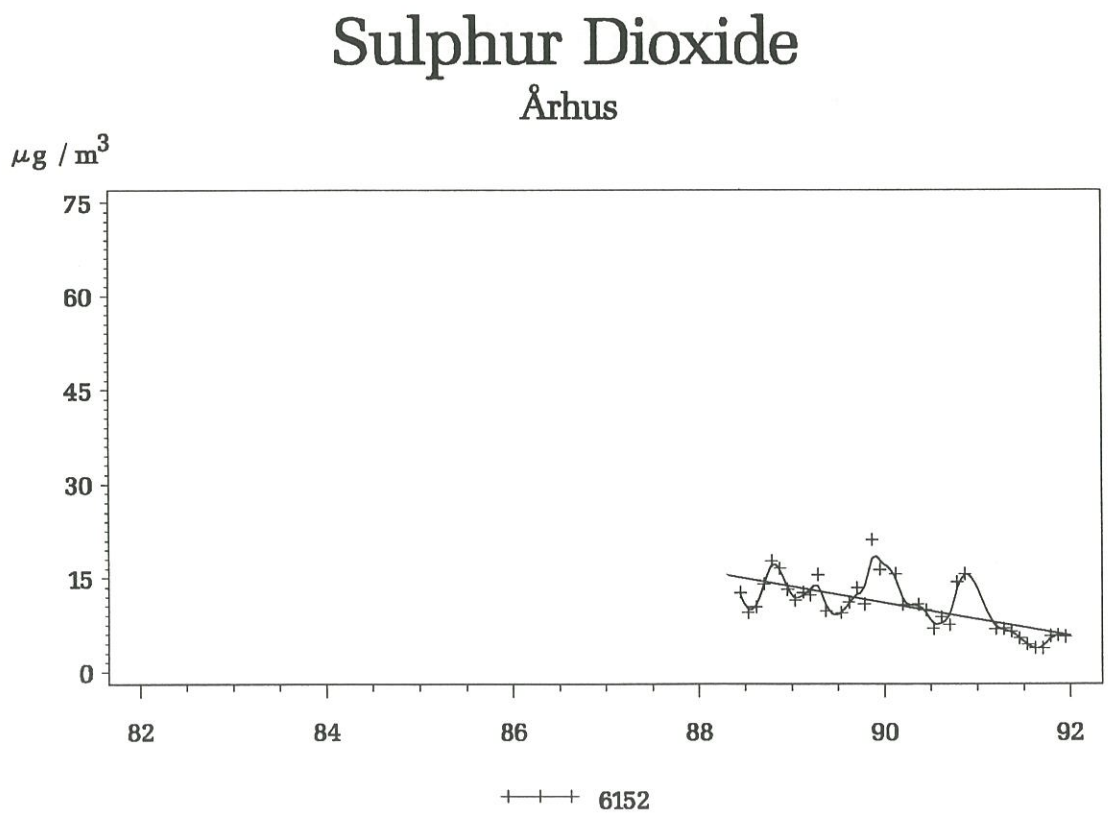
Figur 4.17. Time series for Sulphur Dioxide (µg/m^3) in Fredericia.



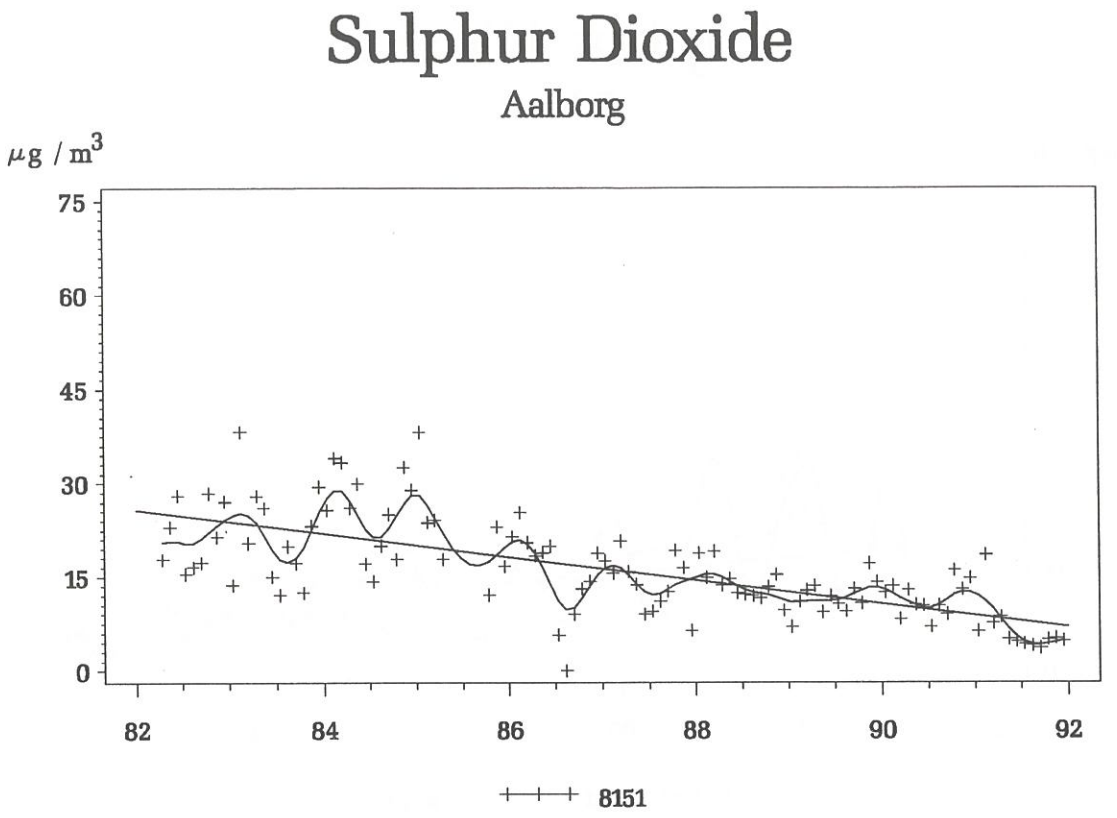


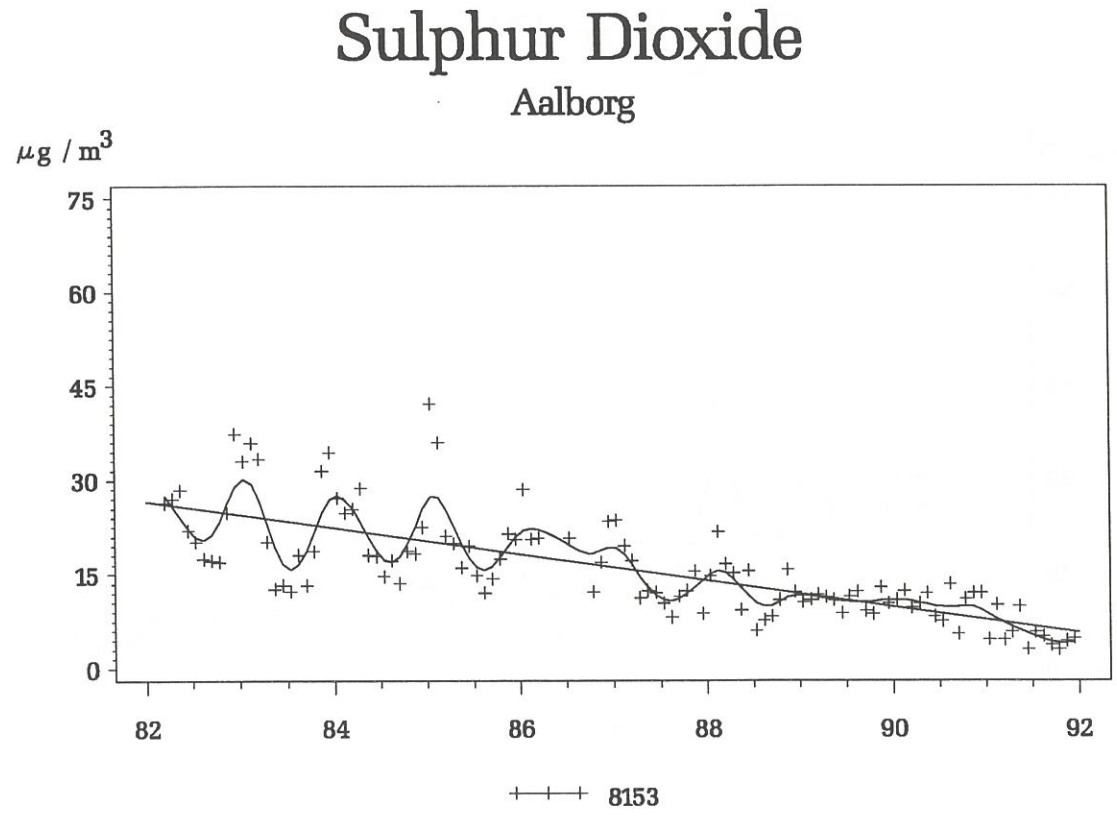
Figur 4.18. Time series for Sulphur Dioxide (µg/m^3) in Esbjerg.



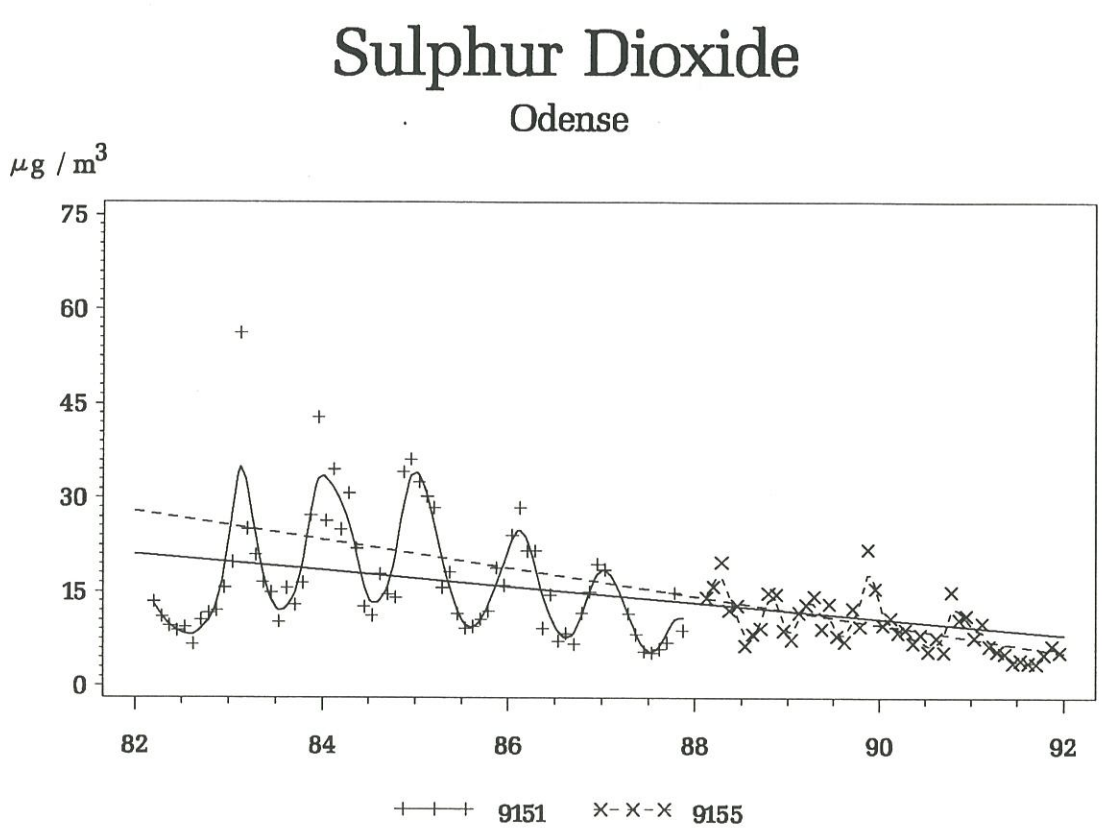


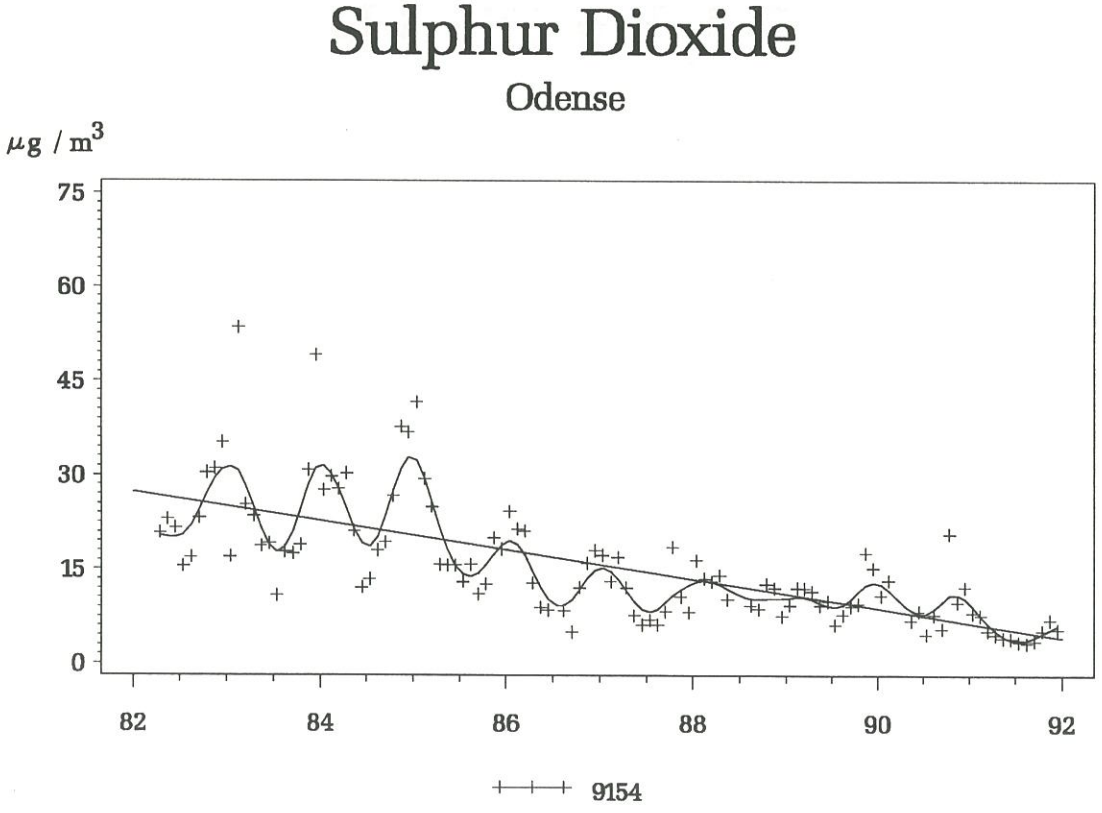
Figur 4.19. Time series for Sulphur Dioxide (µg/m^3) in Århus.



