Ministry of Environment and Energy
National Environmental Research Institute

The Danish Air Quality Monitoring Programme

Annual Report for 1999

NERI technical Report No. 357
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Data sheet

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Summary

The programme

The Danish Air Quality Monitoring Programme (LMP) was started in 1982 as the first nation-wide urban air pollution monitoring programme in Denmark. The programme has been adjusted to the pollution pattern by two revisions. The present phase (LMP III) was started in 1992. From 2000 a new phase will be started including i.a. measurements of PM_{10} and Benzene at several locations. The present report describes the results from 1999 and updates the trends from the start of the programme in 1982. Measurements are performed at twin sites in the cities of Copenhagen, Odense and Aalborg. One of the sites is at kerb side at a street with heavy traffic and the other is on the roof of a building a couple of hundreds meters from the street site. Two rural sites, one outside Copenhagen and one at Keldsnor in the southern part of the country are also included. NO, NO_2, SO_2, total suspended particulate matter (TSP) and elements in the aerosols are measured at the street sites. O_3 and the meteorological parameters, wind direction, wind speed, temperature, relative humidity and global radiation are measured at the roof sites. Additional measurements of CO at street and NO and NO_2 at roof sites are now performed continuously in order to improve the knowledge about the NO, NO_2 and O_3 problem complex. At the rural site outside Copenhagen the same program is conducted as at the street stations with the inclusion of O_3. Only NO, NO_2 and O_3 are reported from the other rural site.

Limit values

Air quality limit values have been implemented in Denmark for NO_2, SO_2, TSP in order to protect human health. All limit values are based on EU limit values, which also include a limit value for Pb. A set of threshold values for O_3 came into force in March 1994. They were laid down with consideration of the protection of both human health and plants. The new EU Directives introduce revised standards for NO_2, SO_2, particles (PM_{10}) and Pb. They are implemented through the first "daughter" Directive to the Air Quality Framework Directive. It was adopted by the EU council in April 1999. The new limit values shall be in force from January 2001 (18 month after the publication) and all member states must comply with the new limit values before 2005 or 2010 in accordance with the daughter directives. The new limit values were set to protect human health and ecosystems. They are based on the present knowledge about the toxicity of the species. Further EU standards are prepared for O_3, Benzene and the heavy metals Ni, As and Cd.

Nitrogen oxides

The measured NO_2 concentrations were about a factor of two lower than the limit value, while they are close to the values in the new EU Directive. The trend for NO_2 indicates the latest years a weak decrease. The introduction of three way catalytic converters (TWC) on all new petrol driven cars from October 1990 reduces mainly the NO emission. As a result of this the observed NO concentrations are significantly decreasing. O_3 is at present a limiting factor for the formation of NO_2 at street level and it remains to be seen to what

extent the NO_2 concentrations will be reduced at highly polluted locations as result of the TWC's.

Ozone

Some of the threshold values for O_3 were frequently exceeded. The average O_3 concentrations are almost the same at all sites. The average levels are, especially during the winter, lowest at winds from south-easterly directions. The highest peak concentrations were also observed at south-easterly winds. While O_3 is the limiting factor for formation of NO_2 at street level, NO is the limiting factor at roof level and in background areas. After some years with slightly decreasing O_3 concentrations an increase was observed in 1999. This is probably mainly due to the meteorological conditions in 1999.

Sulphur dioxide and TSP

The SO_2 concentrations have been continuously decreasing since 1982. In 1999 they were only about 1/10 of the limit values. They are also far below the new values proposed by the EU commission. The amount of TSP shows a slightly decreasing trend. as a result of better combustion control. The concentrations of TSP were approximately 1/3 of the limit value. The measured values are not directly comparable with the limit values laid down in the new EU Directive. These are based on PM_{10}. Measurements of PM_{10} at one traffic site show concentrations that exceed the proposed "short term" limit value. It is estimated that the PM_{10} concentrations are roughly 75% of the TSP.

Ultra-fine particles

Focus has been on the potential health risk of ultra-fine particles i.e. particles with a diameter <0.1 µm. Measurements at three of the LMP stations shows that these particles constitute a minor part of the total PM_{10} mass, but that the number concentration peaks below 0.1 µm. The traffic is the main source for these particles in urban areas. Diesel driven cars have the highest emission; but the gasoline cars contribute with comparable amounts.

Heavy metals

The Pb pollution has been reduced with about a factor of 50 since 1982 as a result the removal of Pb from gasoline. The development has outdated the limit value, which is more than a factor of 100 higher than the measured concentrations. Also the proposed new limit value is far above the measured concentrations in Denmark. The measured concentrations for Ni, As and Cd are well below the expected new limit values. The traffic is no longer then main source for Pb, but other heavy metals as Cr and Cu, are mainly traffic related in urban areas. The main source is probably dust from brake pads.

Carbonmonoxide and benzene

CO and benzene are mainly emitted from petrol driven cars. The concentrations are consequently highest close to the streets. The TWC remove the main part of both CO and benzene from the exhaust. The concentrations are at present steadily decreasing, due to the increasing fraction of cars with TWC's and reduction of the benzene content in petrol. A new EU Directive, which sets limit values for CO and benzene, is in preparation. The measured CO concentrations are well below the expected limit values, while the present benzene concentrations at some places exceed the expected limit value, which should be met before 2010.

1 Introduction

LMP III

The third Danish Air Quality Monitoring Programme (LMP III) was started in 1992. The programme comprises an urban monitoring network with stations in three Danish cities. The results are used for assessment of the air pollution in urban areas. The programme is carried out in co-operation between the National Environmental Research Institute (NERI), the Danish Environmental Protection Agency, the Greater Copenhagen Air Monitoring Unit and the County of Funen (for the city of Odense) and the Municipality of Aalborg. NERI is responsible for the practical programme in co-operation with the Agency of Environmental Protection for the City of Copenhagen, the county of Funen, and the Department for the Environment and Urban Affairs, Aalborg. The results are currently published in quarterly reports in Danish and they are summarised in annual reports in English. Statistical parameters and some actual data are accessible at the Internet at the address www.dmu.dk/AtmosphericEnvironment/netw.htm. Selected data are also available at tele-text, Danish National Television.

Previous programme

The programme was revised considerably during 1992 compared to the previous phase (LMP II) (Palmgren, Kemp, Manscher 1992). The installation of the new equipment took place during 1992 and the beginning of 1993. All instruments were updated as planned during 1993 (Kemp 1993) and are now running continuously except for short interruptions due to technical problems.

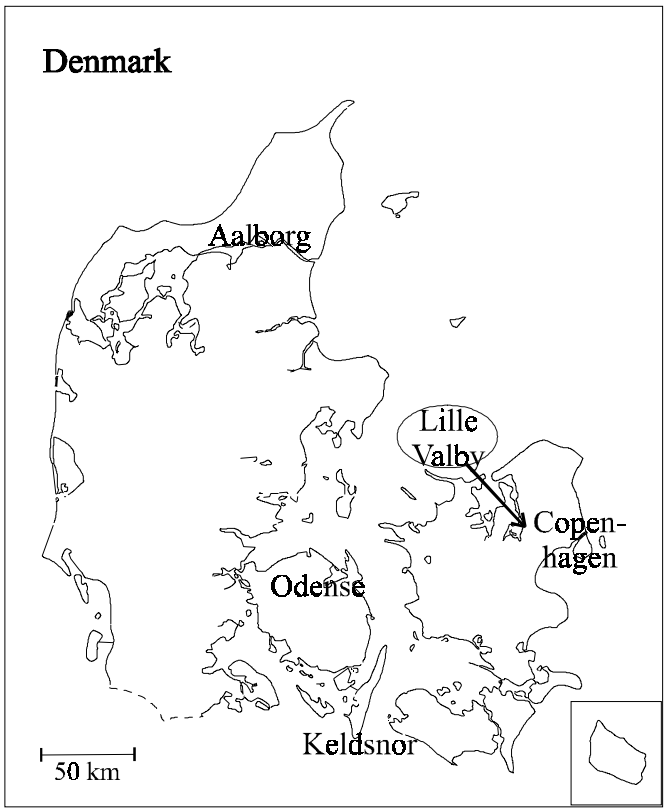


Figure 1.1 Cities and the background sites in the LMP network

Table 1.1 LMP III stations in 1999. TSP is the total suspended particulate matter collected on a filter and determined by weighing. The station type refers to the classification given in Kemp, 1993. BTX is measurements of benzene, toluene and xylene with a monitor. The meteorological measurements comprise wind direction, wind speed, ambient temperature, relative humidity and global radiation.

		Measuring Programme	
	Station type	½ hour average	24 hour average
Copenhagen/1257	Main (Traffic)	NO, NO_2, CO, O_3, BTX	SO_2, PM10, TSP, Elements
Copenhagen/1259	Roof (Urban background)	(O_3), meteorology,NO,NO_2	-
Odense/9155	Main (Traffic)	NO, NO_2, CO	SO_2, TSP, Elements
Odense/9154	Additional (Traffic)	-	SO_2, TSP, Elements
Odense/9159	Roof (Urban background)	O_3, meteorology, NO,NO_2	-
Aalborg/8151	Main (Traffic)	NO, NO_2, SO_2, CO	SO_2, TSP, Elements
Aalborg/8159	Roof (Urban background)	O_3, meteorology, NO,NO_2	-
Lille Valby/2090	Rural background	NO, NO_2, O_3	SO_2, TSP, Elements
Keldsnor/9055	Rural background	NO, NO_2, (O_3)	-

Measurement programme

The measurement programmes at stations in operation during the major part of 1999 are shown in table 1.1. The map (figure 1.1) shows where the sites are located. All sites and measurement methods are described in Kemp, 1993 and NERI, 1998. The ½-hour measurements of SO_2 at Copenhagen/1257, Odense/9159 and Lille Valby/2090 as well the monitor measurements of O_3 at Copenhagen/1259 were closed during 1999. At Copenhagen/1259 O_3 is measured with DOAS. Year 1999 is the last in the LMP III.

Annual statistics, trends, phenomenology

The annual statistics and episodes are summarised for all groups of species. The results are compared with Danish limit and guide values and WHO guideline values. At present limit values are set for SO_2, suspended particulate matter, NO_2 and Pb in Denmark. A set of threshold values for O_3 was introduced in 1994 by the implementation of an EU directive (EEC 1992). The trends since the start of the first LMP programme in 1982 are illustrated using results from Aalborg/8151 and Odense/9154. These are the only stations, which have been in operation since 1982.

Smog warning

A permanent smog warning system including NO_2, SO_2 and O_3 was introduced from the beginning of 1994. For NO_2 and SO_2 warnings will be transmitted, if the concentrations exceed 350 µg/m^3 for more than three consecutive hours and no immediate decrease is expected. According to the ozone directive (EEC 1992) information will be broadcasted, if the hourly average concentration of O_3 exceeds 180 µg/m^3. An alarm will be broadcasted, if the hourly average concentration exceeds 360 µg/m^3.

1999 reports

The 1999 results are found in quarterly reports (Danmarks Miljøundersøgelser 1999a, 1999b, 2000a 2000b). The results obtained

during 1999 are summarised in the present report in form of annual statistics and trends. Results for CO and benzene, toluene and xylene, which cover a whole years measurement, has been included. A overview of results from the LMP programmes together with a description of model calculations and an evaluation of the health aspects is found in Palmgren et al., 1997. A description of the Danish air quality monitoring programmes and selected results are shown on the internet (NERI 2000).

New limit values implemented by the EU commission

The present Danish limit values are almost identical with the limit values laid down in the EU directives (EEC, 1982, 1985, 1989, 1992). These limit values and the monitoring techniques and strategies are under revision. The new EU legislation consists of a framework directive (EC 1996), giving general rules for network design and limit value strategies, and a number of daughter directives giving limit values, reference methods and monitoring strategies for specific pollutants. The limit values will primarily be based on the known health effects of the species. The framework directive has already been adopted. The first daughter directive, including limit values for NO_2, SO_2, particulate matter (PM_{10}) and lead, was adopted in April 1999 (EC 1999) by the EU council. The EU Commission is preparing directives covering O_3, benzene, CO and the heavy metals Cr, As and Cd. The new limit values in relation to the measured concentrations are discussed for the single species in the following chapters.

LMP IV

During 2000 the LMP IV program will be implemented. It will be designed to meet the requirements of the new EU directives. More focus will be put on organic pollutants and particulate matter in the air.