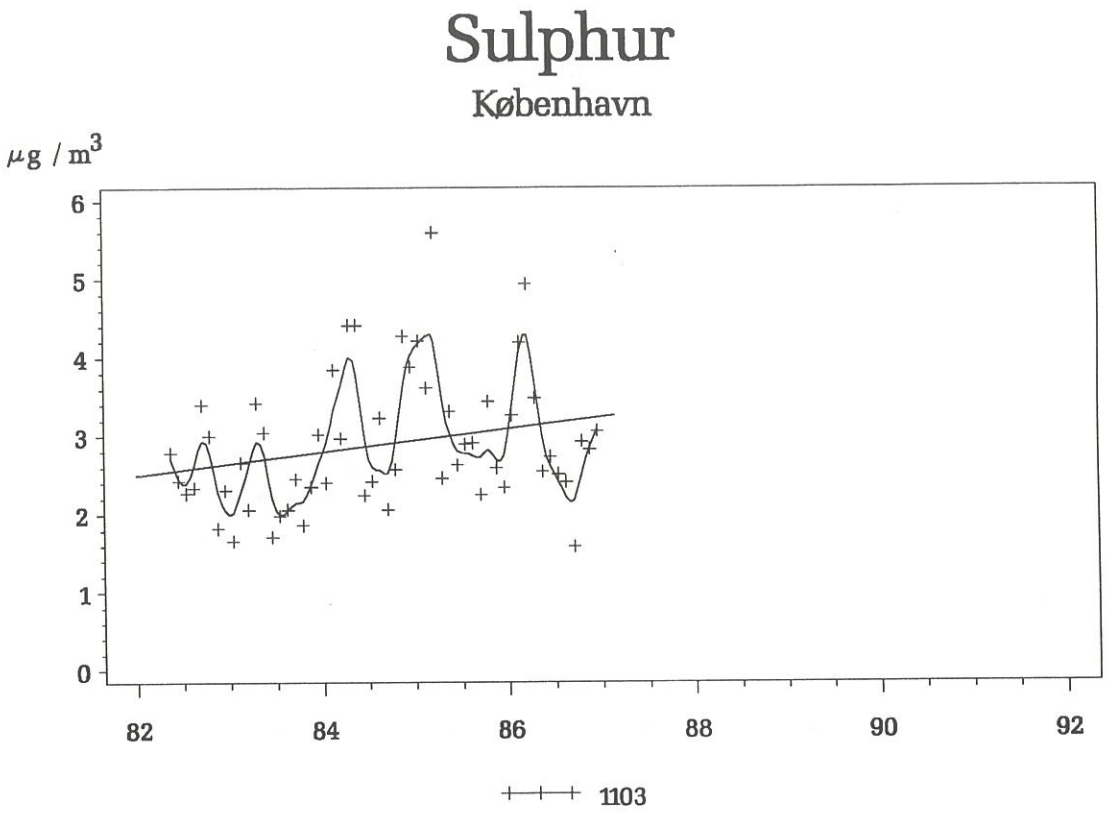


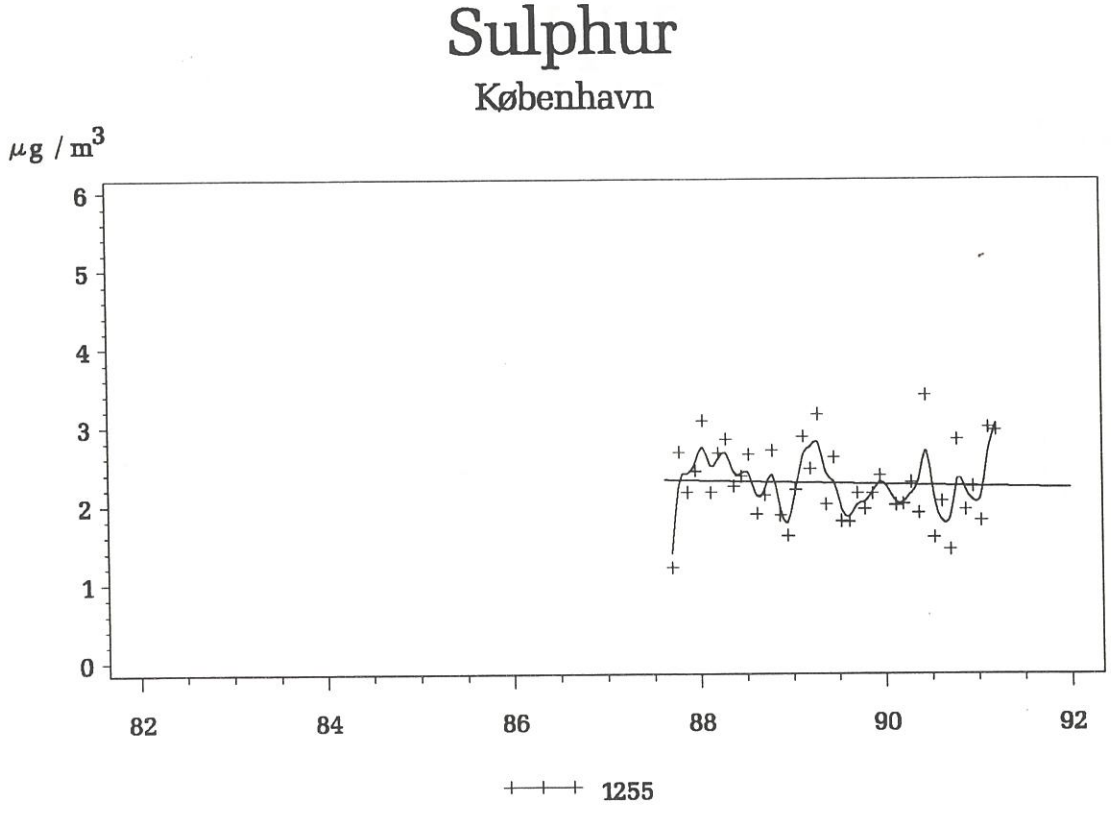
Figur 4.20. Time series for Sulphur Dioxide (µg/m^3) in Aalborg.



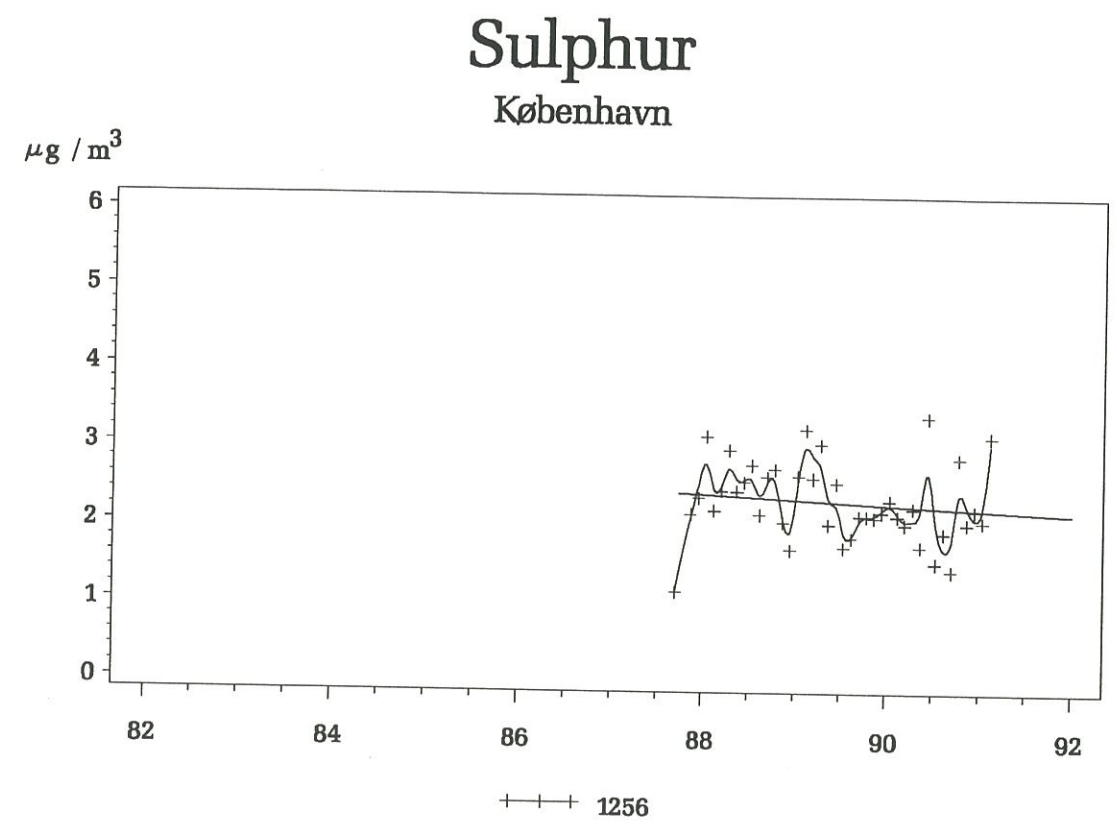


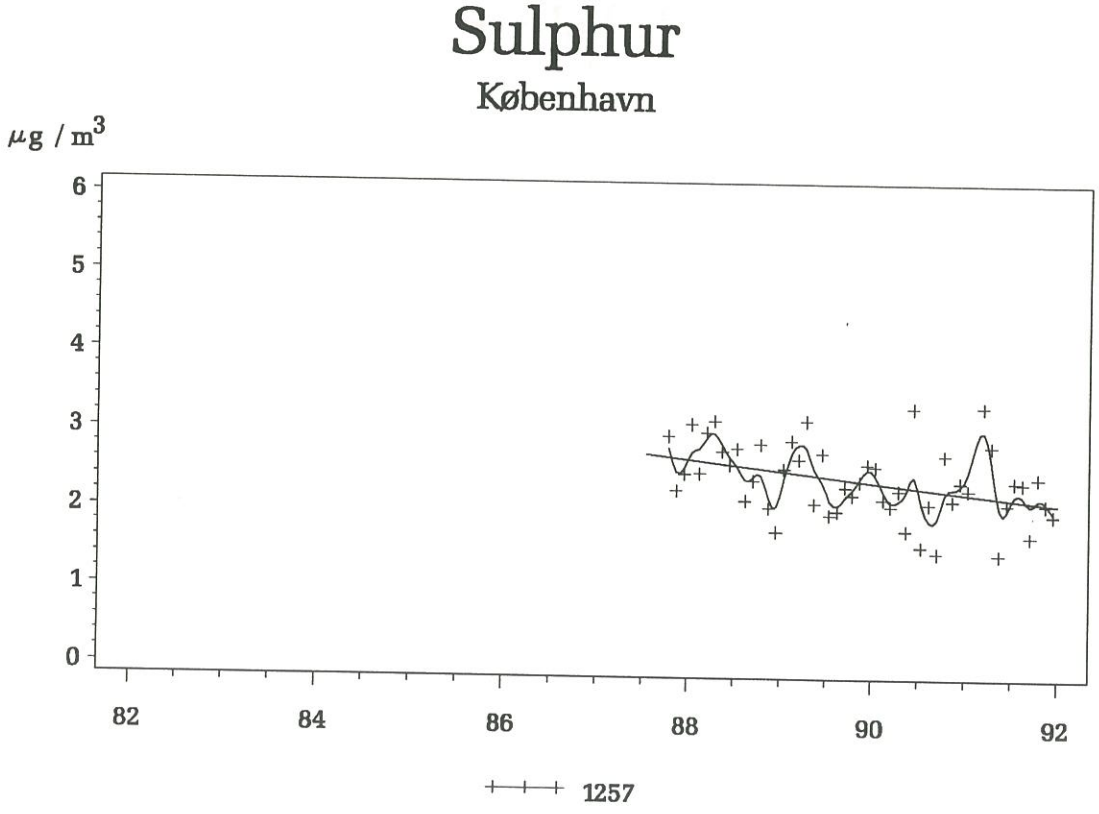
Figur 4.21. Time series for Sulphur Dioxide (µg/m³) in Odense.