Other air quality networks in Denmark

Beside the LMP two other air quality monitoring networks are in operation in Denmark. The Greater Copenhagen Air Monitoring Unit is responsible for a network in the Greater Copenhagen area. A number of pollutants are measures at five sites. The measurements are comparable with the LMP measurements and the two programmes supplement each other in Copenhagen (HLU 1999). A network in rural areas (the Danish Background Monitoring Program) was established in 1978. At present gas and aerosol measurements are performed at six stations while various ions are determined in precipitation collected at 12 sites. The aim is i.a. to study acidification and eutrofication of the forests, farmland, Danish sea and freshwater areas (Ellermann et al. 2000).

2 Nitrogen Oxides

Source

The term NO_x denotes usually the sum of NO and NO_2. NO_x is emitted from combustion processes. The most important sources in Denmark are motor vehicles and power plants. The main part of the direct emission from traffic consists of NO (more than 90%). The emitted NO is oxidised in the atmosphere to NO_2 and further to HNO_3/NO_3 (nitrate), or e.g. PAN. If the O_3 concentration is sufficiently high the conversion of NO takes place almost instantaneously, whereas the reaction time for the formation of HNO_3/NO_3 is of the order of several hours. The exact reaction rates depend very much on the actual concentrations, the photochemical activity and the temperature.

Measurements

During 1999 continuous measurements of NO and NO_2 were performed at all stations except Odense/9154. More than 90% of the possible results are valid for all stations except the two in Aalborg, where only about 70% are available (see table 2.1).

2.1 Annual statistics

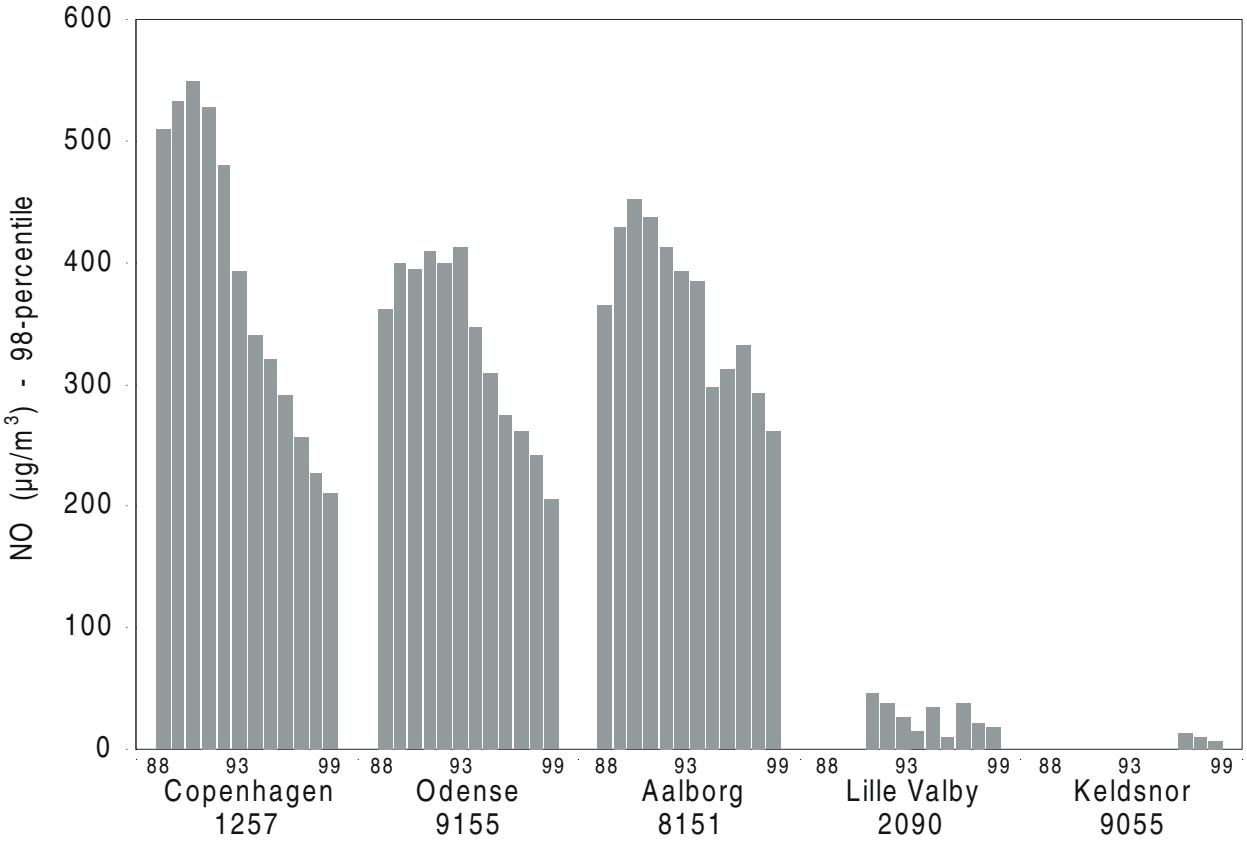
Limit values

The limit value for Denmark is 200 µg/m^3 for the 98-percentile of hourly average values of NO_2 measured over one year. The guide values are 135µg/m^3 for the 98-percentile and 50 µg/m^3 for the median (Miljøministeriet 1987, EEC 1985). At least 75% of the possible measurements have to be available for a valid comparison with the limit and the guide values. The 98-percentiles and the medians of NO and NO_2 since 1988 are found in figure 2.1 and 2.2. The statistical parameters corresponding to the limit and guide values are found in table 2.1 together with the annual averages. The 99.8-percentile represents the 17th or 18th largest value. It may be representative for the peak concentration, with exception of a few extremes.

The limit values were not exceeded in 1999, but the results from Copenhagen exceeded the WHO guide value (40 µg/m^3) and was close to the Danish guide values for the median.

New limit values

The EU Directive 1999/30/EC (EC, 1999) sets new limit values for NO_2 for the protection of human health and vegetation. The limit values for protection of human health are in force 18 months after adoption, but shall be met before January 2010. The annual average for protection of health was slightly exceeded in 1999 at Copenhagen/1257 (table 2.1). It is expected that the ambient concentrations will decrease in the coming years due to already taken measures, e.g. introduction of TWC.



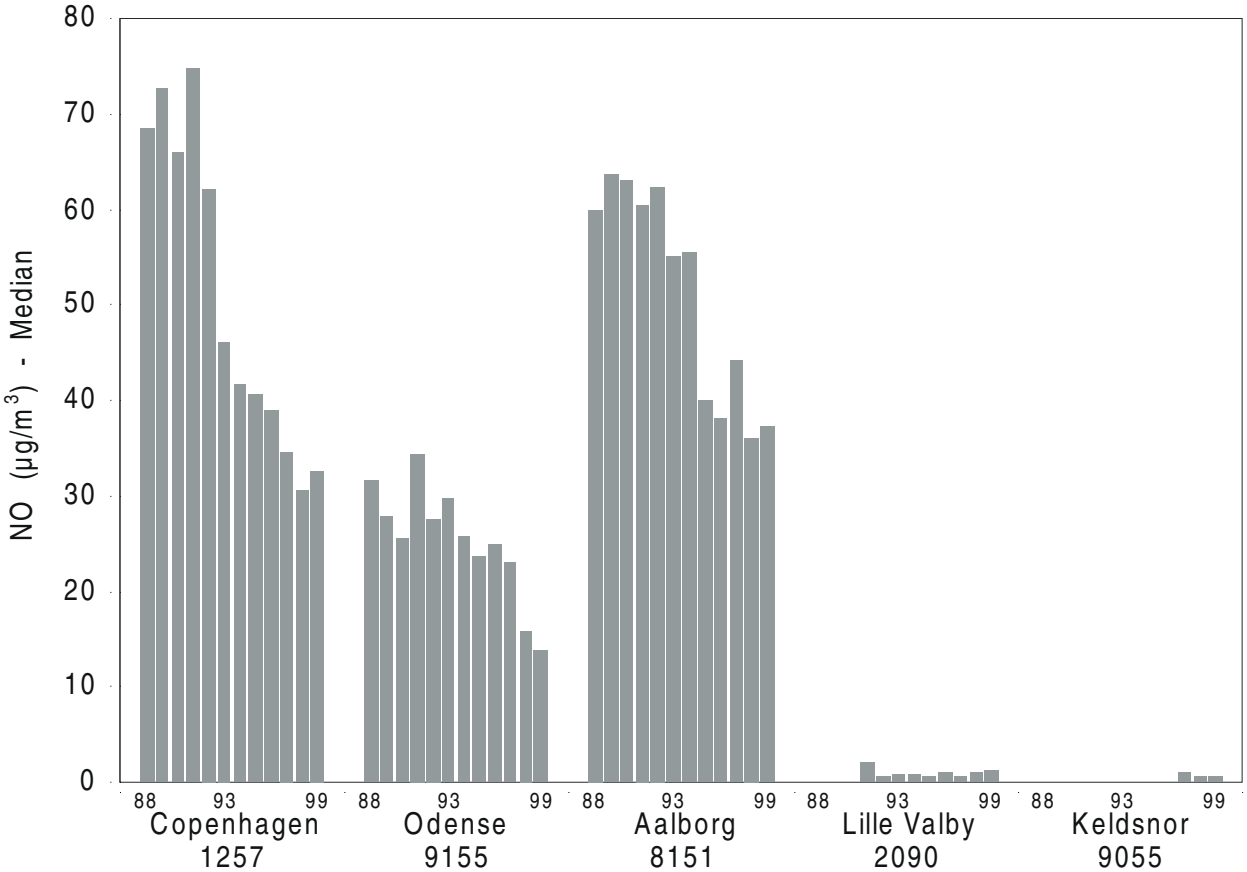


Figure 2.1 Medians and 98-percentiles for NO from 1988 to 1999.

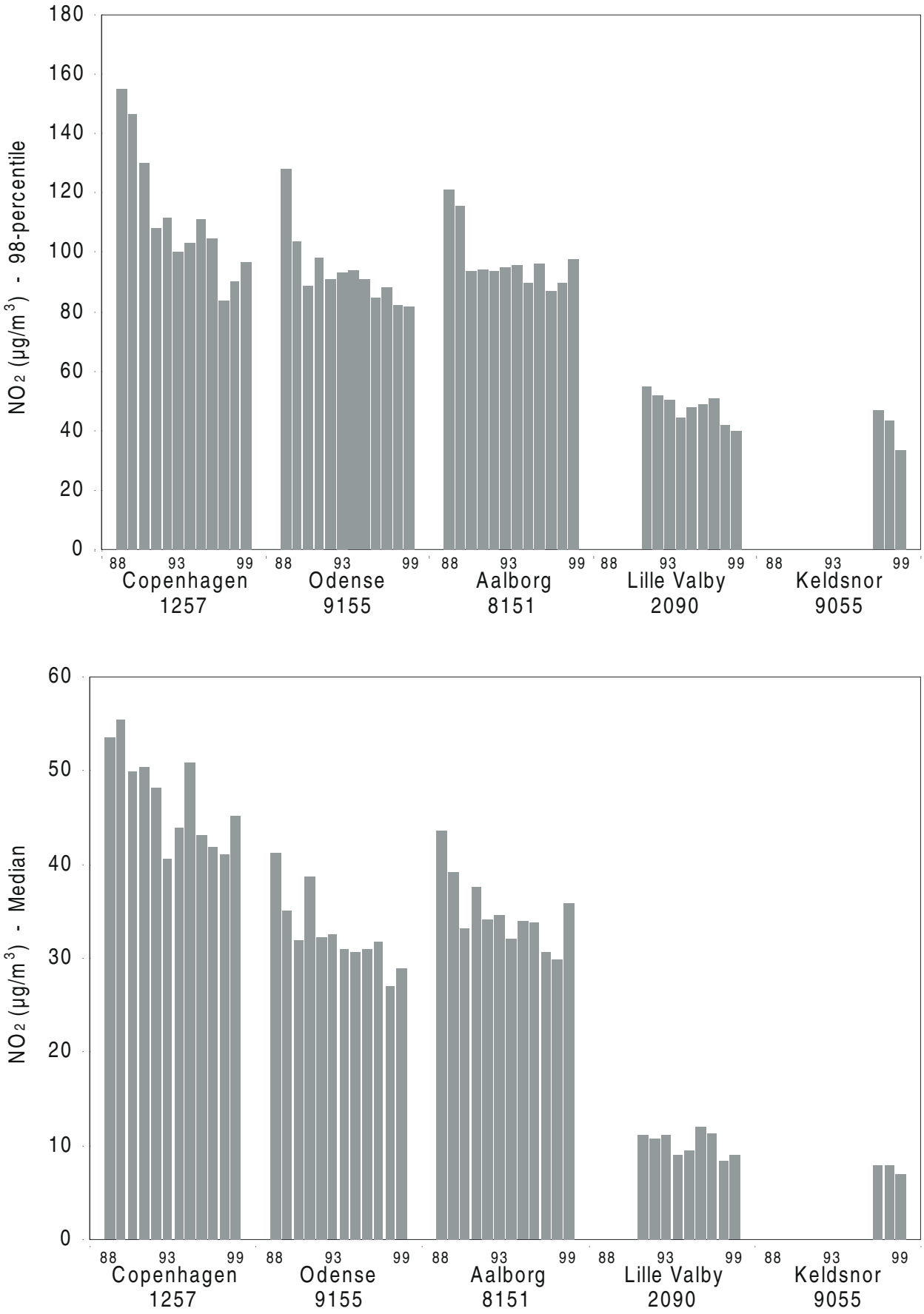


Figure 2.2 Medians and 98-percentiles for NO_2 from 1988 to 1999

NO vs. NO_2

The high concentration of NO compared to NO_2 at traffic stations illustrates that the NO is not a limiting factor for the formation of NO_2 at streets. Under ordinary conditions almost all NO has been oxidised a few hundred meters away at the urban background sites.

Low NO in Odense

It has been observed every year that the average and median concentrations for NO at Odense/9155 is relatively low. It is a result of the location of the station on the north-east side of the street, which is perpendicular to the prevailing south-western wind direction. Due to the street canyon effect the station will not be exposed directly to the pollution from the passing traffic during the prevailing south-westerly winds (Kemp, Palmgren, Manscher 1996).

Table 2.1 The values are calculated for all measurements from 1999 and based on hourly average values. Values below the detection limit are included as one half of the detection limit (more than half of the NO values from Lille Valby/2090 and Keldsnor/9055 are below the detection limit). All stations did yield sufficient results for a valid comparison with the limit and guide values. The number of measurements is listed in the second column. The limit values and Danish guide values are found in Miljøministeriet, (1987), the new values in EC, (1999) and the WHO guide value is given in WHO (2000).

		NO (µg(NO)/m^3)				NO_2 (µg(NO_2)/m^3)			
Station	Num-ber	Ave-rage	Me-dian	98-perc	99.8-perc	Ave-rage	Me-dian	98-perc	99.8-perc
Traffic sites:									
Copenhagen/1257	8290	50	33	211	375	47	45	97	118
Odense/9155	7722	34	14	205	375	33	29	82	104
Aalborg/8151	8699	61	37	262	428	40	36	98	117
Urban background:									
Copenhagen/1259	7849	5	1	40	122	25	22	65	91
Odense/9159	8555	6	2	41	138	20	17	53	75
Aalborg/8159	7456	7	2	71	232	18	14	55	85
Rural:									
Lille Valby/2090	8591	2	(1)	18	51	12	9	40	56
Keldsnor/9055	8107	1	(1)	6	23	9	7	33	51
Limit values	>6570	-	-	-	-	-	-	200	-
New limit values		-	-	-	-	40	-	-	200
Guide value (DK)		-	-	-	-	-	50	135	-
Guide value (WHO)		-	-	-	-	40	-	-	-

2.2 Episodes

Smog warning

NO_2 is included in the national smog warning system. A warning will be issued if the concentration exceeds 350 µg(NO_2)/m^3 for more than three consecutive hours, and if an immediate improvement is not expected.

Table 2.2 shows the highest values measured at the four stations. NO is included for comparison. The values are calculated according to

the provisions in the warning system. Neither the WHO guide line value nor the warning limit were exceeded in 1999.

Table 2.2 Maximum concentrations of NO (not included in the smog warning system) and NO_2 in 1999. For comparison with the warning limit the lowest 1 hour values are identified for every consecutive three hours. (The warning criteria: that the concentration should exceed 350 µg/m^3 for consecutive three hour is the same as that the lowest hourly value within the three hour period exceeds 350 µg/m^3). The highest of these values during the whole year are listed under "max. 3 hour". The values under "max. hour" are the absolute one hour maximum values. The indication of time is the beginning of the periods.

NO	Max. 3 hour (µg(NO)/m^3)	Day hour	Max. hour (µg(NO)/m^3)	Day hour
Traffic sites:				
Copenhagen/1257	399	990331: 8	671	990817: 6
Odense/9155	457	990211: 8	641	990211: 8
Aalborg/8151	576	991108:15	617	991108:17
Urban background:				
Copenhagen/1259	131	991221: 8	243	990331: 8
Odense/9159	191	990211: 8	254	991111: 8
Aalborg/8159	234	991220:20	425	991220:21
Rural:				
Lille Valby/2090	43	990327: 5	121	991111: 9
Keldsnor/9055	29	991123:23	38	991123:23
Warning limit				
	Max. 3 hour (µg(NO)/m^3)	Day hour	Max. hour (µg(NO)/m^3)	Day hour
Traffic sites:				
Copenhagen/1257	125	990528: 6	147	990528: 7
Odense/9155	107	990211: 8	122	990211: 9
Aalborg/8151	118	990805: 6	136	990403: 8
Urban background:				
Copenhagen/1259	113	990402:21	116	990402:22
Odense/9159	82	990211: 9	95	990211:10
Aalborg/8159	80	990222:18	110	990222:19
Rural:				
Lille Valby/2090	62	990331:19	65	990402:21
Keldsnor/9055	52	991222: 3	75	990729:20
Warning limit	350			
Guide value (WHO 2000)			200	

The highest concentrations of NO are usually seen in the winter month at all stations, however local road or building construction

may influence the maximum values. The peak concentrations of NO_2 are most frequent in spring or summer due to the higher background values of O_3.

No major episodes in 1999

Figure 2.3 show a typical spring episode. Around April 1st a period with clear sky and wind, mainly from south-east at low speed, gave rise to transport of O_3 from Central Europe during daytime. In the nights the wind almost disappears and a low mixing layer allows the local NO_x emissions to gradually spread out. The highest NO_2 concentrations are observed a couple of hours earlier in Copenhagen than at the semi-rural station Lille Valby/2090. All O_3 is removed during night due to deposition and formation of NO_2.

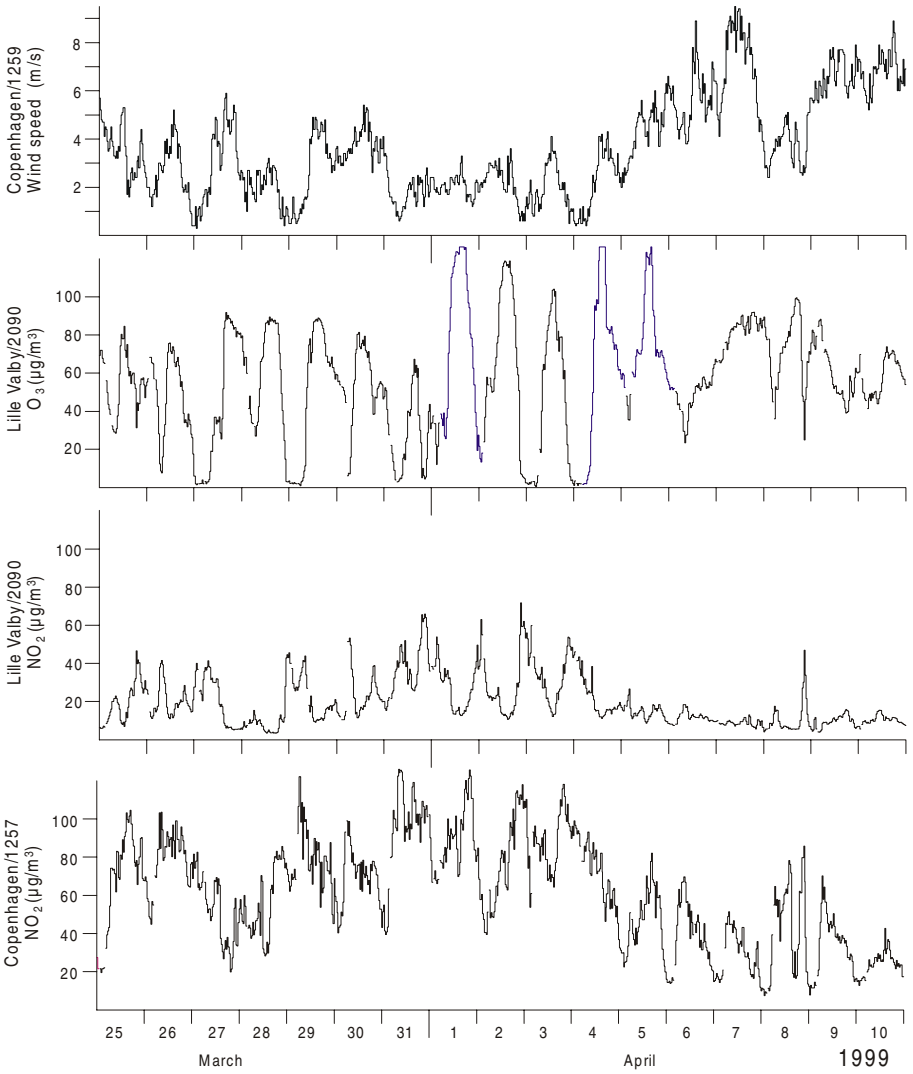


Figure 2.3 The first spring episode in 1999.

2.3 Trends

Percentiles

The annual percentiles and average values for NO and NO_2 measured at Aalborg/8151 are shown in figure 2.4. The level of NO was almost constant in the period from 1982 to 1991, but decreased during the following years synchronously with the increasing number of cars with TWC.

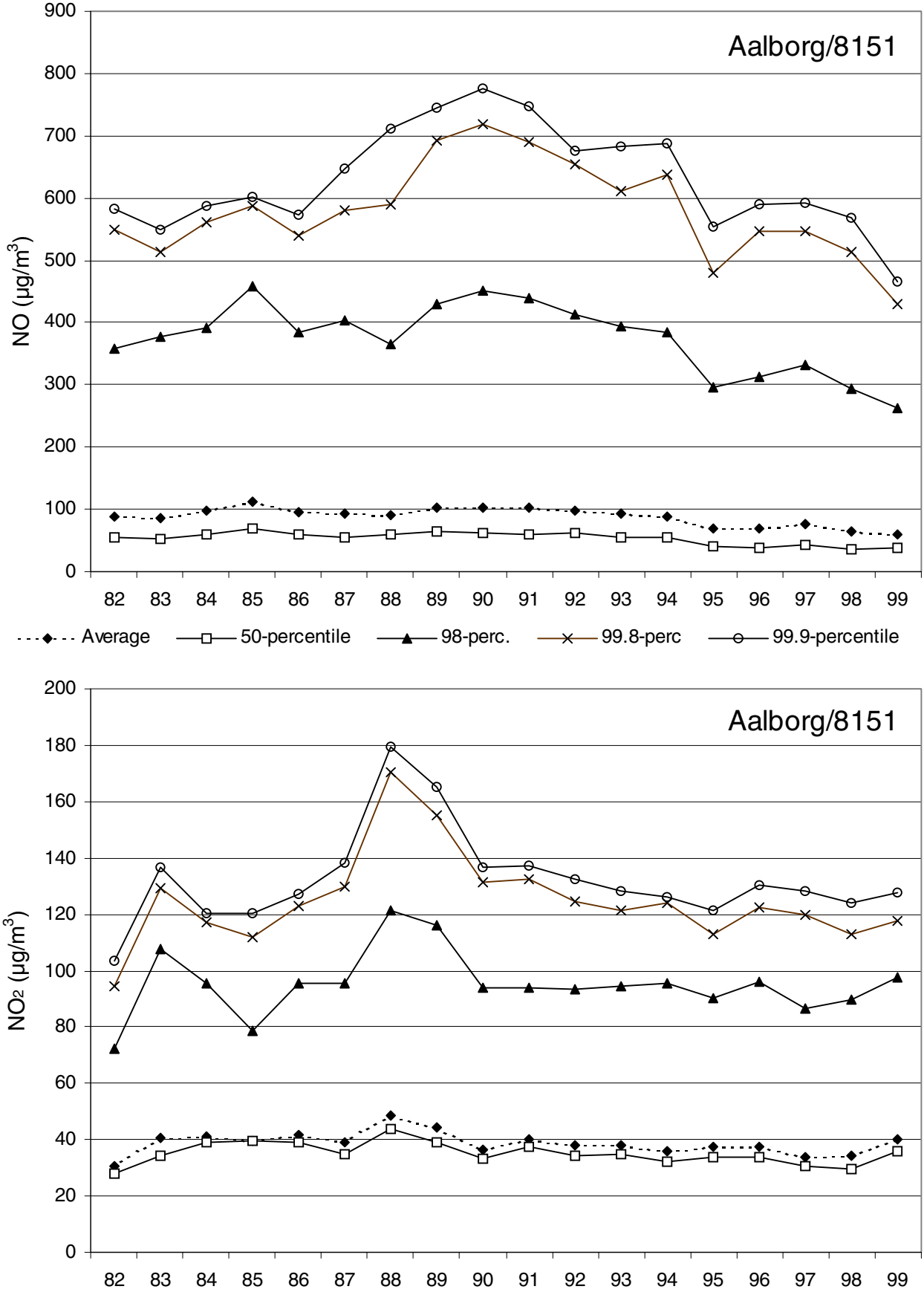


Figure 2.4 Trends for annual 99.9-, 99.8-, 98-, 50-percentiles, and average value based on hourly average concentrations of NO_2 and NO measured at Aalborg/8151.