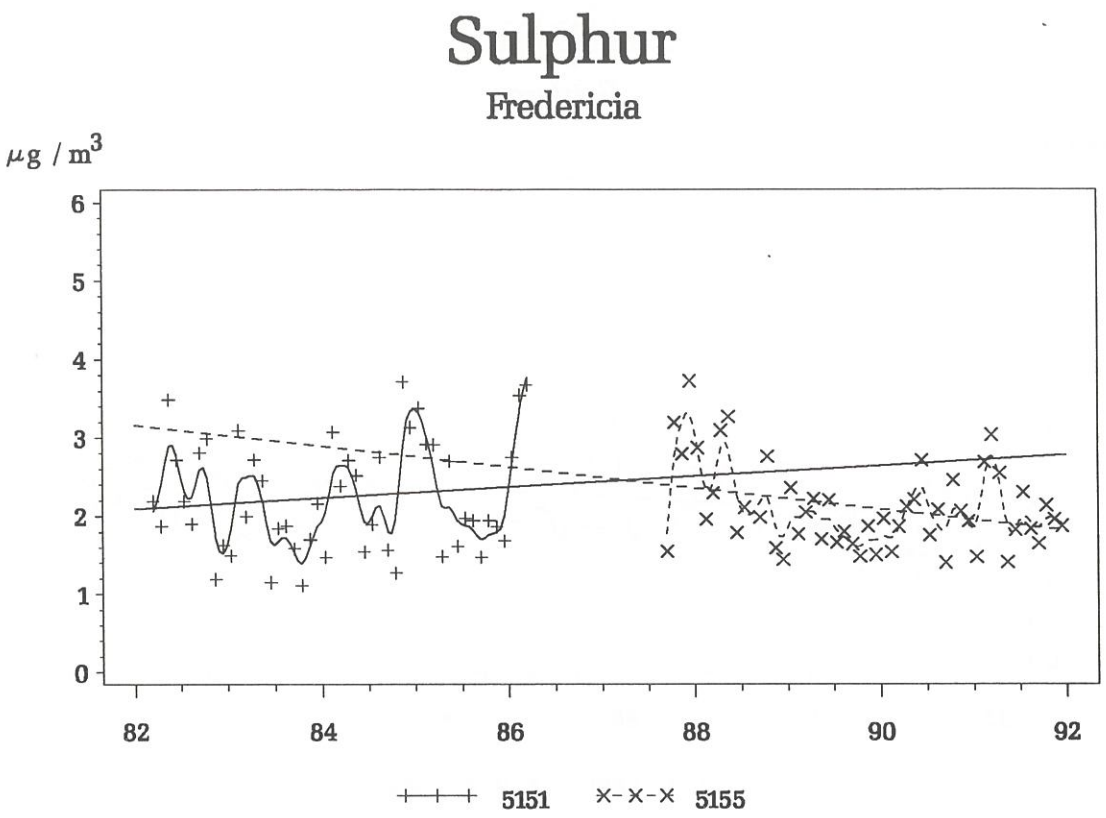


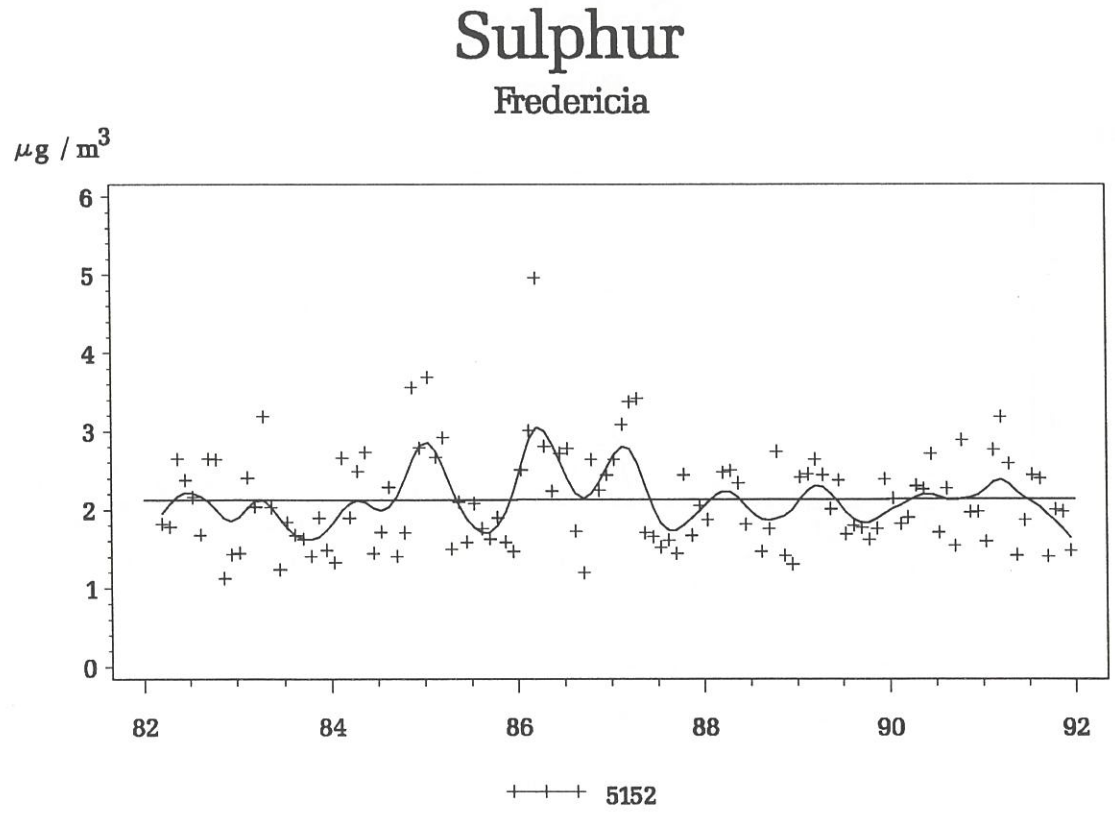
Figur 4.22. Time series for Sulphur (μg/m³) in Copenhagen.



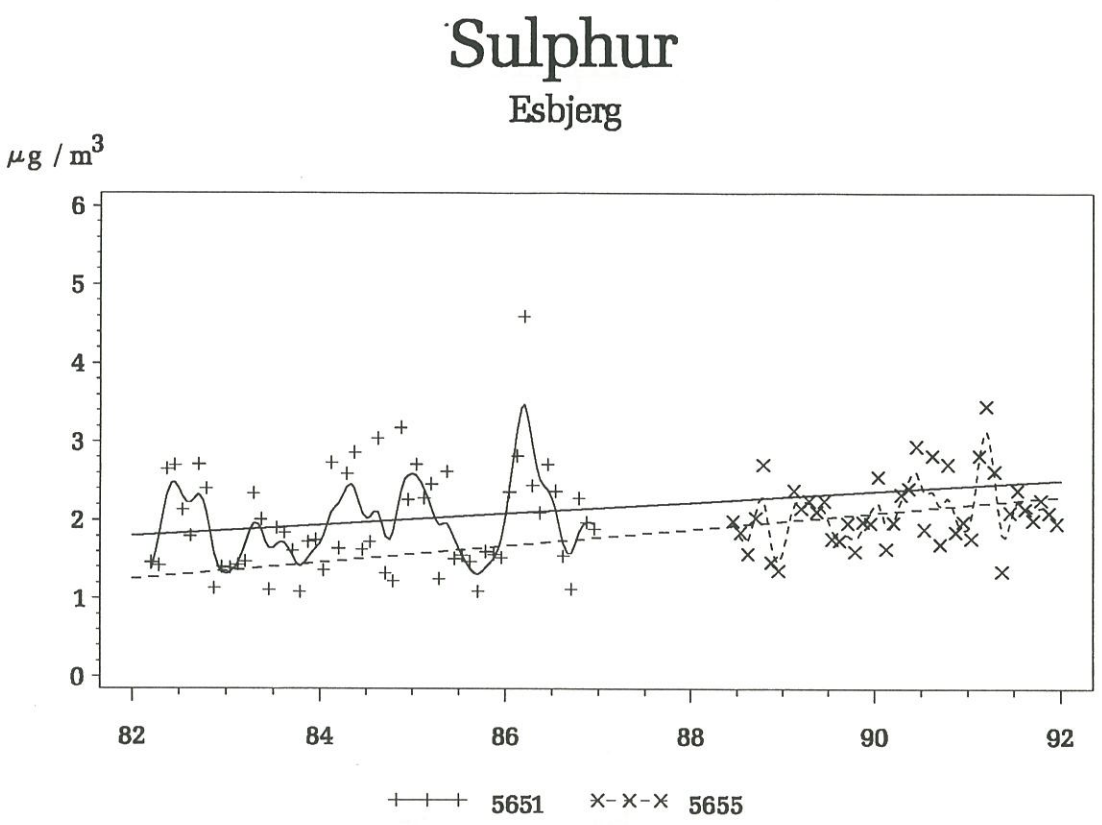


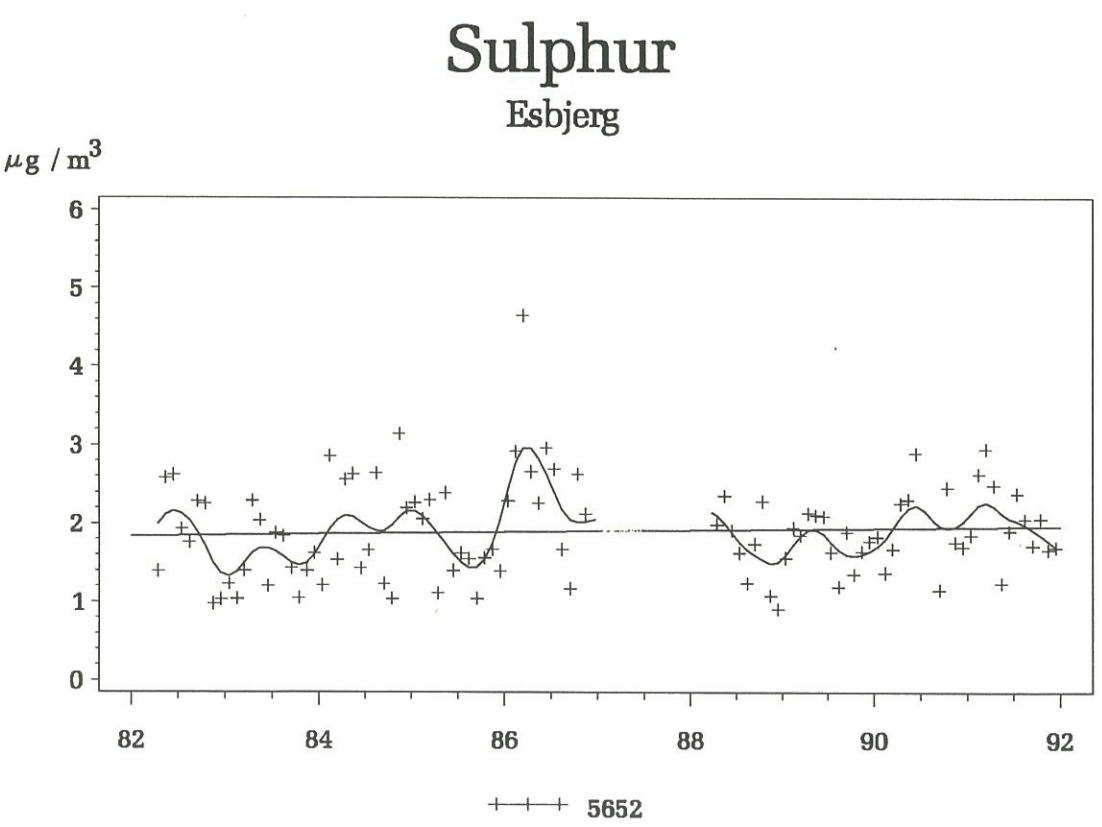
Figur 4.23. Time series for Sulphur (µg/m³) in Copenhagen.



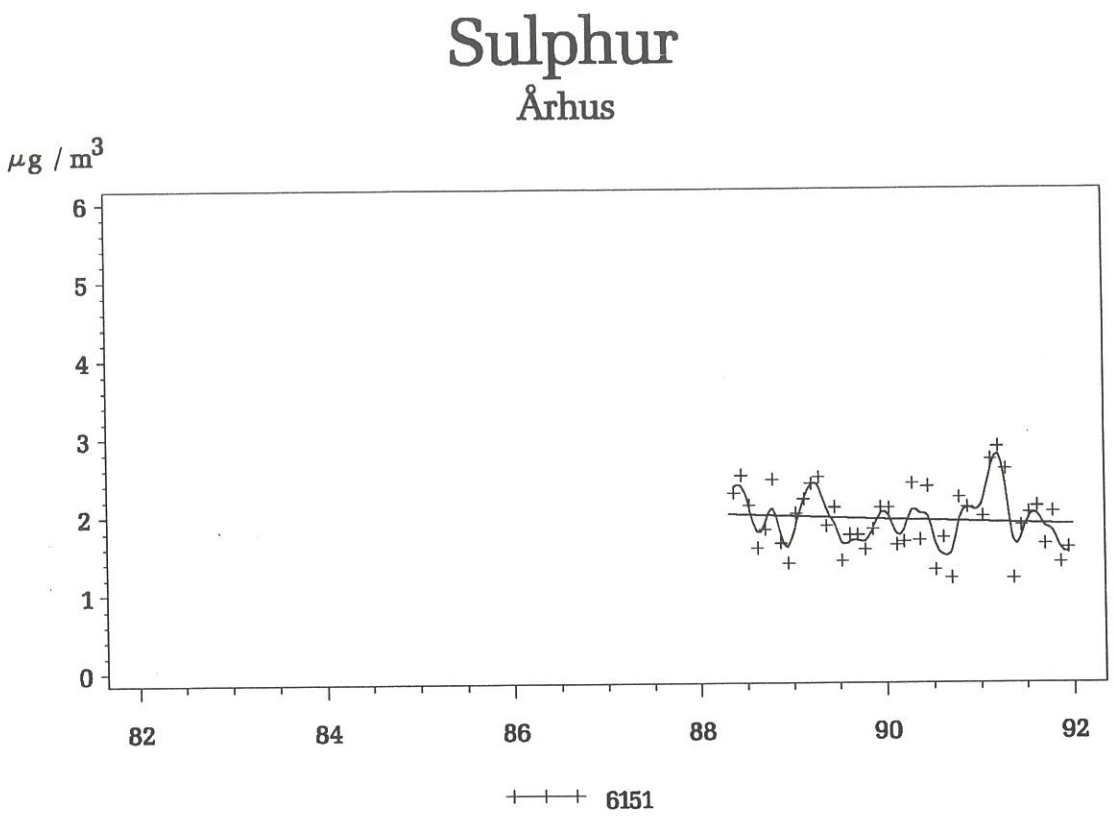


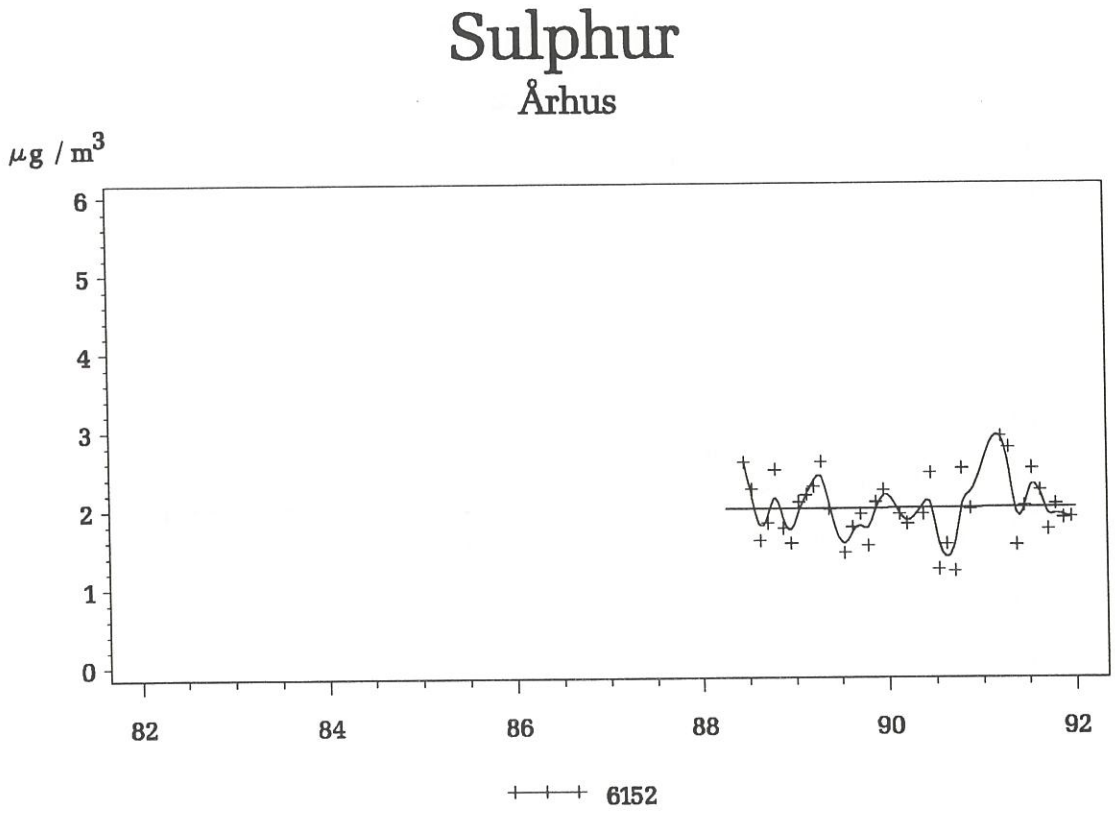
Figur 4.24. Time series for Sulphur (µg/m³) in Fredericia.



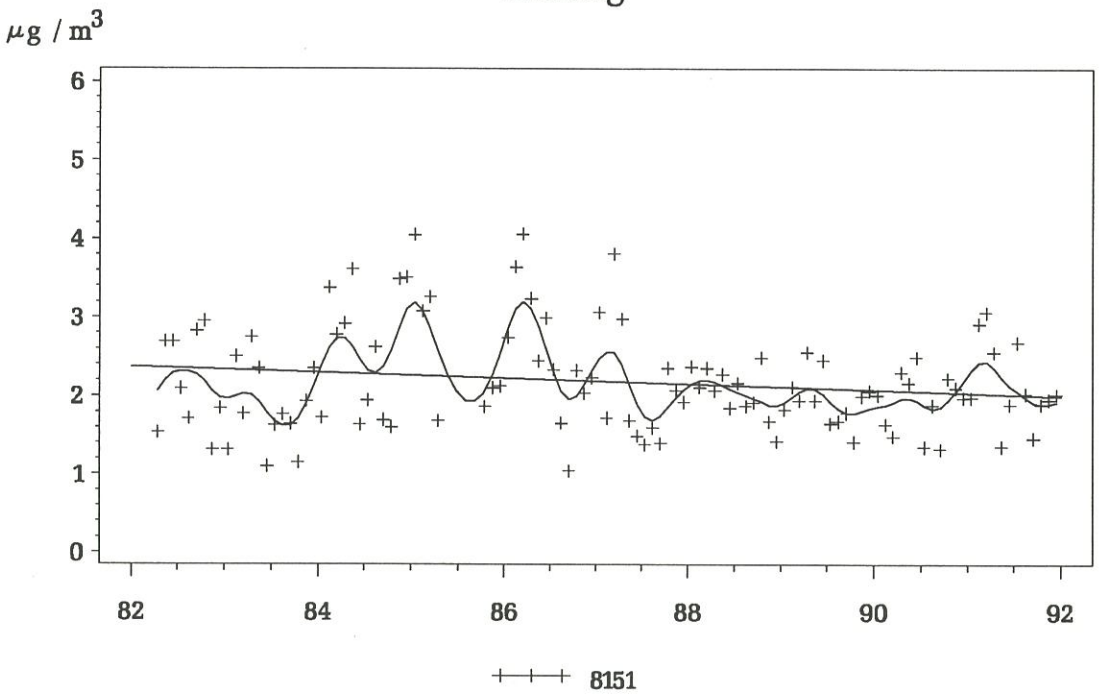


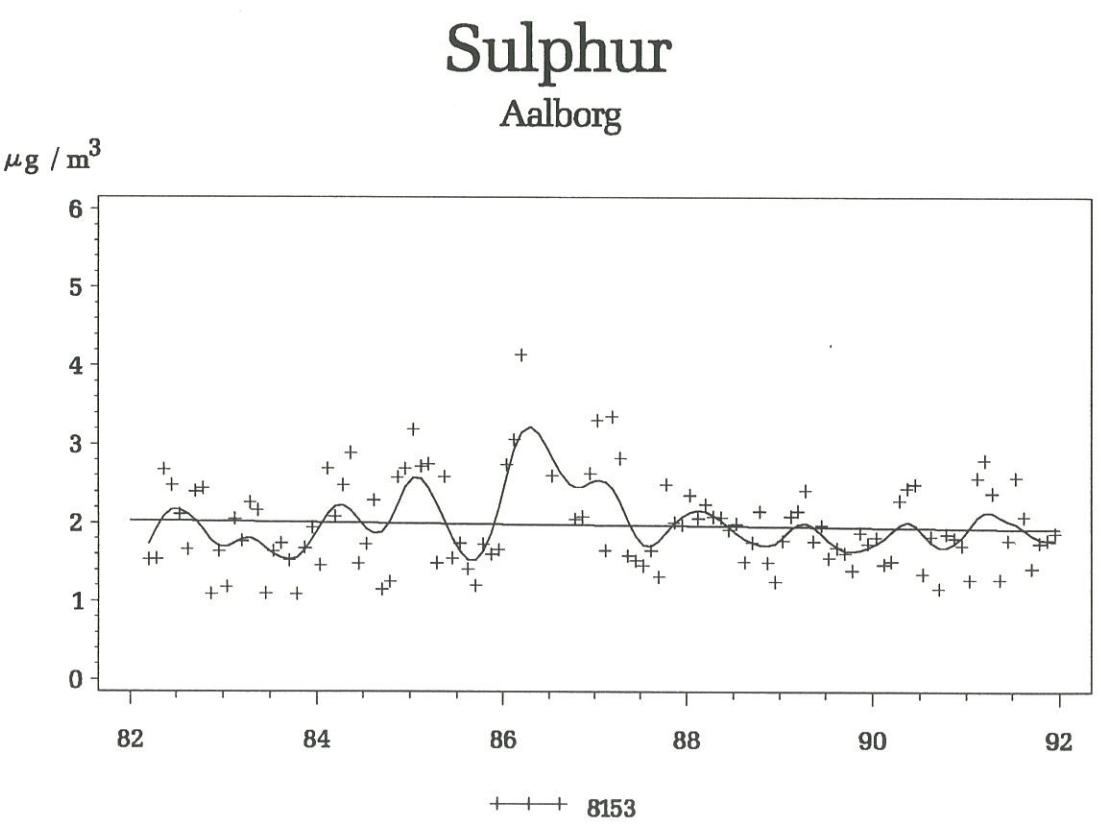
Figur 4.25. Time series for Sulphur (µg/m³) in Esbjerg.



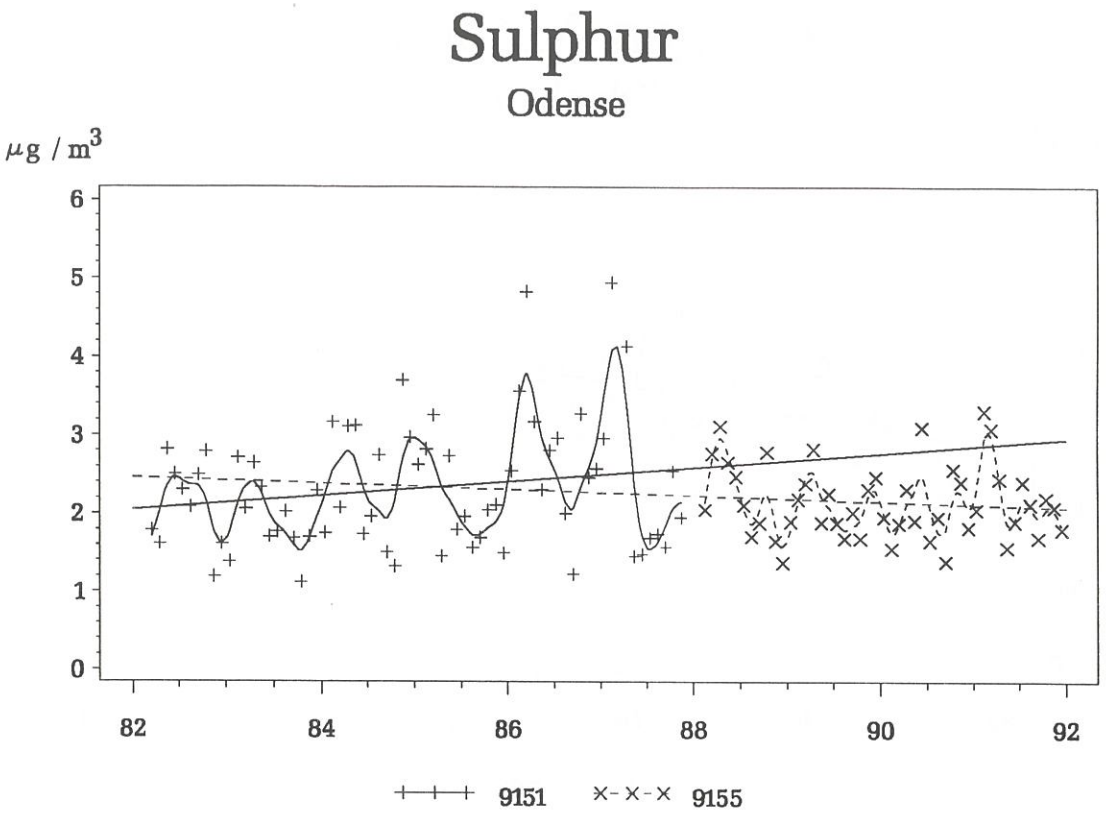


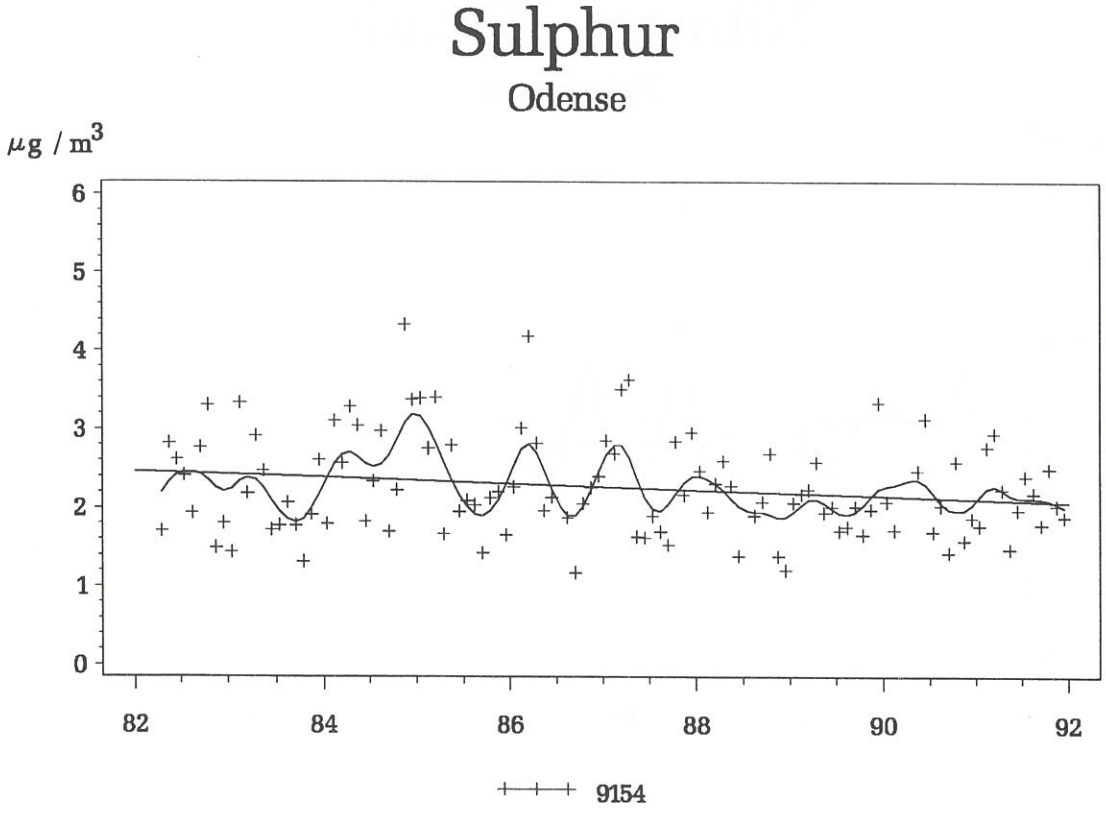
Figur 4.26. Time series for Sulphur (µg/m^3) in Århus.