The slight decrease of the average and median concentrations of NO_2 the previous 5-6 years is broken in 1999. But the change in 1999, which is caused by the increased O_3 concentrations (see chapter 3.3), is probably not an indication of a change in the generally decreasing trend. The decrease is however much less than for NO and hardly statistically significant. It is still almost obscured by the meteorological variations from year to year.

The skewness of the distributions is obviously much greater for NO than for NO_2. The ratios between the 98-percentile and the median are approximately 7 for NO, whereas they are only 2-3 for NO_2. This indicates that at least at high NO concentrations the NO_2 formation is limited – probably by the O_3 concentrations.

Averages

The trend of the monthly average values and the annual variation are shown in figure 2.5. The variation for NO and NO_2 seems to be (to some extent) opposite in the sense that years with high NO concentrations correspond to years with low NO_2 concentrations and vice versa. There is a distinct annual variation for NO with low concentrations during the summer, when the emissions are lower and the oxidation rate is higher. The variation is much less pronounced for NO_2.

Copenhagen vs. other cities

Figure 2.1 indicate that the NO concentrations have been decreasing more in Copenhagen than in the two smaller cities Odense and Aalborg. The reason for this is not obvious, but it may be caused by a reduction of the urban background levels, which are higher in Copenhagen than in the two other cities.

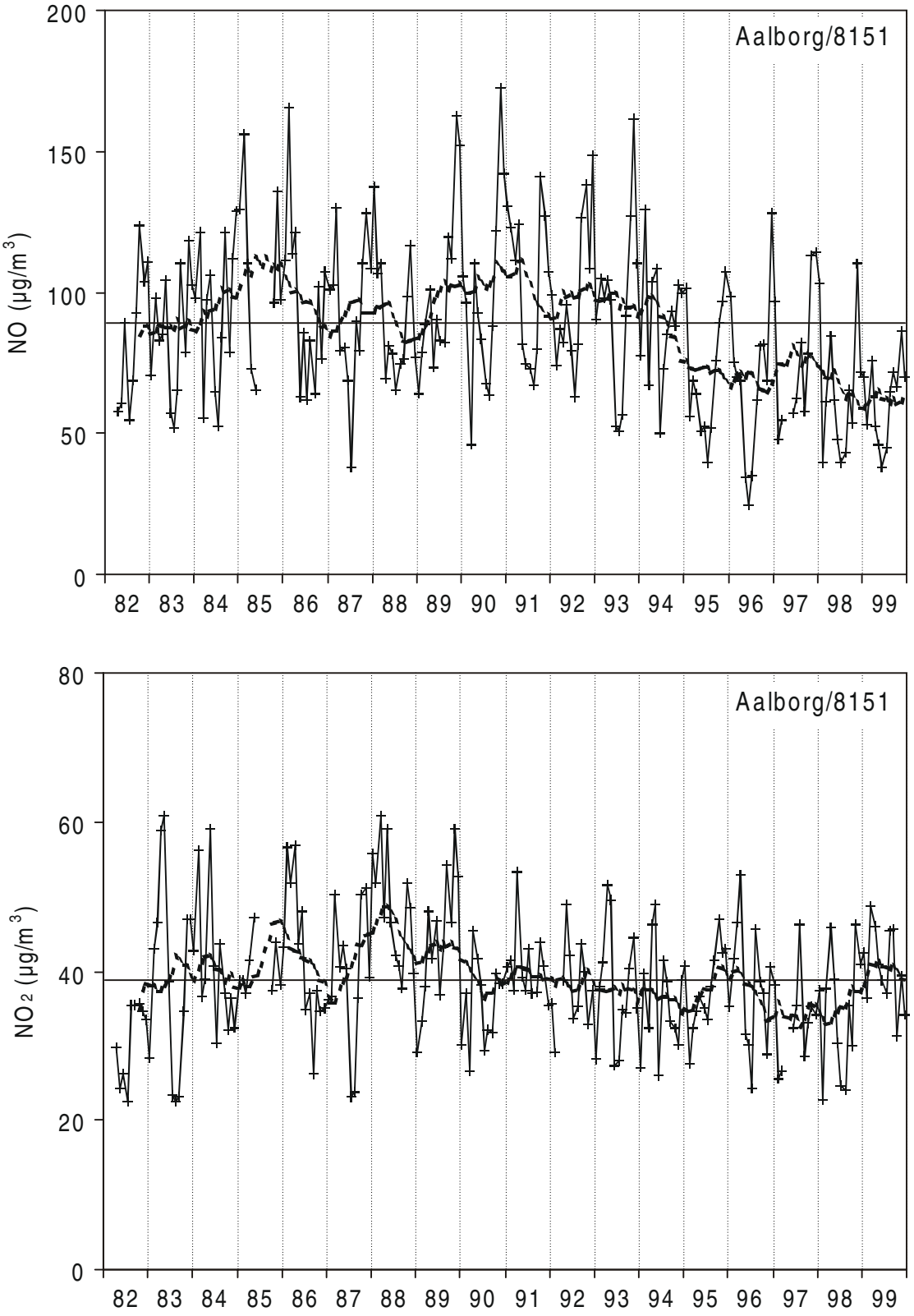


Figure 2.5 Trend for NO and NO_2 measured at Aalborg/8151. The points are measured monthly averages, the dotted curve is a moving average over 13 month and the straight line is the average over all years.

3 Ozone

Measurements in 1999

Measurement of O_3 was started mid 1991 at the rural station (Lille Valby/2090). At the end of 1992 the urban background stations (Kemp 1993) were equipped with monitors. The measurements started at Odense/9159 in August 1992 and at Aalborg/8159 in December 1992. At Copenhagen/1259 continuous measurements started in February 1993 with a DOAS instrument and a monitor was installed in April 1993. The O_3 measurements at Keldsnor/9055 began in January 1995 (see table 1.1). Further O_3 measurements were performed from April 1994 at the street station Copenhagen/1257 in connection with the TOV programme (Berkowicz et al. 1996). The monitor measurements at Copenhagen/1259 were stopped in July 1999; apart from that almost complete series of results are available from all stations.

Sources and formation

The O_3 in the lower troposphere is formed as a secondary pollutant mainly by photochemical reactions involving i.a. volatile organic compounds (VOC) and NO_2. An important parameter for the reaction velocity is the ambient temperature. The VOC may either be of anthropogenic or natural origin. Oxidation of NO is the main reaction for the reduction of O_3 in urban areas. The climatic conditions and the emission patterns in Denmark result in a net decomposition in urban areas due to the high NO emissions from combustion processes, whereas the presence of O_3 in background areas, especially during the summer, exceeds the levels of reducing compounds.

3.1 Annual statistics

Threshold values

The EEC directive on air pollution by O_3 (EEC 1992) is implemented in Denmark through a governmental regulation (Miljøministeriet, 1994). It obligates the member states i.a. to perform measurements of O_3 at localities where the threshold values given in the directive are likely to be exceeded, and where it is possible that human individuals or vegetation are exposed to O_3 pollution.

Concentrations depending on site type

The levels at the rural and the urban background stations are almost the same. The concentrations in street level are considerably lower, due to reaction with i.a. NO. This confirms that the NO_x concentrations in general are lower than the O_3 concentrations away from the busy streets, i.e. there is not sufficient NO to remove a significant part of the O_3 close to trafficked streets.

Exceedings

The measured values are compared to the threshold values in table 3.1. In May the meteorological conditions were favourable for O_3 formation and transport. The temperature was relatively high and the air was in periods coming from Central Europe. Otherwise there were only few occasions with high concentrations. The values in 1999 were generally higher than the corresponding 1998 values. The max. 24 hour and the max. 8 hour threshold values were exceeded on many occasions in 1999 at all stations. There were exceedances in all

month of the year. It is always the 24 hour threshold that is exceeded in the winter. The threshold at 65 µg/m^3 is very close to the background of the Northern Hemisphere. As shown previously (Kemp, Palmgren and Manscher, 1996) the O_3 concentrations always reach this level at wind speeds above 10 m/s, especially from Northwest.

Table 3.1 Annual average values, percentiles and maximum values for O_3 measured in 1999 compared to threshold values. (Miljøministeriet 1994, EEC 1992). The eight-hour values are calculated in accordance with the EEC directive, as a non-overlapping moving average; they are calculated four times a day from the eight hourly values between 0 and 9, 8 and 17, 16 and 1, 12 and 21.

O_3 (µg/m^3)	Average	Median (hour)	98-perc. (hour)	99.9-perc. (hour)	max. 24 hours	max. 8 hours	max. 1 hour
Urban Background:							
Odense/9159	51	52	104	152	107	146	180
Aalborg/8159	51	53	99	134	98	125	142
Rural:							
Lille Valby/2090	59	60	115	167	118	174	188
Keldsnor/9055	66	66	115	145	113	130	154
Traffic:							
Copenhagen/1257	33	31	77	99	77	92	119
Threshold value	-	-	-	-	65	110	200
Average number of exceedances per station (excl. Copenhagen/1257)	-	-	-	-	100	10	0

AOT40

UN-ECE uses the concept of critical levels to assess the effects of O_3 to agricultural crops and ecosystems (UN-ECE 1996). The effect parameter is calculated as the accumulated O_3 exposure above a threshold value of 40 ppb (≈ 80 µg/m^3), the so-called AOT40. The excess is expressed as the number of ppb·h above 40 ppb. The guideline for crops is based on integration over all daylight hours during May-July, while the guideline for forest is based on the whole period April-September. Since the AOT40 values represent "difference concentrations" only a slight change of the absolute level may affect the AOT40 value drastically. It is not unusual that the AOT40 value is changed by a factor of two from year to year as seen for the AOT40(forest) from 1998 to 1999. The values from the same year are however remarkably close for stations over the Northern Europe as illustrated by the results from UK and Sweden (table 3.2) from the same two years.

The measured values in 1999 exceeded the UN-ECE guide values (table 3.2). Apart from the episode in August, which added an extra amount to the forest related AOT40, a steady increased was observed during the whole seasons in 1999 (fig. 3.1).

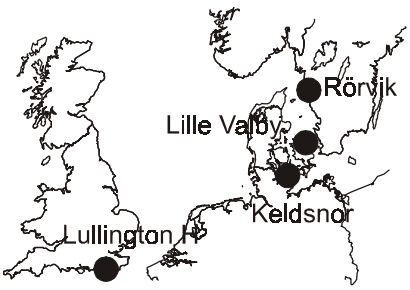


Table 3.2 AOT40 values (UN-ECE 1996). Unit ppb·h. The AOT40(forest) from the UK station at Lullington Heath is calculated from the information found at www/aeat.co.uk/netcen/airqual/ and the AOT40(forest) from the Swedish station at Rörvik is from www.ivl.se/miljo/projekt/ozon/.

Station	1995	1996	1997	1998	1999
AOT40 (crops)					
Lille Valby/2090	4700	1513	1409	3008	3400
Keldsnor/9055	2850	2540	-	4263	5900
Guide v. UN-ECE				3000	3000
AOT40 (forest)					
Lille Valby/2090	8850	5510	5152	4940	13100
Keldsnor/9055	6800	8020	-	7040	14000
Lullington Heath				7700	13900
Rörvik				6400	12500
Guide v. UN-ECE					10000

3.2 Episodes

Threshold values

The EEC directive makes it mandatory to inform the population, if the hourly average concentration of the O_3 exceeds 180 µg/m^3 and to issue a warning, if the hourly average concentration exceeds 360 µg/m^3. The information or warning shall include the following information:

- Date, hour and place of the occurrence of concentrations in excess of the above mentioned threshold values.
- Reference to the type(s) of community values exceeded (information or warning).
- Forecasts of the change of concentrations, geographical area concerned and the duration.
- Population concerned.
- Precautions to be taken by the population concerned.