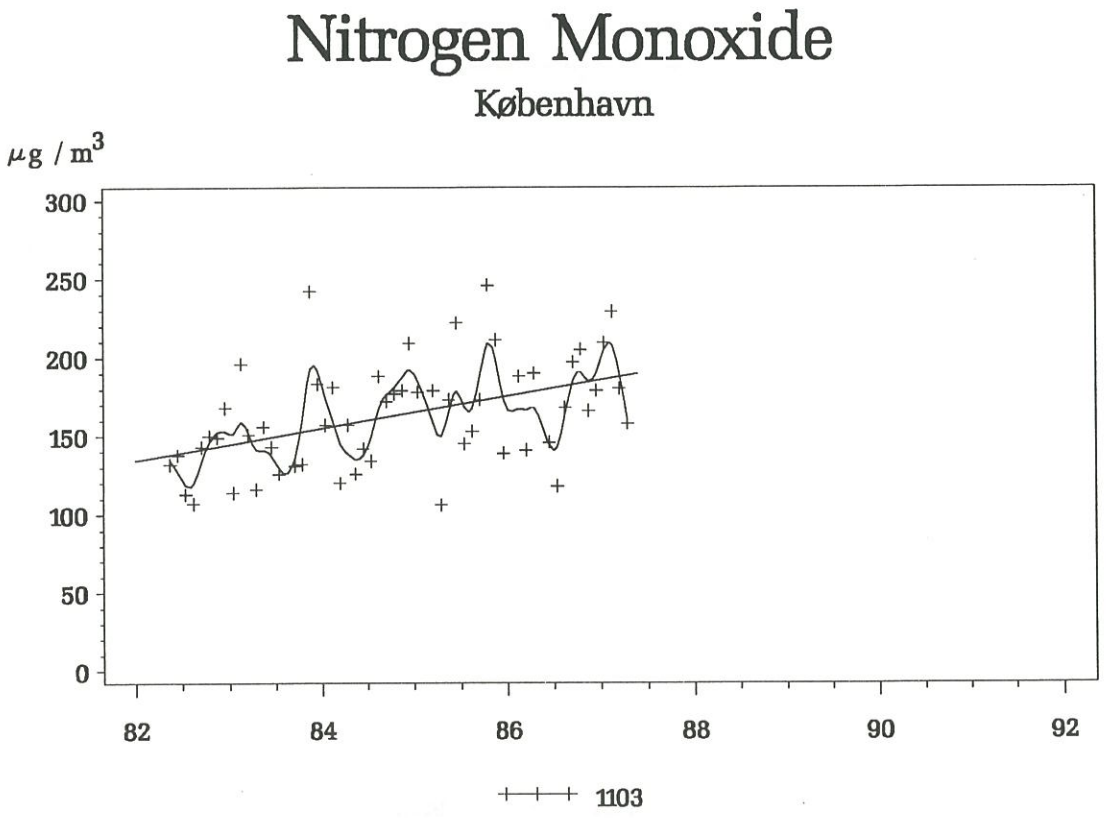


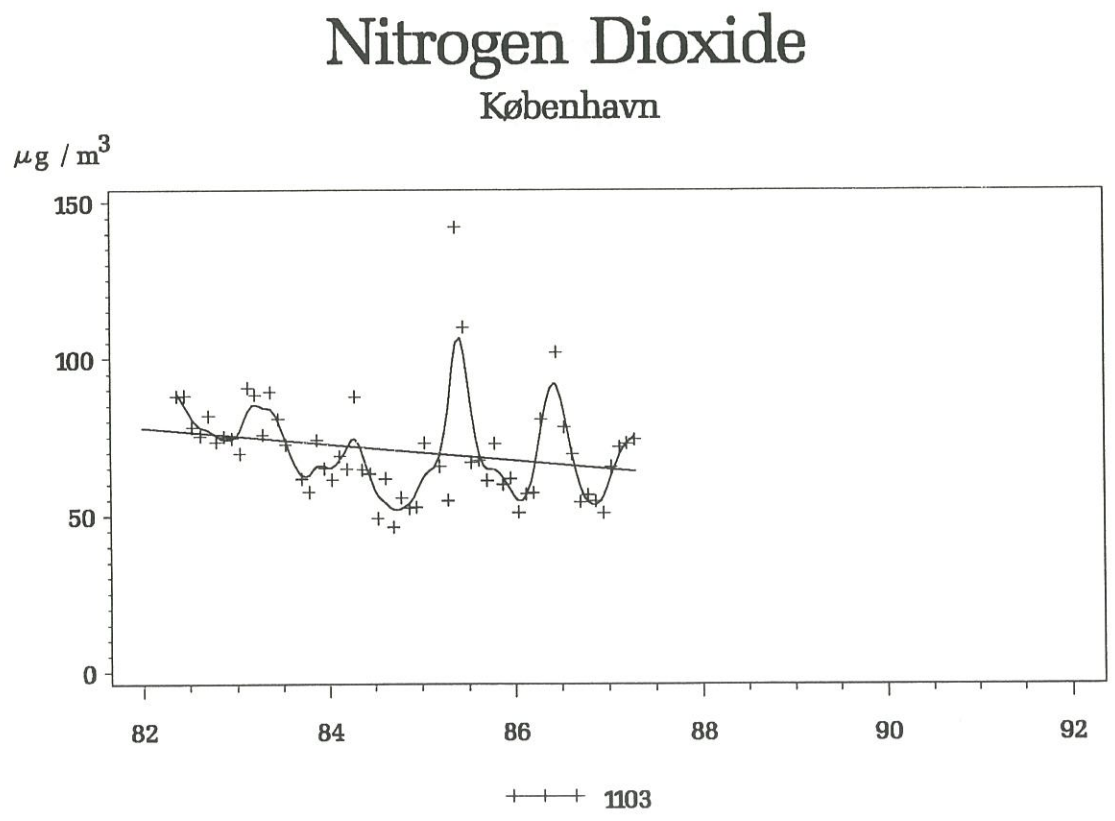
Figur 4.27. Time series for Sulphur (µg/m^3) in Aalborg.



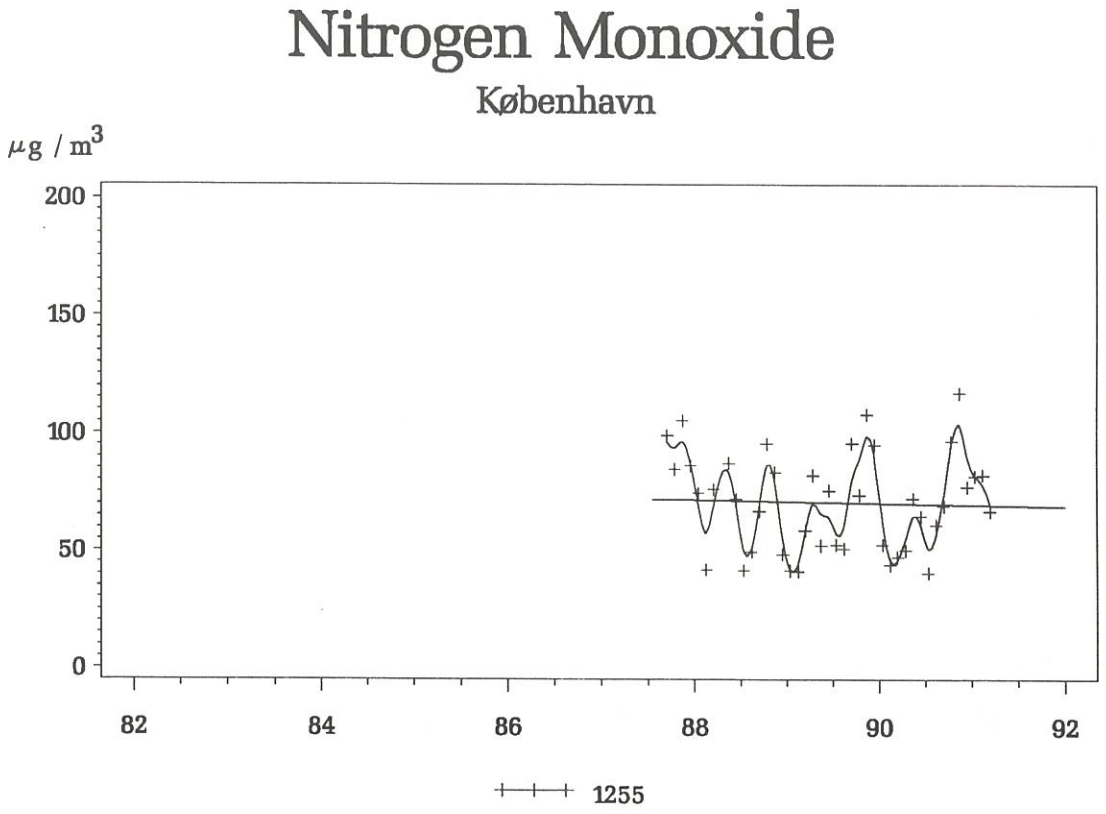


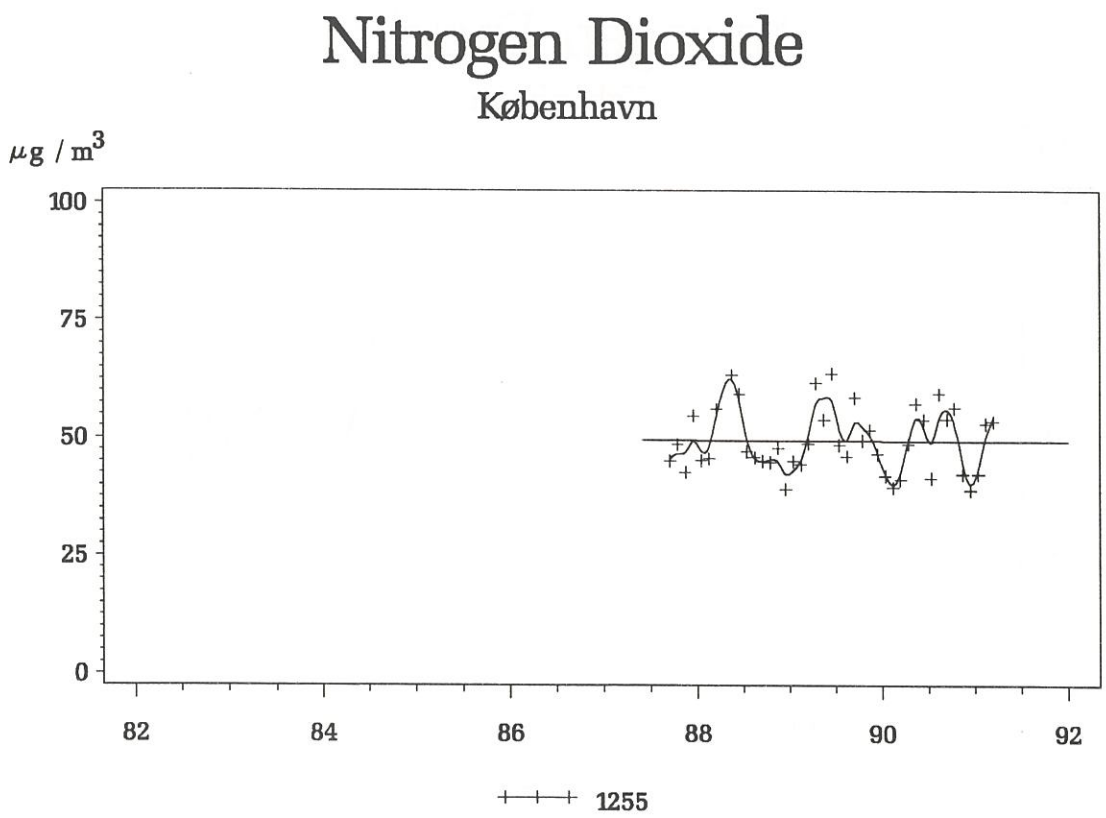
Figur 4.28. Time series for Sulphur (µg/m³) in Odense.



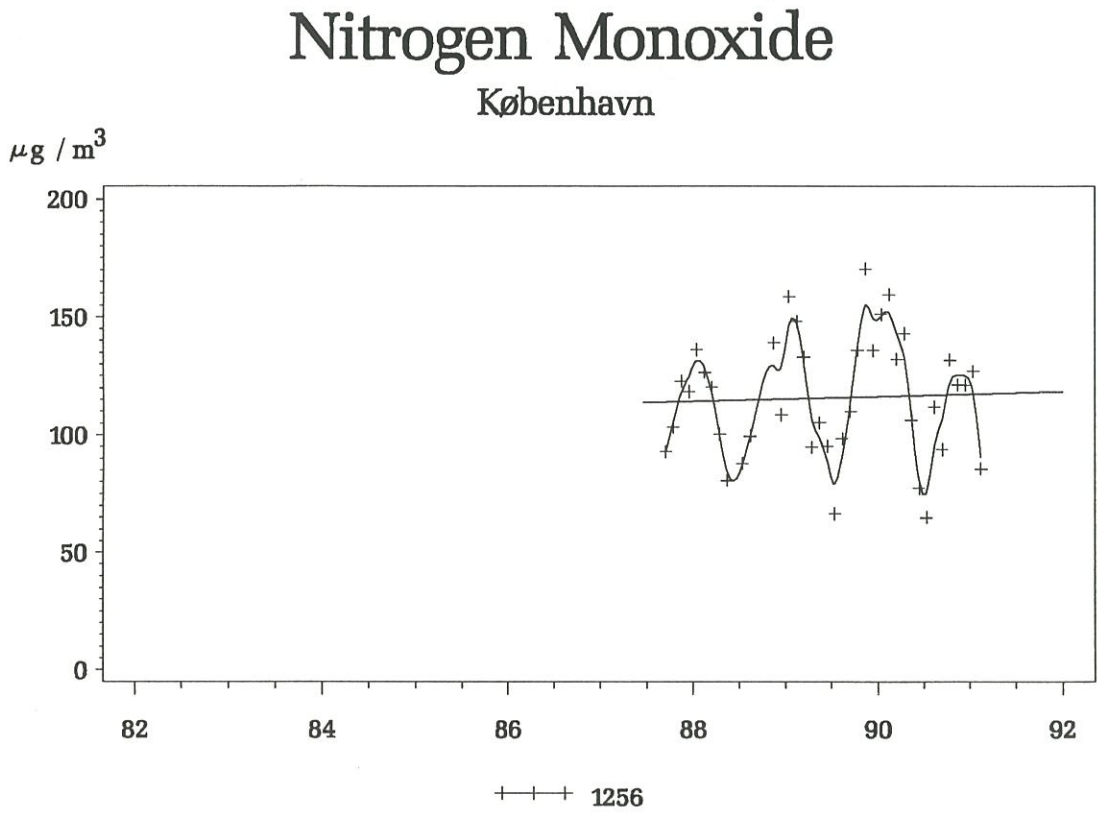


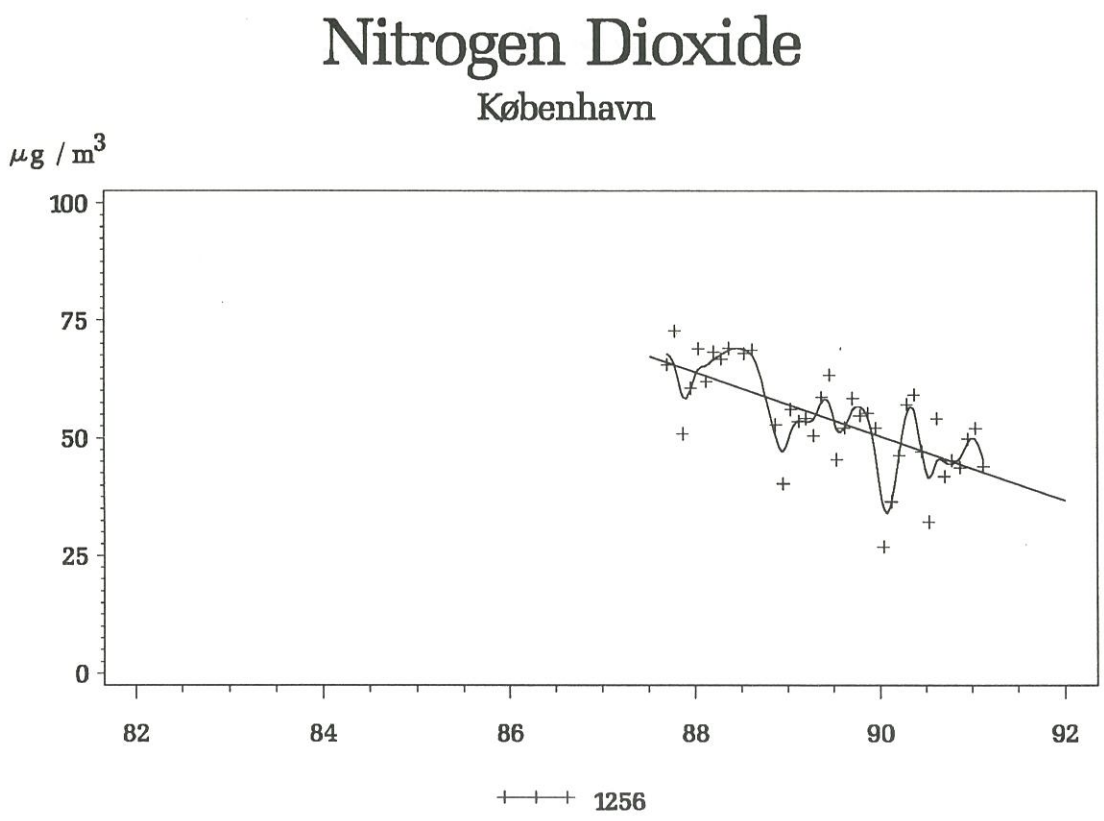
Figur 4.29. Time series for Nitrogen Monoxide and Nitrogen Dioxide (μg/m^3) in Copenhagen.



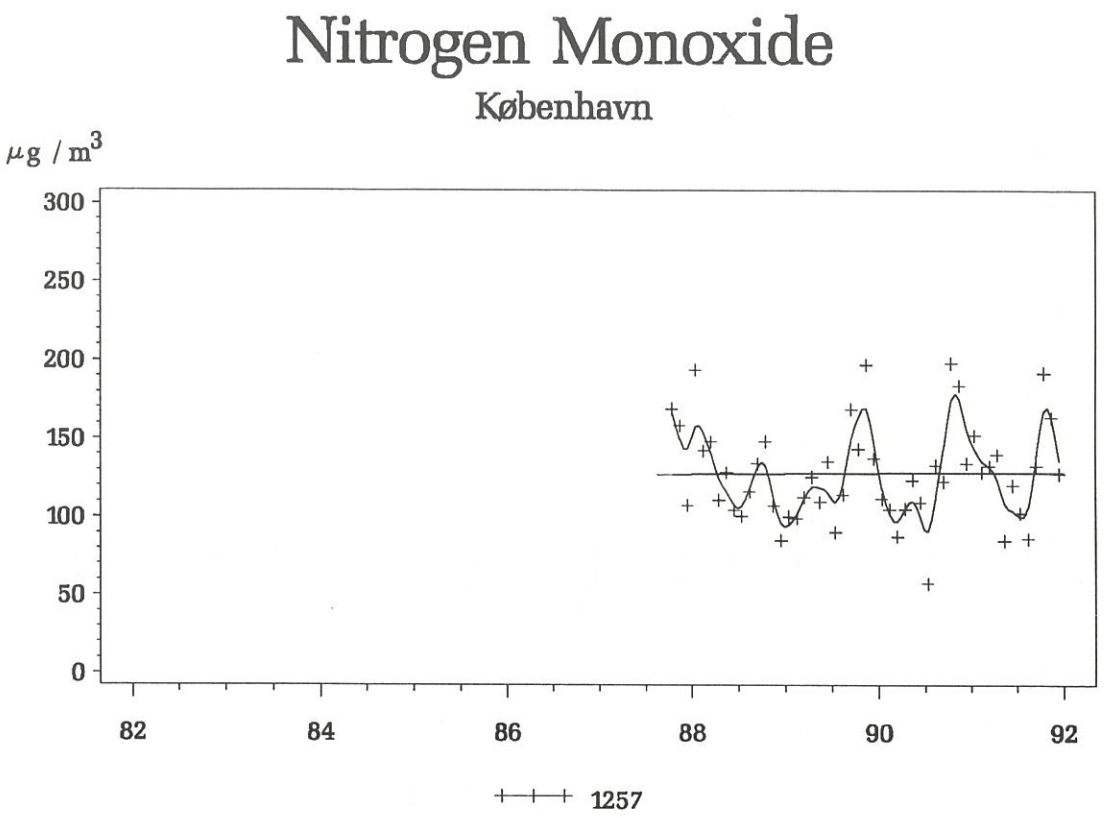


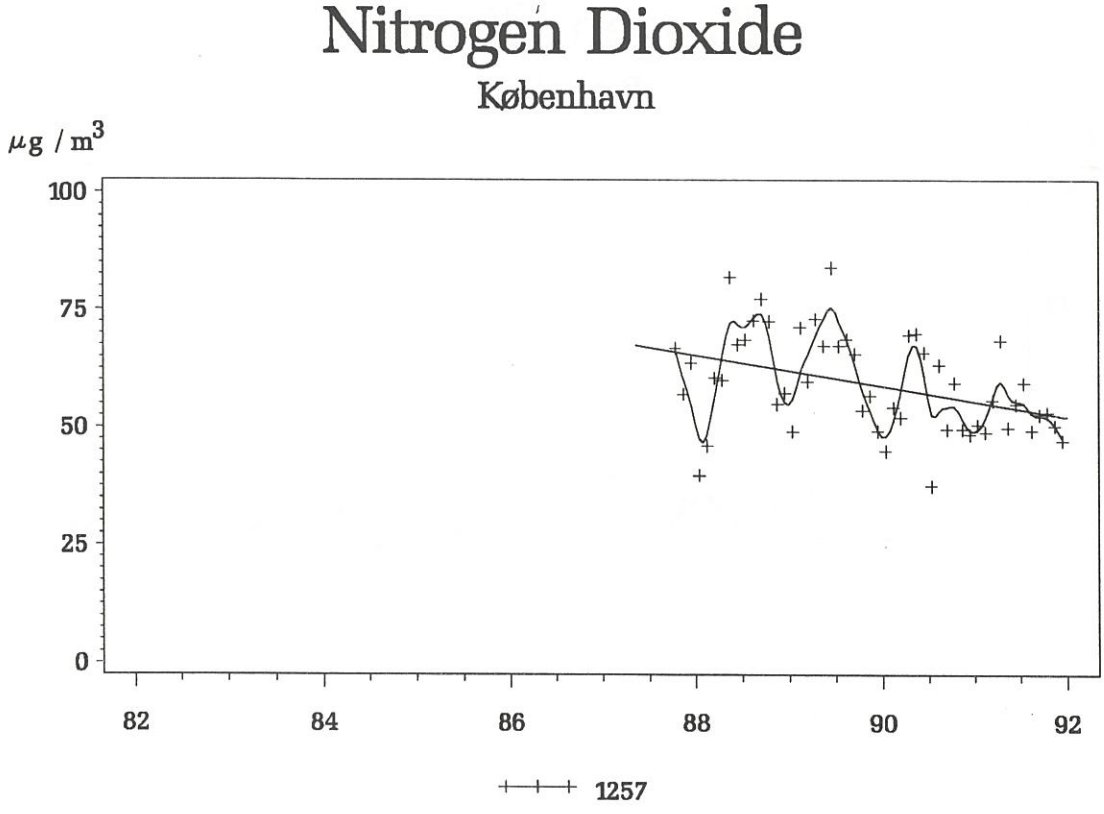
Figur 4.30. Time series for Nitrogen Monoxide and Nitrogen Dioxide (μg/m³) in Copenhagen.