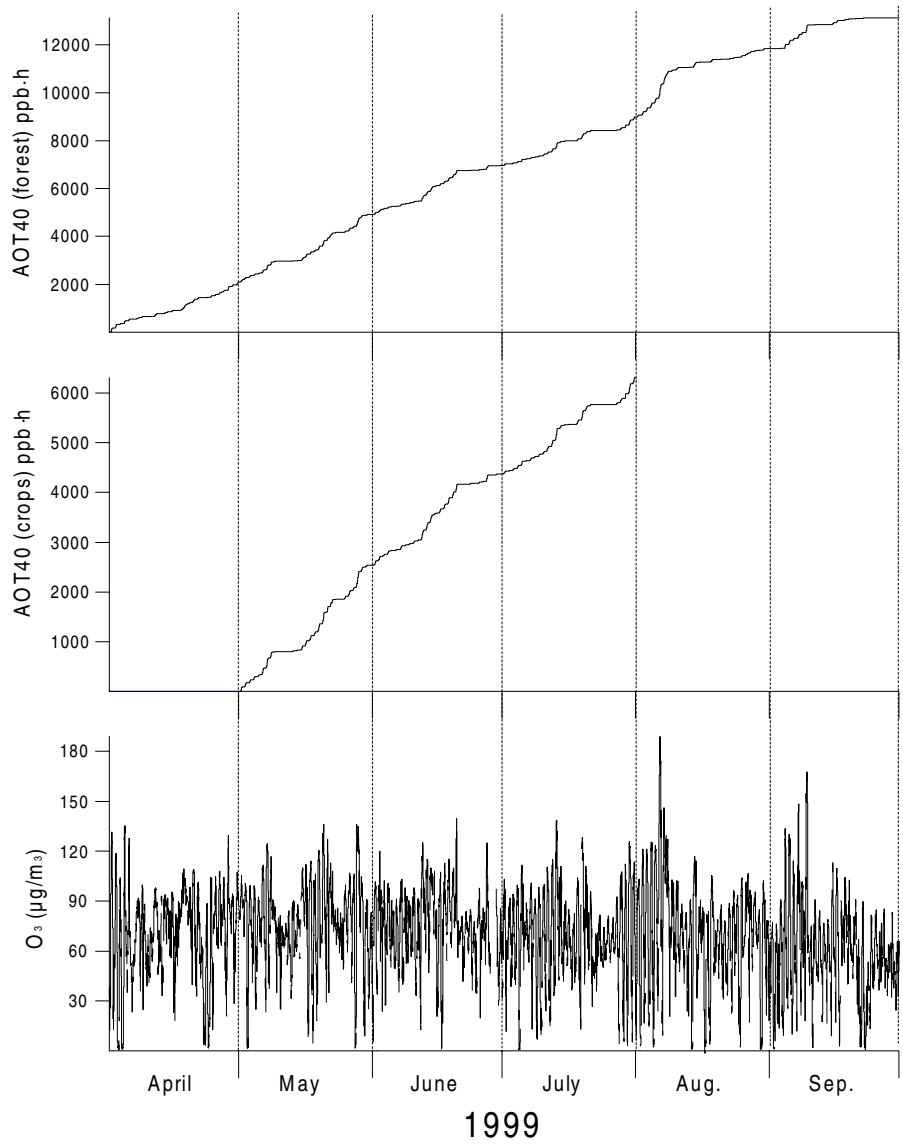


Figure 3.1 O_3 results from Lille Valby/2090. The lower curve is the measured ½-hour averages, while the two upper curves are the cumulated AOT40 values representing the values for crops and forest.

Press releases

No press releases due to exceedance of the information threshold at 180 µg/m^3 were send out. The highest hourly concentration in Denmark of O_3 was in 1999 measured to 188 µg/m^3 at Lille Valby/2090 August 6. The information threshold was exceeded for two consecutive hours that day. It was however not revealed before the final quality control took place. Apart form this single incidence no concentrations above 180 µg/m^3 were observed.

3.3 Trend

Trend for O_3 and O_x

More than seven years of data are now available from Lille Valby/2090. The development is illustrated by the annual averages and 98-percentiles in figure 3.3. The results are divided according to wind direction sectors. The sector from 40 to 160^0 represent the continental contribution with an addition of some regional contribution from the Copenhagen area, the sector from 180 to 240^0 represents the western part of Continental Europe, while the background contribution is assumed to be found in the sector from 260 to 20^0.

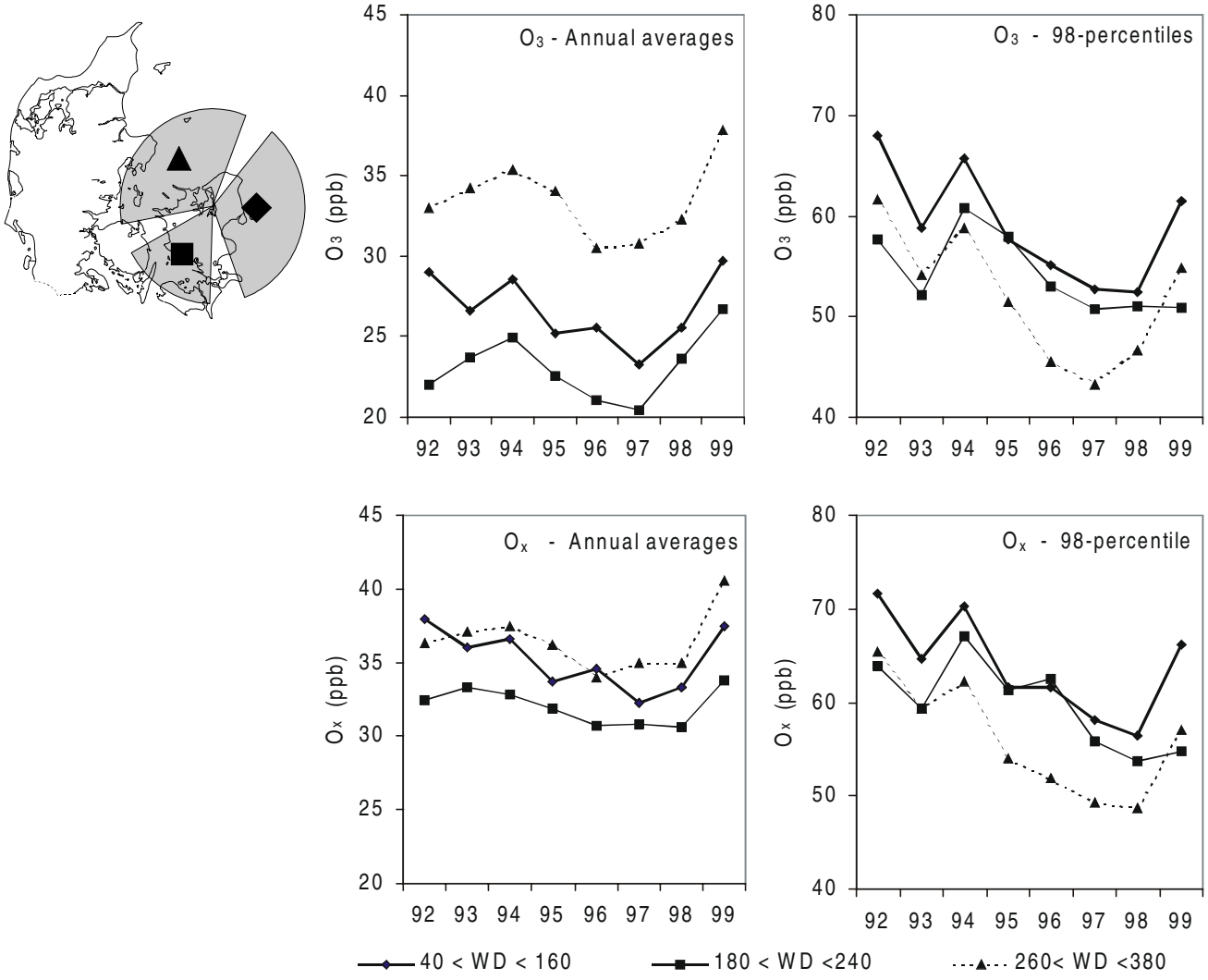


Figure 3.3 Annual averages and 98-percentiles measured at Lille Valby/2090 for O_3 and O_x (O_3+NO_2). The wind sectors for the single curves are shown on the map. The signatures on the map correspond to the points on the curves.

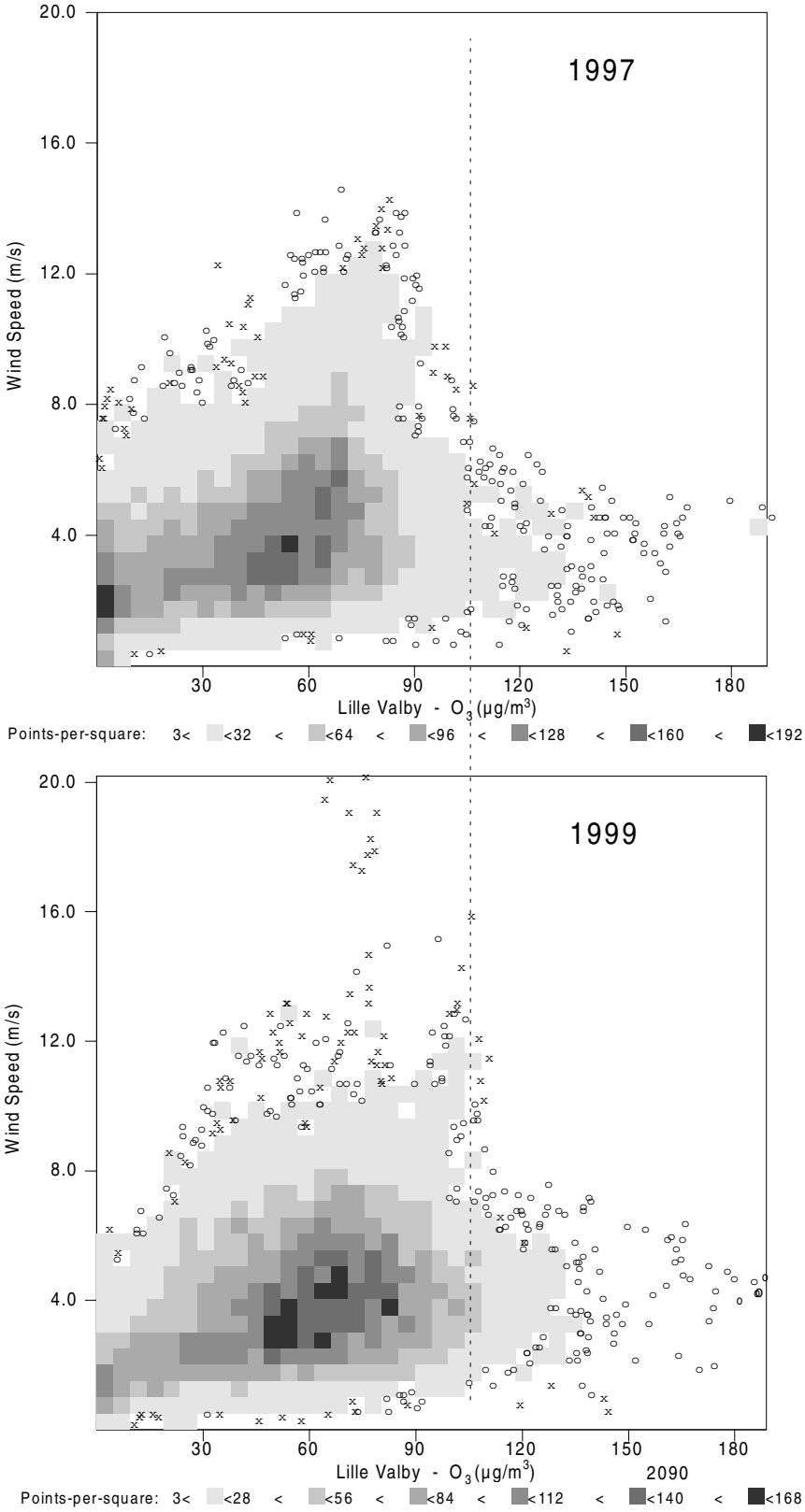


Figure 3.4 Relationship between O_3 concentrations and Wind speed in 1997 and 1999, which illustrates the change of the "hemispheric background" as indicated for 1999 by the dotted line. The plot area is divided into 40x40 squares. The squares are shaded according to the scales below the graphs if more than three points is found within a square. Single points are shown as x for winter and o for summer measurements.

The highest average concentrations of O_3 were observed in the background sector, while the O_x concentrations were remarkably

alike for all three sectors. This is in agreement with the results reported in the previous years (Kemp and Palmgren, 1999). Further it can be noted that the difference between O_3 and O_x is very small for the 98-percentiles.

The decreasing trend from 1992 to 1997, was broken with the 1999 results. It may be caused by different meteorological conditions in 1999 compared to the previous years. Windy periods in January and December and warm weather in August 1999 gave relatively high concentrations. Previously it has been suggested that the highest concentrations measured at high wind speeds (> 8 m/s) may be interpreted as the "hemispheric background" level (Kemp, Palmgren and Manscher, 1996). Fig. 3.4 shows that the hemispheric background has changed from around 90 µg/m^3 to around 110 mg/m^3 in 1999.

3.4 Smog Warners Data Exchange

A provisional system has been implemented for "on-line" exchange of O_3 measurement results between several north European countries. Every day before 10 am GMT the hourly maximum value for O_3 from the previous day and the latest available result from a number of sites in Austria, Belgium, The Czech Republic, Denmark, Finland, Germany, Holland, Luxembourg and UK are send by e-mail to AEA in UK. Since last year the network is extended with four stations in The Czech Republic and two more stations in UK (cf. fig. 3.5).

At 10 am GMT all collected results are returned to the participating organisations. The data exchange was established by the Technical Working Group on Data Exchange and Forecasting for Ozone episodes in Northwest Europe, which was initiated at the Ministerial Conference on Tropospheric Ozone in Northwest Europe held in London in May 1996. The purpose with the data exchange is primarily to supply data, which can be used for validation of models for forecasting of O_3 episodes. These episodes will usually cover a large part of the North Europe and will pass the area within a few days.

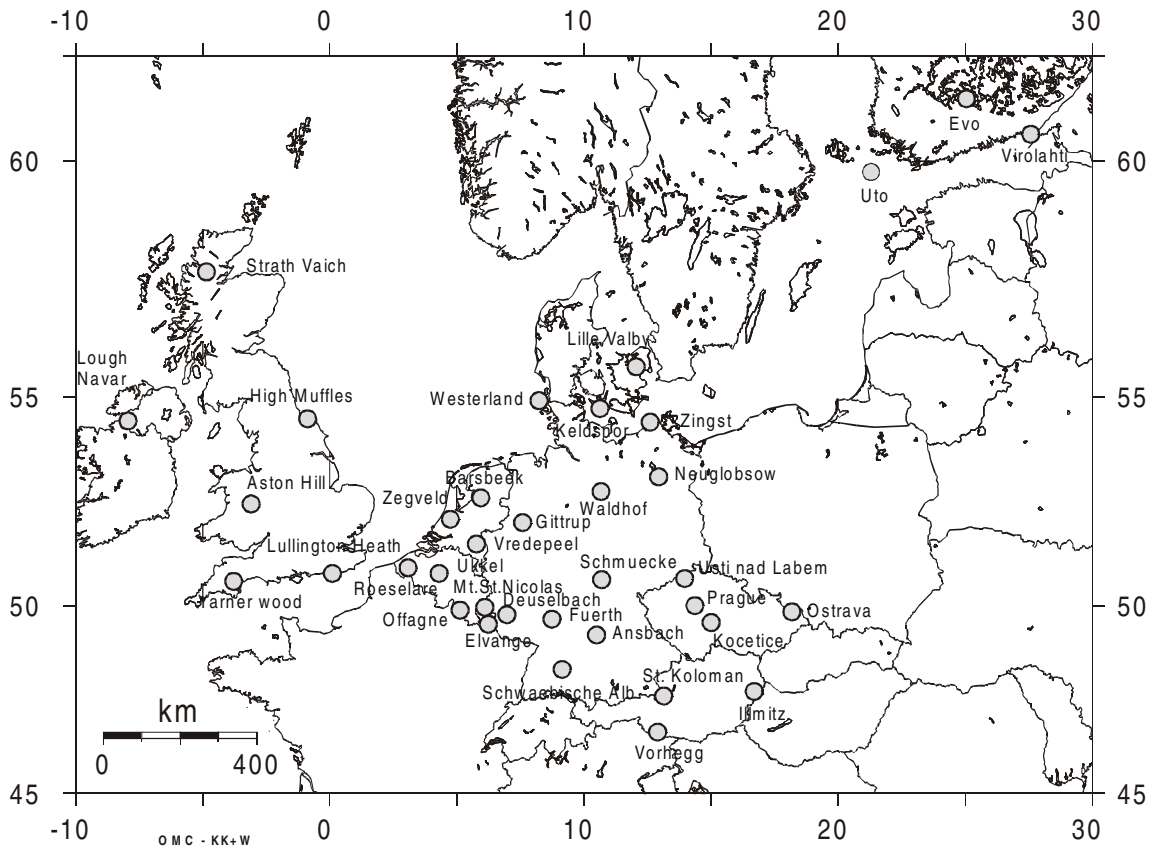


Figure 3.5 Sites participating in Smog Warners Data Exchange.

4 Sulphur compounds

Sources

Sulphur is determined in gas phase as SO_2 and as the elemental content in particulate matter. The main source of SO_2 is combustion of heavy oil and coal. Sulphur in particulate matter is expected to be sulphate (either HSO_4^- or SO_4^{2-}). The two main sources of the sulphate are the oxidation of SO_2 to H_2SO_4 and sulphate directly emitted from the sea. Sea spray will only contribute significantly to the sulphate at the stations in the Danish Air Quality Monitoring Programme during strong wind from west and north-west. The oxidation time for SO_2 in the atmosphere is of the order of one day meaning that the collected particulate sulphur to a large extent has been emitted from sources several hundred km from the stations, while the SO_2 may be of local origin as well as long range transported.

Measurements

In 1999 the concentrations of both SO_2 and particulate S were determined as 24-hour average values at the three main stations (Copenhagen/1257, Odense/9155 and Aalborg/8151), the additional station (Odense/9154) and the background station (Lille Valby/2090). During the year the ½-hour SO_2 measurements were stopped at all stations but Aalborg/8151.

4.1 Annual statistics

Limit values

The measured SO_2 concentrations in Denmark are listed in table 4.1 together with the limit values (Miljøministeriet 1986). The set of limit values is a simplified, but more stringent, version of those laid down by EEC (EEC 1980, 1989). The medians and 98 percentiles are shown in figure 4.1. At all stations the measured values were well below the limit and guide values. The winter concentrations were somewhat higher than the values for the whole year.

Table 4.1 SO_2 and particulate sulphur pollution in Danish cities. The values are calculated for all valid 24 hour results from 1999. The winter is defined as the three first and three last months of the year. The number of measurements for SO_2 is given for the whole year/as well as for the winter.

Station	SO_2 ($\mu g(SO_2)/m^3$)							Particulate S ($\mu g(S)/m^3$)		
	Number	Median year	Median winter	Max. 3 days	98-perc.	Max. day	Average	Number	Average	Max. day
Copenhagen/1257	356/177	3.4	3.8	13	12	24	4.0	356	1.22	5.6
Odense/9155	360/180	1.3	1.4	5.9	5.9	10.9	1.7	361	1.21	4.4
Odense/9154	362/180	1.3	1.4	5.0	6.6	11.7	1.7	362	1.21	4.3
Aalborg/8151	358/179	1.4	1.3	5.2	6.3	11.6	1.8	358	1.18	4.6
Lille Valby/2090	363/180	1.1	1.2	5.2	6.6	17	1.5	363	0.99	4.1
Limit value	-	80	130	250	250	-	-	-	-	-
Guide - (EEC 1980)	-	-	-	-		-	40-60	-	-	-
Guide - (WHO 2000)	-	-	-	-	-	125	50	-	-	-

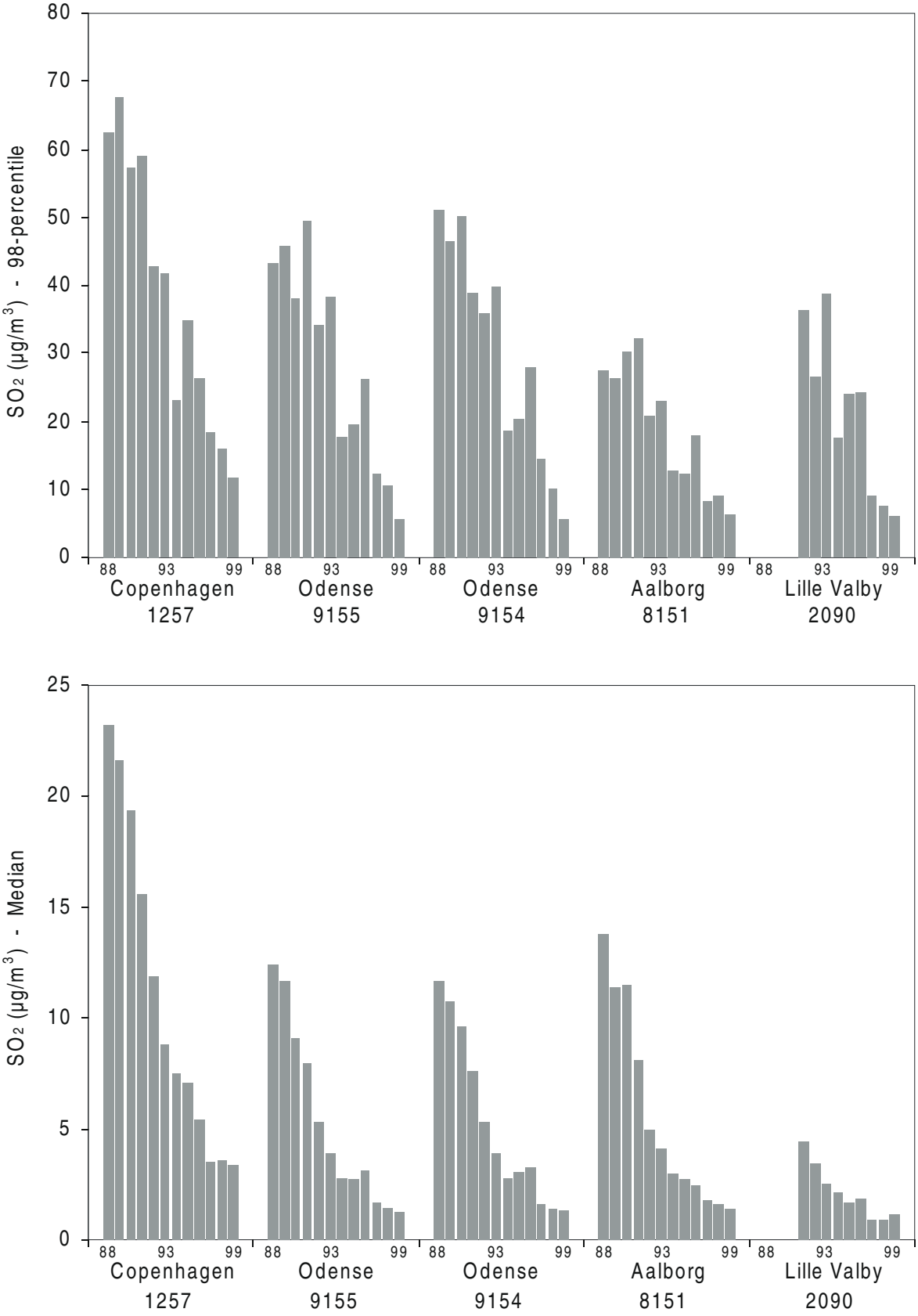


Figure 4.1 Medians and 98-percentiles for SO_2 from 1988 to 1999.

The EU Council has adopted a new set of limit values for i.a SO_2 (EC, 1999) The new values are found in table 4.2 together with the measured results. The new limit values are to be met either in July 2001 or January 1, 2005. All measured values are far below the new values.

Table 4.2 Limit values in EU Directive 1999/30/EC (EC, 1999) and the corresponding measured results from 1999. The values in column 2 and 3 are for protection of human health, while the yearly and winter averages are for protection of ecosystems and shall only be met at rural sites.

SO_2 µg/m^3	25th highest hour	Fourth highest day	Annual average	Winter average
Copenhagen/1257	-	12.7	-	-
Odense/9155	-	6.9	-	-
Odense/9154	-	7.0	-	-
Aalborg/8151	26	8.0	-	-
Lille Valby/2090	-	7.4	1.5	1.6
New limit value	350	125	20	20
To be met	2005	2005	19 July 2001	19 July 2001

New limit values

Drastic decrease in sulphur in 1997

The measured values were considerably lower in 1997 than in 1996. The difference is mainly due to a much smaller contribution from Eastern Europe (Kemp, Palmgren and Manscher, 1998). The effect was observed for both SO_2 and particulate S. The patterns in 1998 and 1999 were much like that in 1997 and slightly decreasing. This indicates that the reduction in the air concentration in fact correspond to a decrease in the emission in the east European countries.

Smog warning

A smog warning system for NO_2, SO_2 and O_3 was implemented in Denmark in April 1994. A warning will be issued for SO_2, if the concentration exceeds 350 µg/m^3 for more than three consecutive hours and an immediate improvement is not expected. The new Directive (EC, 1999) includes an alert threshold at 500 µg/m^3 measured over three consecutive hours. Neither of these values are expected to be exceeded at any time in Denmark. The highest observed value to be compared with the above limits was 29 µg/m^3 in 1999.