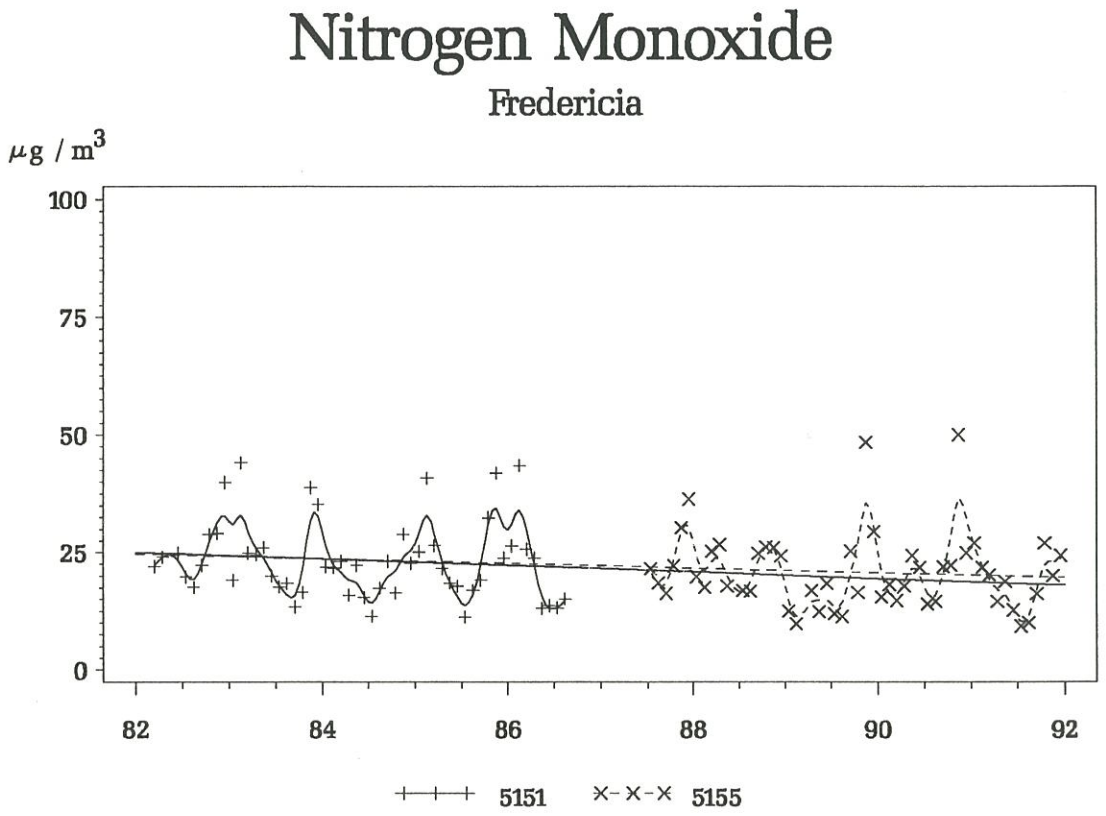


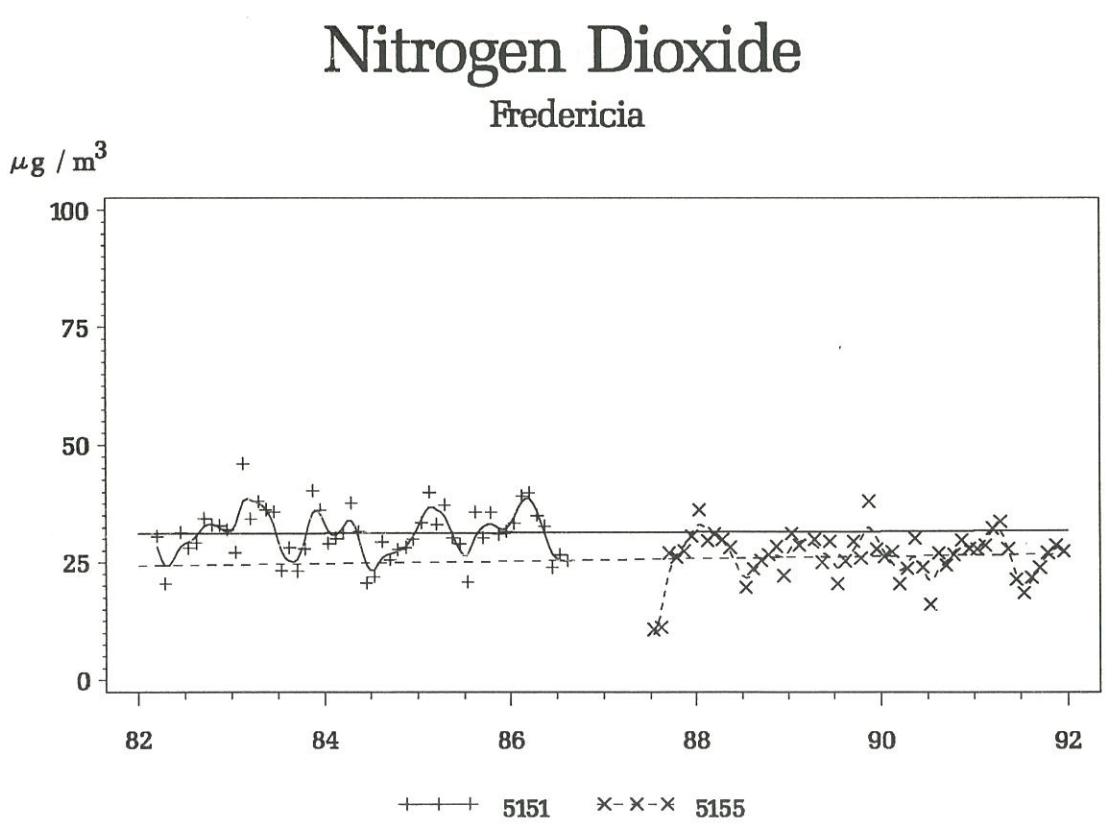
Figur 4.31. Time series for Nitrogen Monoxide and Nitrogen Dioxide (μg/m^3) in Copenhagen.



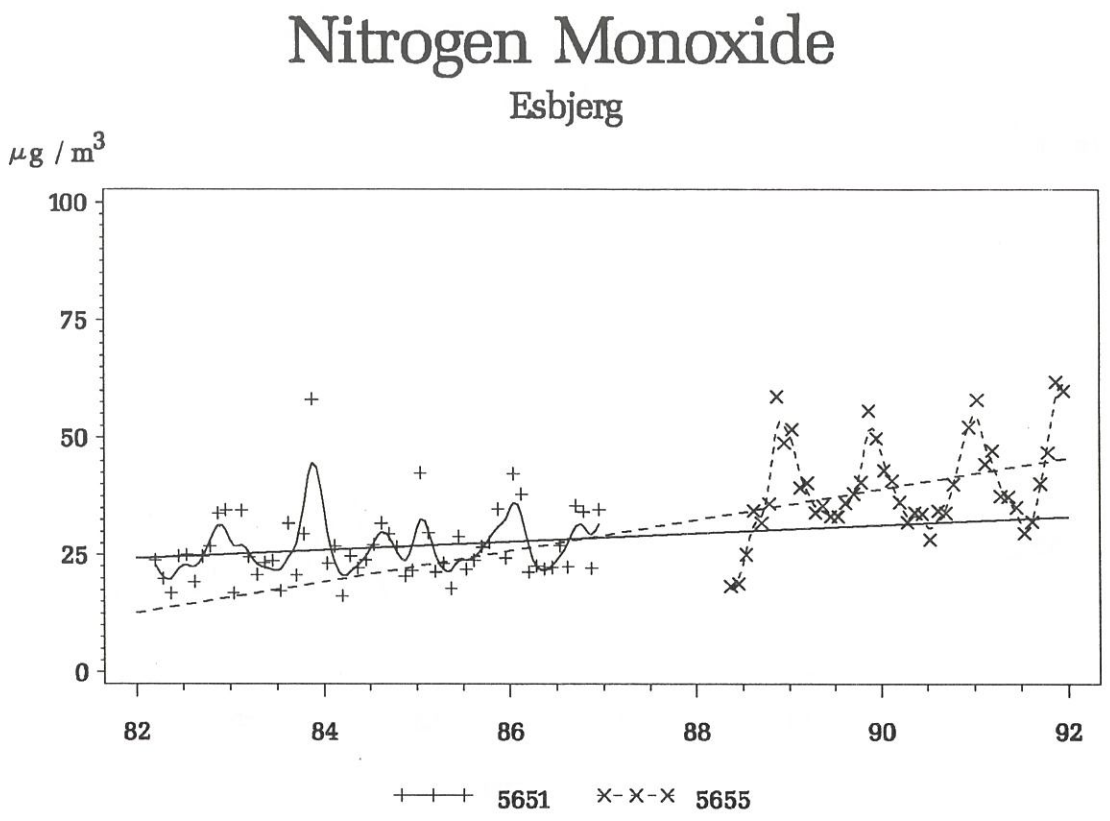


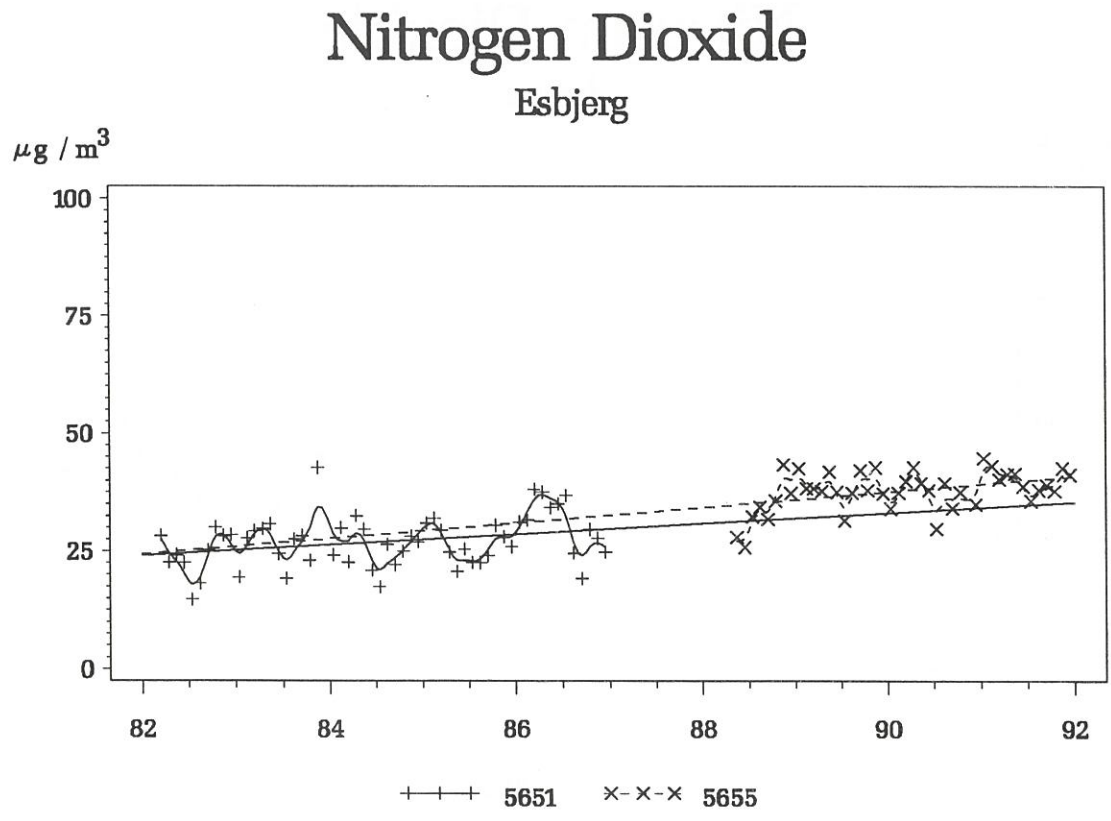
Figur 4.32. Time series for Nitrogen Monoxide and Nitrogen Dioxide (µg/m^3) in Copenhagen.



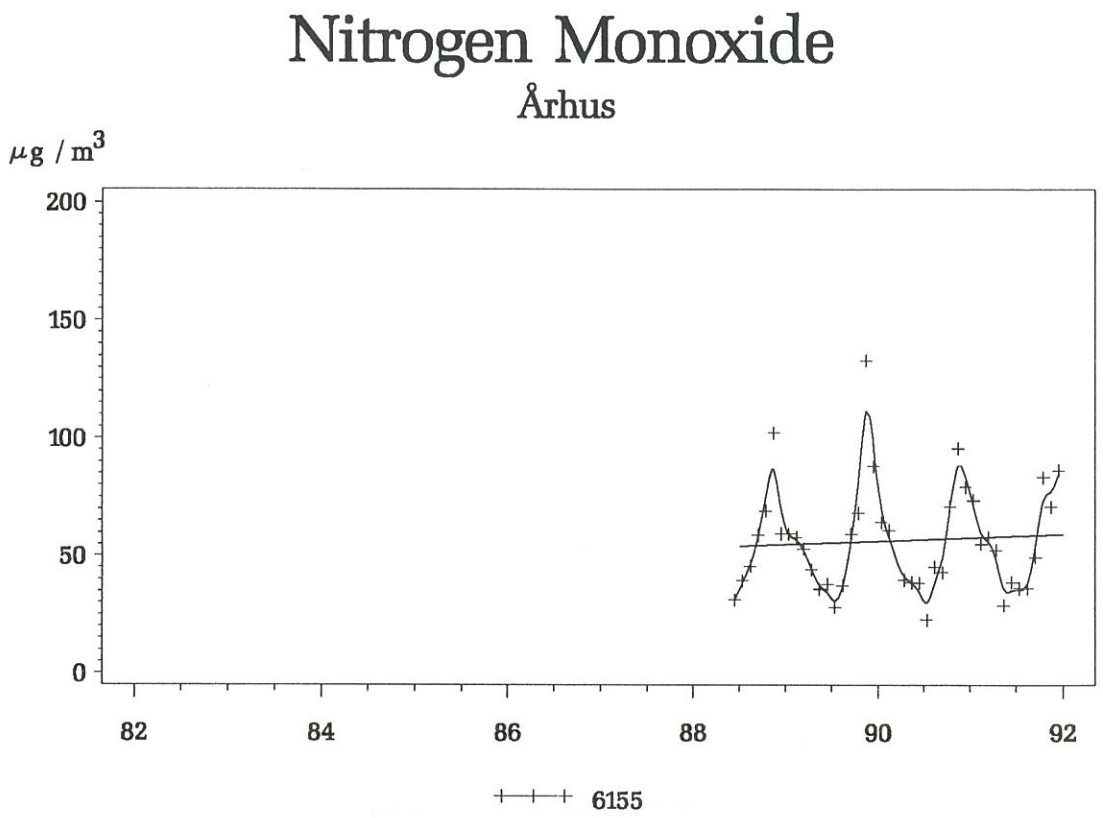


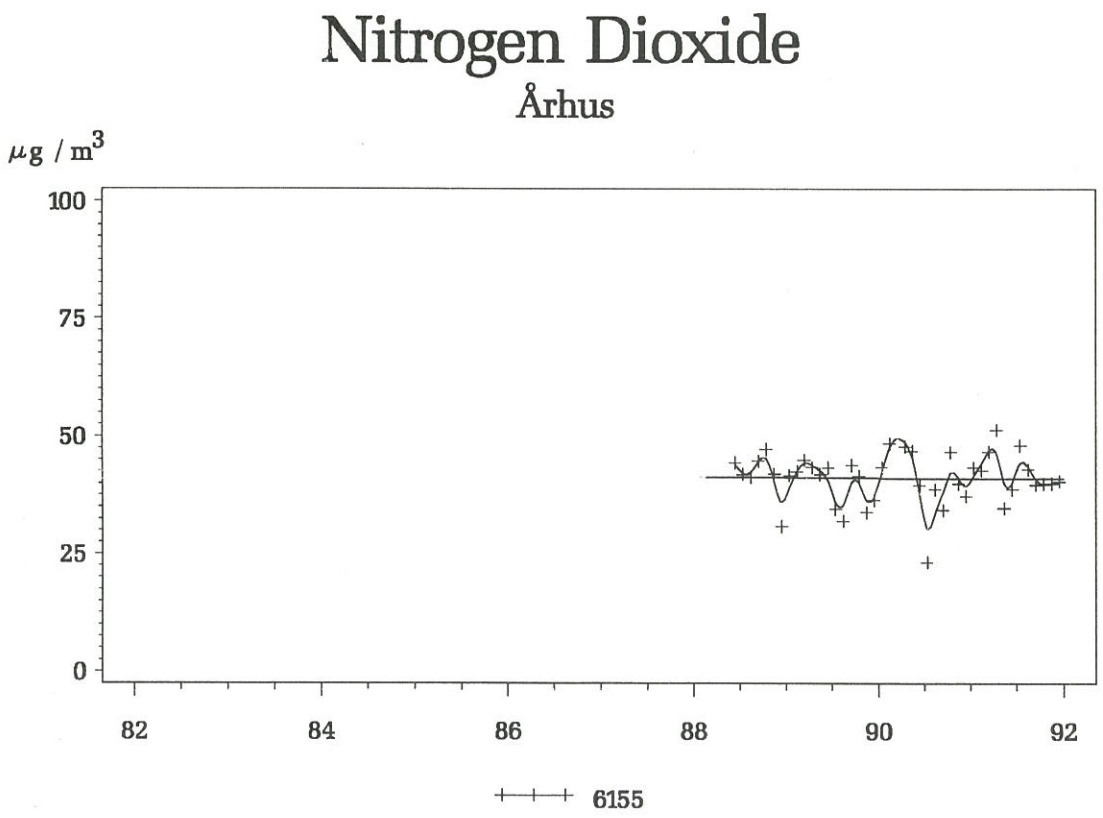
Figur 4.33. Time series for Nitrogen Monoxide and Nitrogen Dioxide (µg/m^3) in Fredericia.



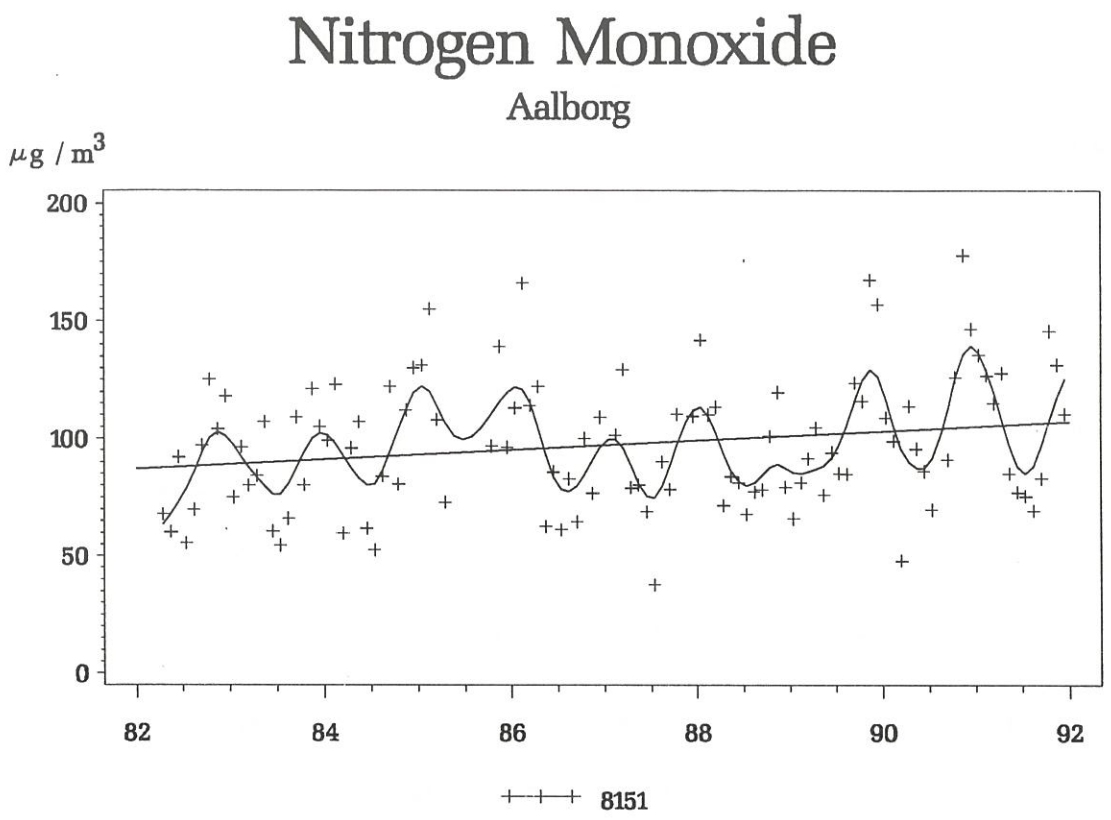


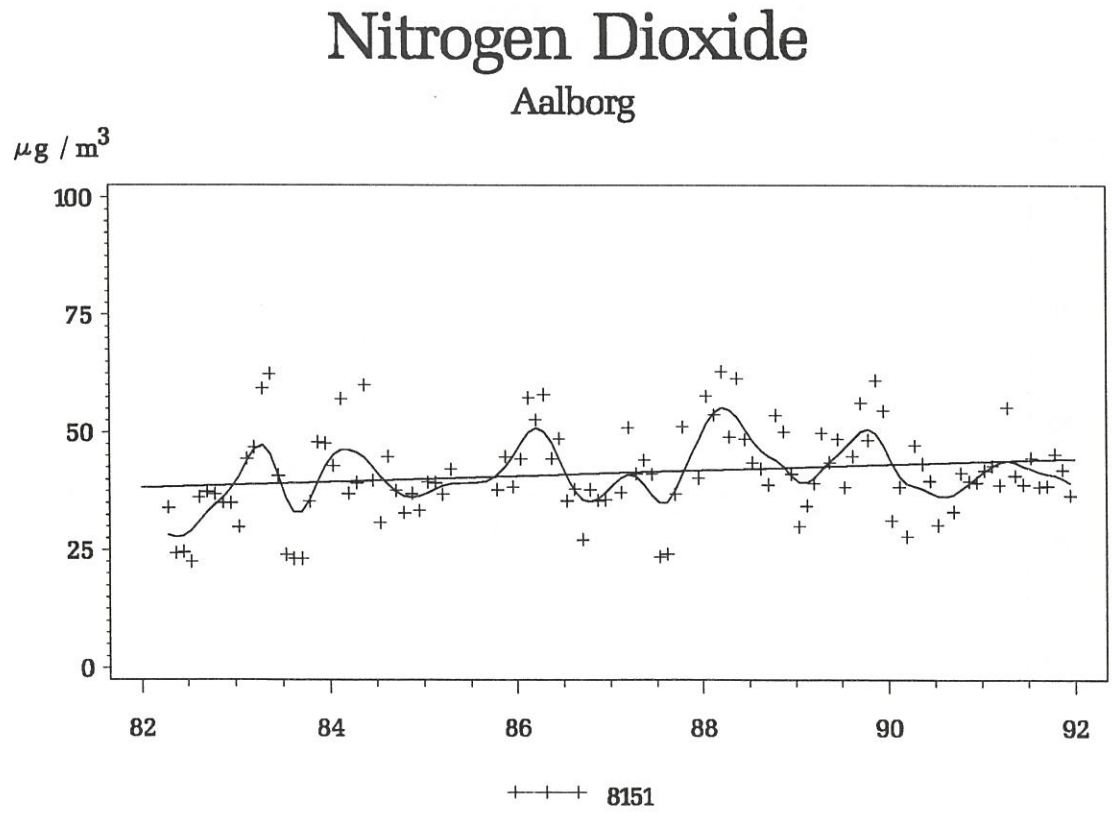
Figur 4.34. Time series for Nitrogen Monoxide and Nitrogen Dioxide (µg/m^3) in Esbjerg.



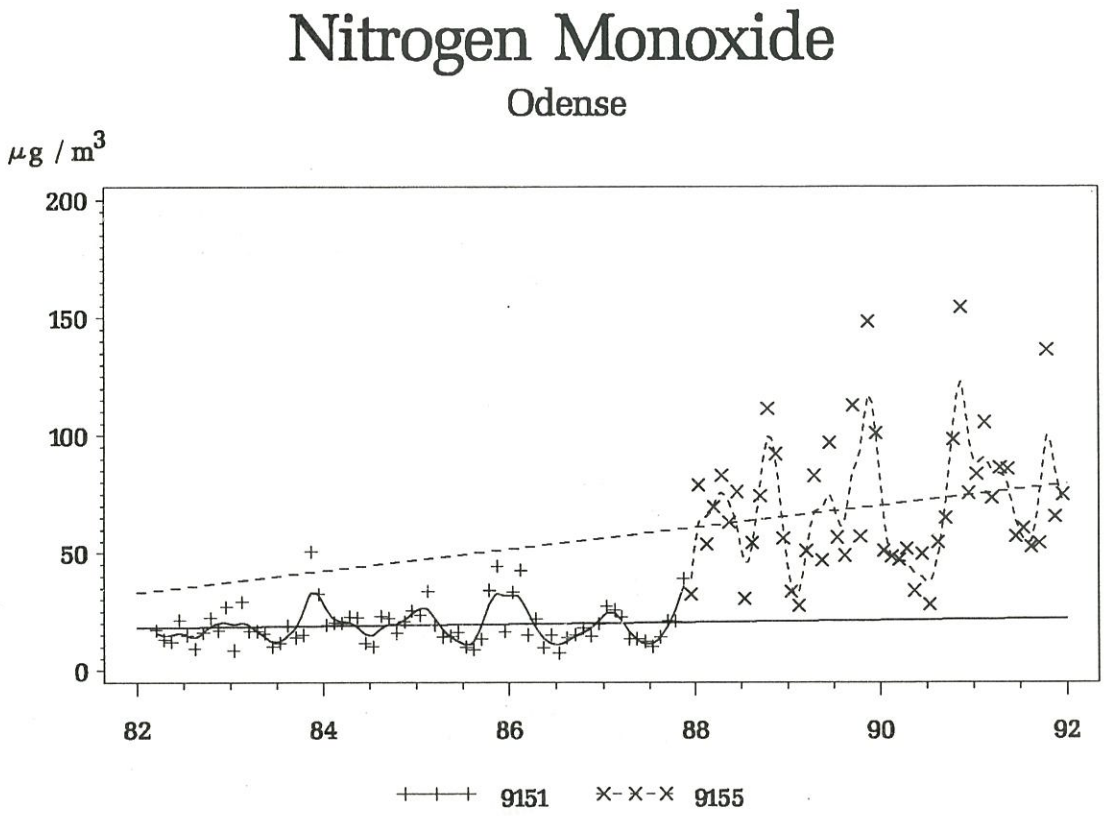


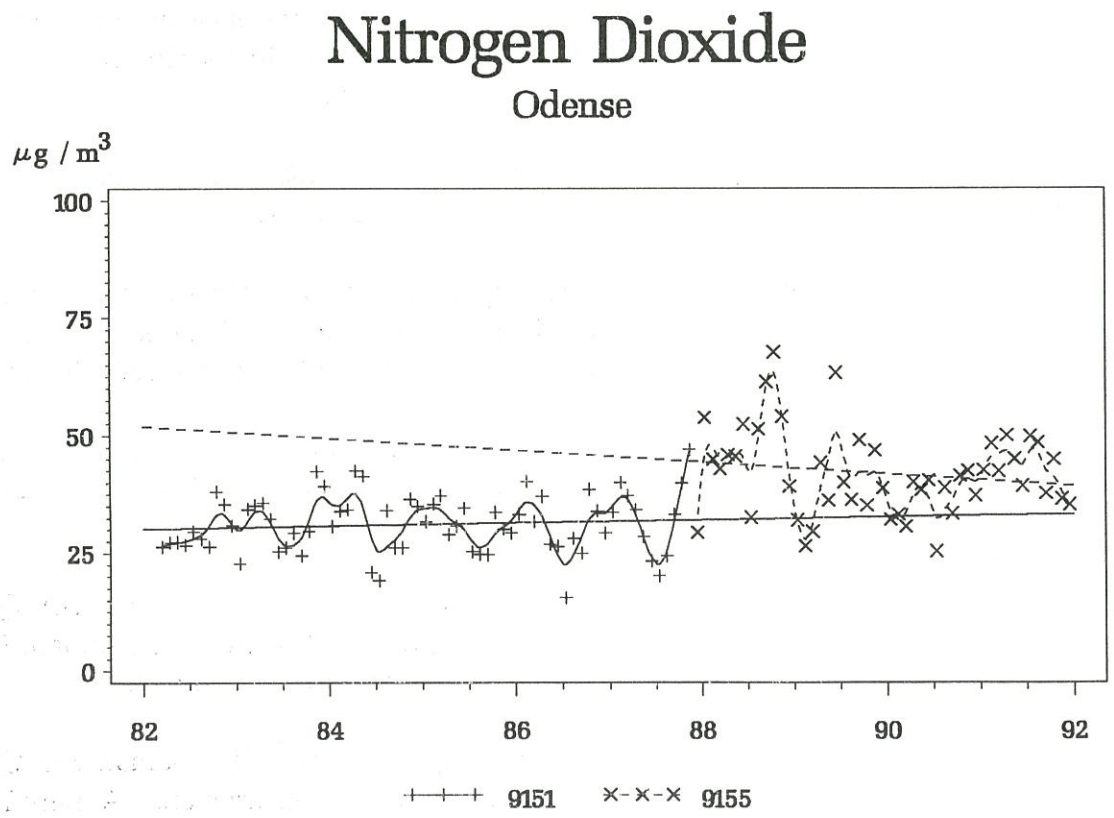
Figur 4.35. Time series for Nitrogen Monoxide and Nitrogen Dioxide (µg/m^3) in Århus.





Figur 4.36. Time series for Nitrogen Monoxide and Nitrogen Dioxide (µg/m^3) in Aalborg.





Figur 4.37. Time series for Nitrogen Monoxide and Nitrogen Dioxide (µg/m^3) in Odense.

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Addresses:

National Environmental
Research Institute
Frederiksborgvej 399
P.O. Box 358
DK-4000 Roskilde
Denmark

Tel: +45 46 30 12 00
Fax: +45 46 30 11 14

Management
Personnel and Economy Secretariat
Research and Development Secretariat
Department of Emissions and Air Pollution
Department of Environmental Ecology
Department of Policy Analysis
Department of Marine Ecology and Microbiology

National Environmental
Research Institute
Vejlsøvej 25
P.O. Box 413
DK-8600 Silkeborg
Denmark

Tel: +45 89 20 14 00
Fax: +45 89 20 14 14

Department of Freshwater Ecology
Department of Terrestrial Ecology

National Environmental
Research Institute
Grenåvej 12, Kalø
DK-8410 Rønde
Denmark

Tel: +45 89 20 14 00
Fax: +45 89 20 15 14

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