Episodes

The emission of SO_2 is, both in Denmark and the neighbouring countries, reduced considerably during the last two decades. Some years ago concentrations during episodes reached frequently levels close to limit and guide values. Today concentrations, even during

the episodes, are well below critical values. As a consequence of this positive development it has been decided to reduce the ½-hour measurements of SO_2 in the LMP program. The station Aalborg/8151 is now the only place where a SO_2 monitor is in operation. Two so-called hot-spot episodes were observed during 1999. The highest hourly maximum at these two occasions were 73 and 62 µg/m^3.

4.2 Trends

Percentiles for SO_2

The annual percentiles and average values based on daily average SO_2 concentrations measured at Aalborg/8151 are shown in figure 4.2. The level of SO_2 has been decreasing since 1982. The reduction is most evident for the "long term" values (median and average values), which are determined by the contributions from a number of local sources, while the long range transport episodes contribute very much to the 98-percentile and the other "short-term" values. After a couple of years with stagnation or even increase of the short term parameters the values for 1997 and on were drastically reduced.

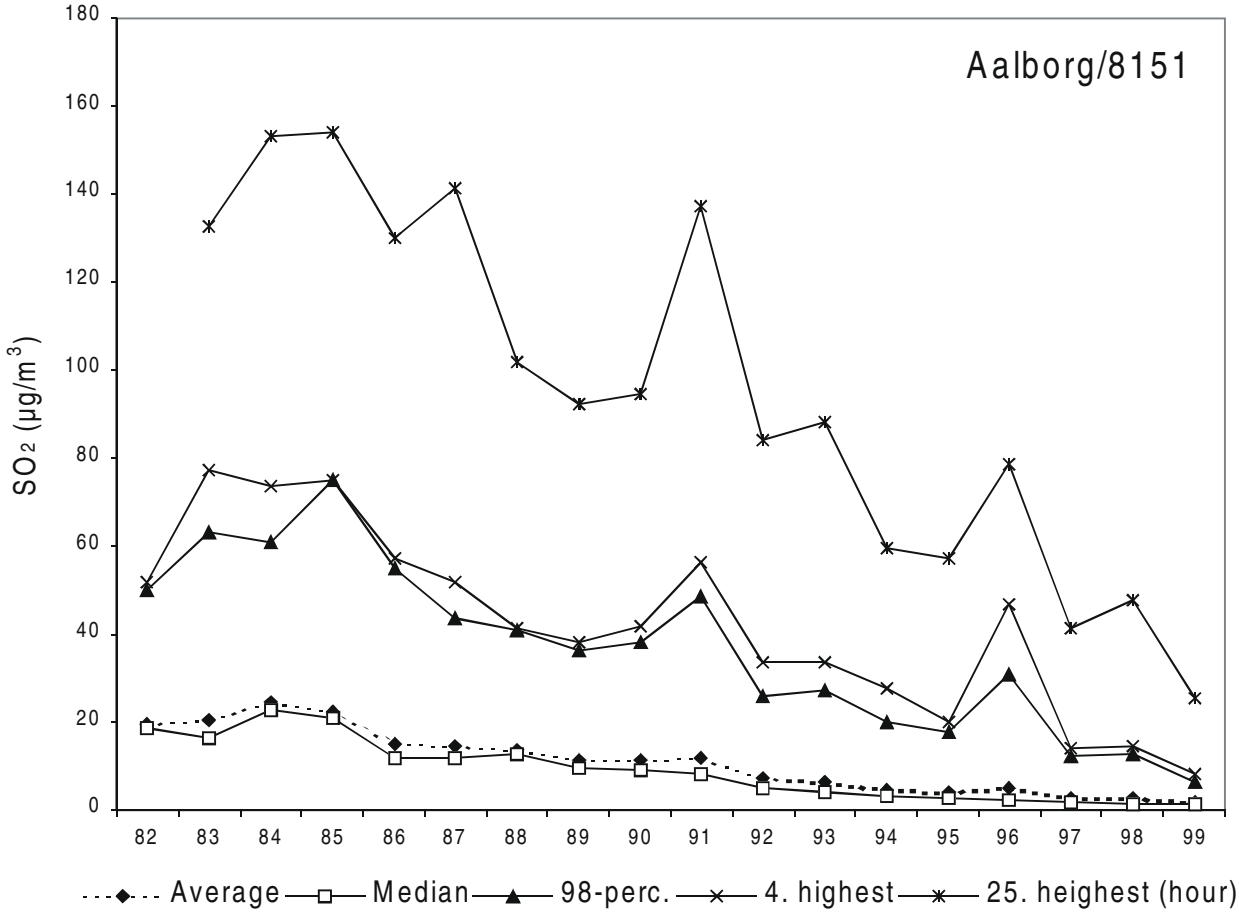


Figure 4.2 Trends for annual average value, median, 98-percentile and forth highest value based on daily results together with the 25th highest hourly average. All results are from Aalborg/8151.

Avarage SO_2 and S concentrations

The trends for the monthly average values at Aalborg/8151 and Odense/9154 are shown in figure 4.3 and 4.4. The average SO_2 concentrations have been reduced with almost a factor of five, since it

peaked around 1984. The steep decrease in 1985-86 was caused by a compulsory reduction of the sulphur content in fossil fuel from January 1986. Better combustion control and increased use of natural gas for domestic and district heating and introduction of lighter diesel oil are the main reasons.

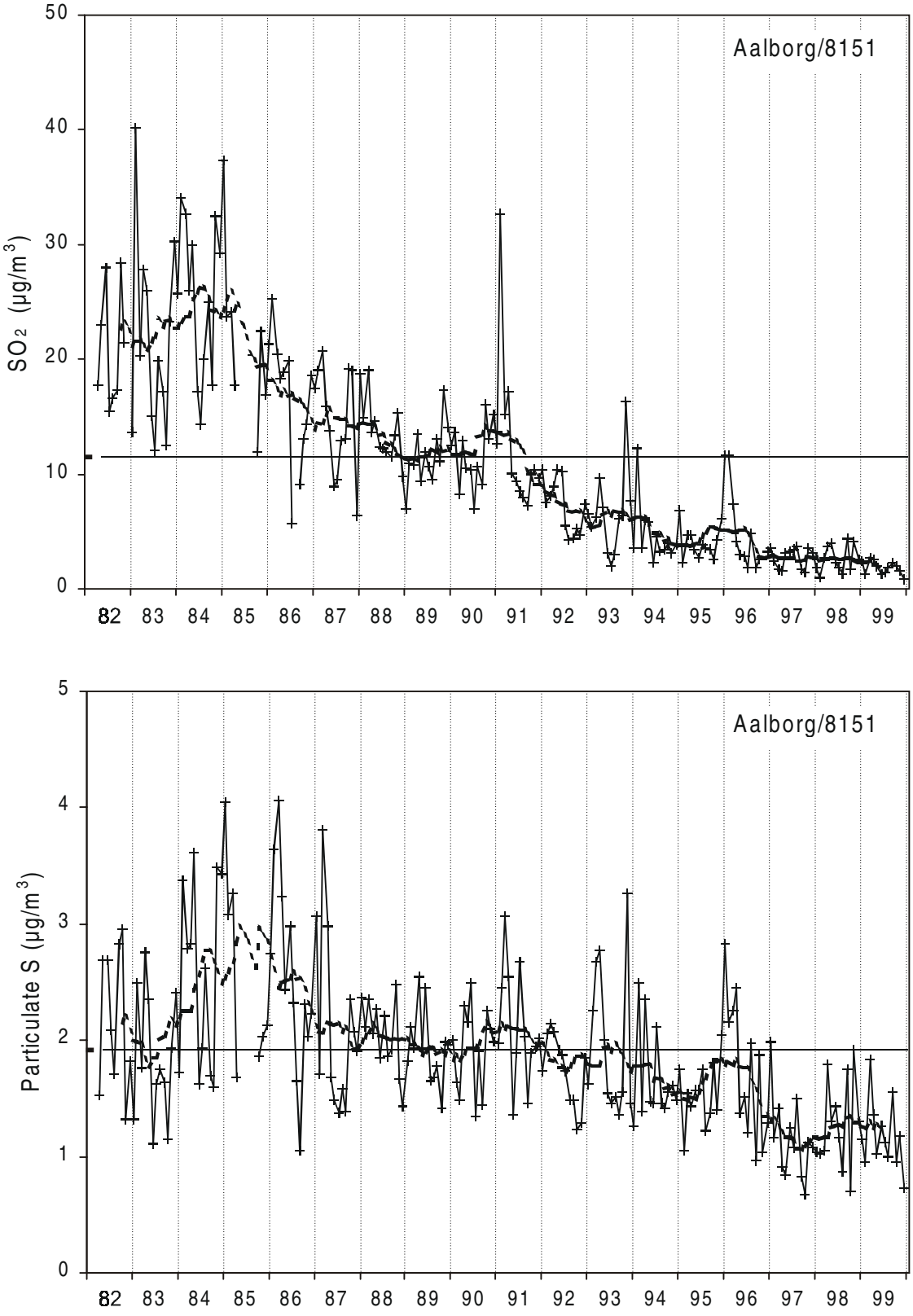


Figure 4.3 Trend for SO_2 and particulate S measured at Aalborg/8151. The points are measured monthly averages, the dotted curve is a moving average over 13 month and the straight line represents the average over the whole period.

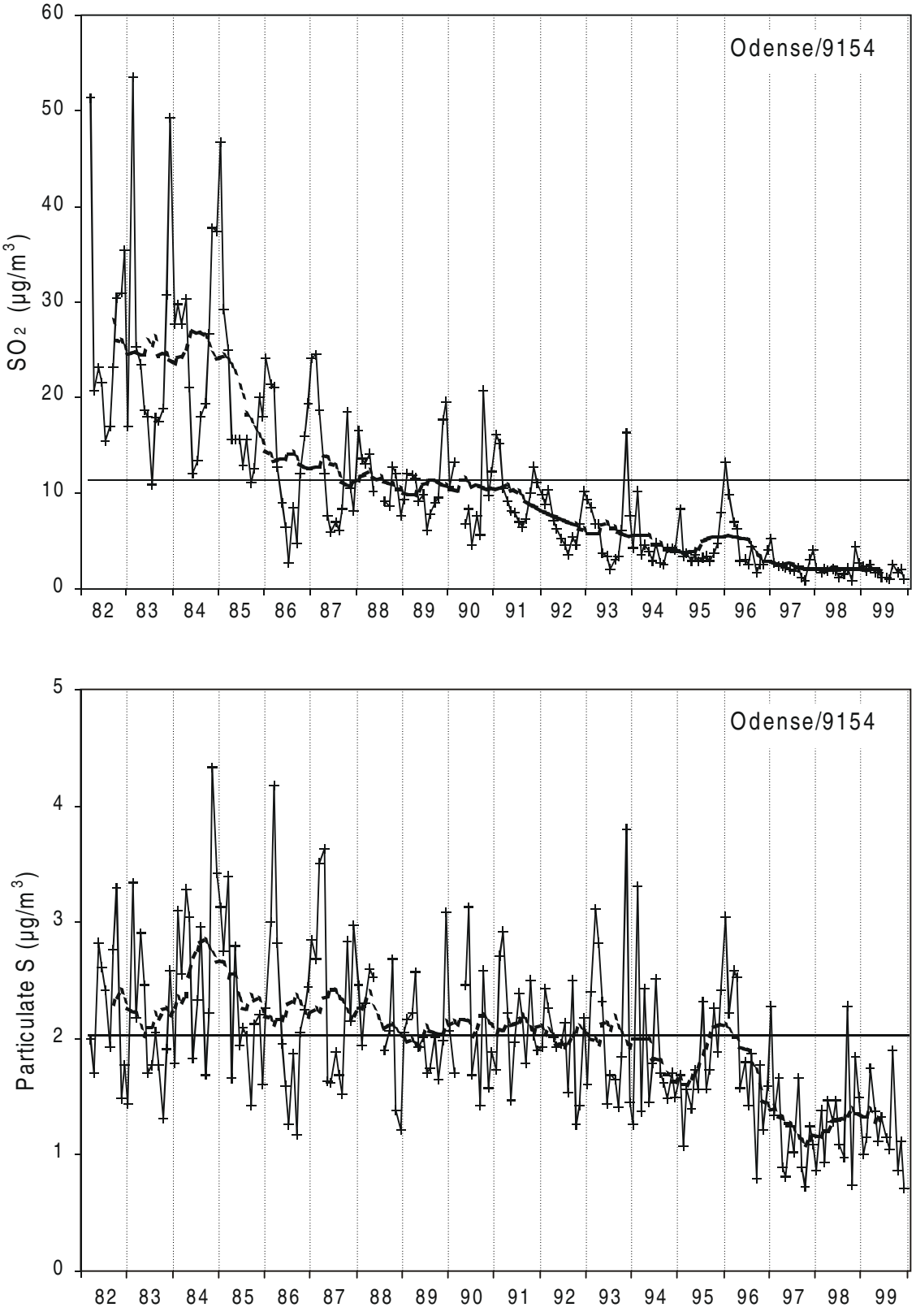


Figure 4.4 Trend for SO_2 and particulate S measured at Odense/9154. The points are measured monthly averages, the dotted curve is a moving average over 13 month and the straight line represents the average over the whole period.

Up to 1996 particulate S has, in contrast to SO_2, shown only a slightly downward trend. This is probably because the sulphur emission has

been reduced more in Denmark than in our neighbouring countries. The amount of particulate S may also be limited by the amount of compounds in the atmosphere that are able to oxidate SO_2. From 1996 to 1997 the concentrations of particulate S were reduced with more than 30 %. This is, as for SO_2, mainly caused by a reduction of the contribution from Southeast (Kemp, Palmgren and Manscher, 1998). It will be interesting to follow the development the following years to see to what extent the decrease in concentrations is a result of reduced sulphur emissions.

5 Suspended particulate matter

The total suspended particulate matter (TSP) is determined by weighing of the aerosol filters. The samplers collect particles up to an aerodynamical diameter of around 25 µm, but this cut-off varies from about 10 to 50 µm depending on the wind speed (Kemp 1993). The TSP samplers will be replaced by PM_{10} samplers at all stations within the next one to two years.

Particle size and sources

The particles are a mixture from the different source types, but the coarse particles (> 2 µm) of windblown dust of local origin are expected to dominate. The fine particle fraction includes contributions of long range transported soil dust and particles from combustion processes, e.g. sulphate and nitrate particles. These particles are in the size range between 0.1 and 1 µm. A major contribution to particulate pollution in urban areas is believed to be attributed to traffic, and especially, to emissions from diesel powered vehicles. Ultra-fine particles emitted from petrol as well as diesel engines are formed at high temperature in the engines, in the exhaust pipe, or immediately after emission to the atmosphere. Some of these particles may be in the so-called nucleation mode (nano-particles < 50 nm). The dominating ultra-fine particle mode has a number concentration peak in the range 50 nm - 100 nm. The particles are formed by coagulation of primary particles, and by condensation of gases on particles. The mass of the ultra-fine fraction is only a small part of the total particulate matter (TSP or PM_{10}). The ultra-fine particles may however be important for the health effects of ambient particles. Although measurements of the ultra-fine particles are not a part of the LMP programme results of some recent measurements are described in section 5.5.

Sites

TSP was in 1999 measured as 24 hour average values at Copenhagen/1257, Odense/9155, Odense/9154, Aalborg/8151 and Lille Valby/2090. The measurements at Lille Valby started in the beginning of 1995. Continuous measurement of PM_{10} was started in July 1998. Sampling in 24 hour intervals is performed using an OPSIS SM200 sampler at Copenhagen/1257. The particles are collected on Membrane filters (Millipore type RA). The PM_{10} is determined both on-line with a build-in β-gauge and gravimetric, using the same procedure as for TSP. The PM_{10} results are described in section 5.4.

5.1 Annual statistics

Limit values

The limit values in force in Denmark (Miljøministeriet 1986) are based on EEC directive (EEC 1980). The limit values and the relevant statistical parameters for 1999 are given in table 5.1. The annual 95-percentiles and average values are shown for 1988-1999 in figure 5.1.

Table 5.1 Average values, 95-percentiles and maximum values for TSP in 1999. The numbers are calculated for 24 hour average values.

		TSP (µg/m^3)			
Station	Number	Average - whole year	95-perc.	Max. value	Day
Copenhagen/1257	356	47	97	159	990331
Odense/9155	359	49	104	269	990129
Odense/9154	362	44	89	125	990331
Aalborg/8151	358	51	99	153	990403
Lille Valby/2090	360	24	54	79	990918
Limit value	min. 100	150	300	-	-

Measured values

The measured values at the urban stations were between 1/4 and 1/2 of the limit values. The 1999 results were not much different from the 1998 results. The general trend has been slightly decreasing since 1988 (figure 5.1) and there is no reason to believe that it will change much within the coming years. A major part of the collected particles are windblown dust and may be considered to be either of "natural" origin, constructions or resuspended particles from the roads. The particles from combustion processes are in the fine particle fraction, and it is expected to decrease in the future due to reduction of the emission as a result of i.e. better cleaning of the smoke from power plants, obligatory TWC on petrol cars and restrictions on the diesel exhaust.

5.2 Episodes

The measured maxima of the daily average values are given in table 5.1. TSP has been measured at the rural station Lille Valby/2090 for almost 5 years. The results are in average around one half of the results from the urban street stations. But during episodes, when pollution is transported from the European continent, the difference is reduced and a major part of the TSP even at street stations are coming from sources outside the cities. This is illustrated on fig. 5.2 for two episodes in September 1999. The maximum concentrations at Odense/9155 on January 29 was a one-day episode. It was probably caused by local activity in the street where the measuring station is placed.

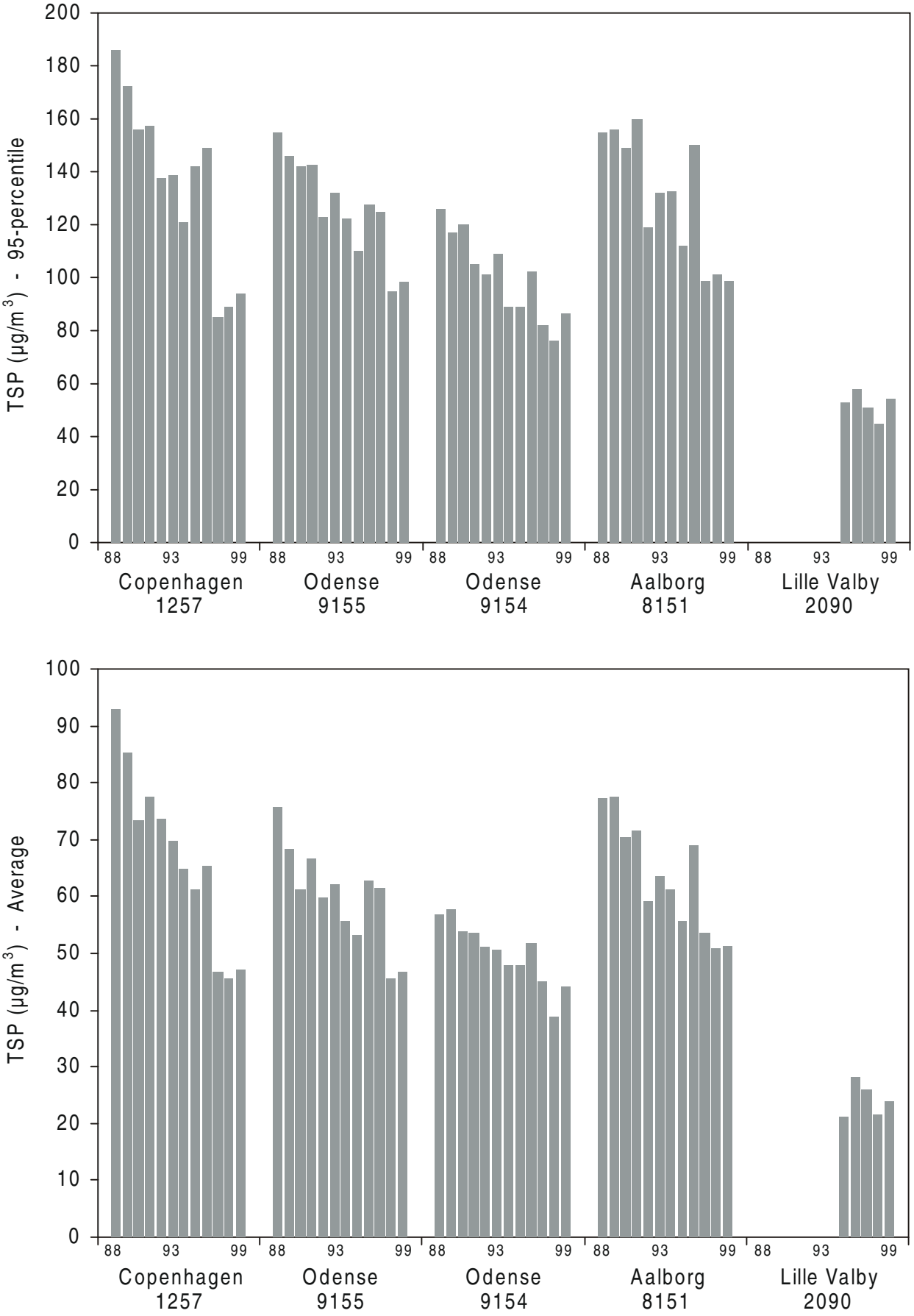


Figure 5.1 Average values and 95-percentiles for TSP from 1988 to 1999.

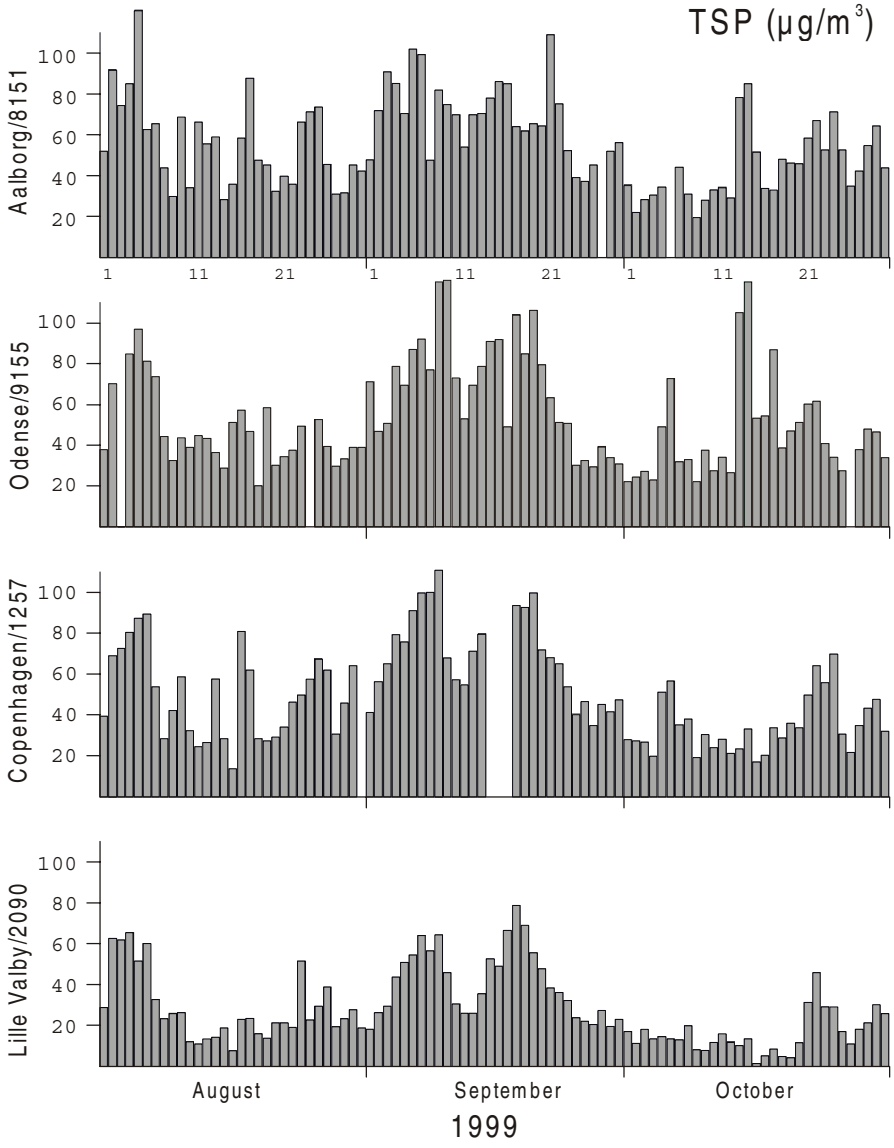


Figure 5.2 TSP episodes in September 1999.

5.3 Trends

Percentiles

The annual percentiles and average values based on daily average TSP concentrations measured at Aalborg/8151 are shown in figure 5.3. The level of TSP has been decreasing since 1986. The ratio between the "short term" values (95- and 98-percentiles) and the "long term" values (median and average) are almost constant in contrast to the case for SO_2 where the decrease was steeper for the long term than for the short term values.

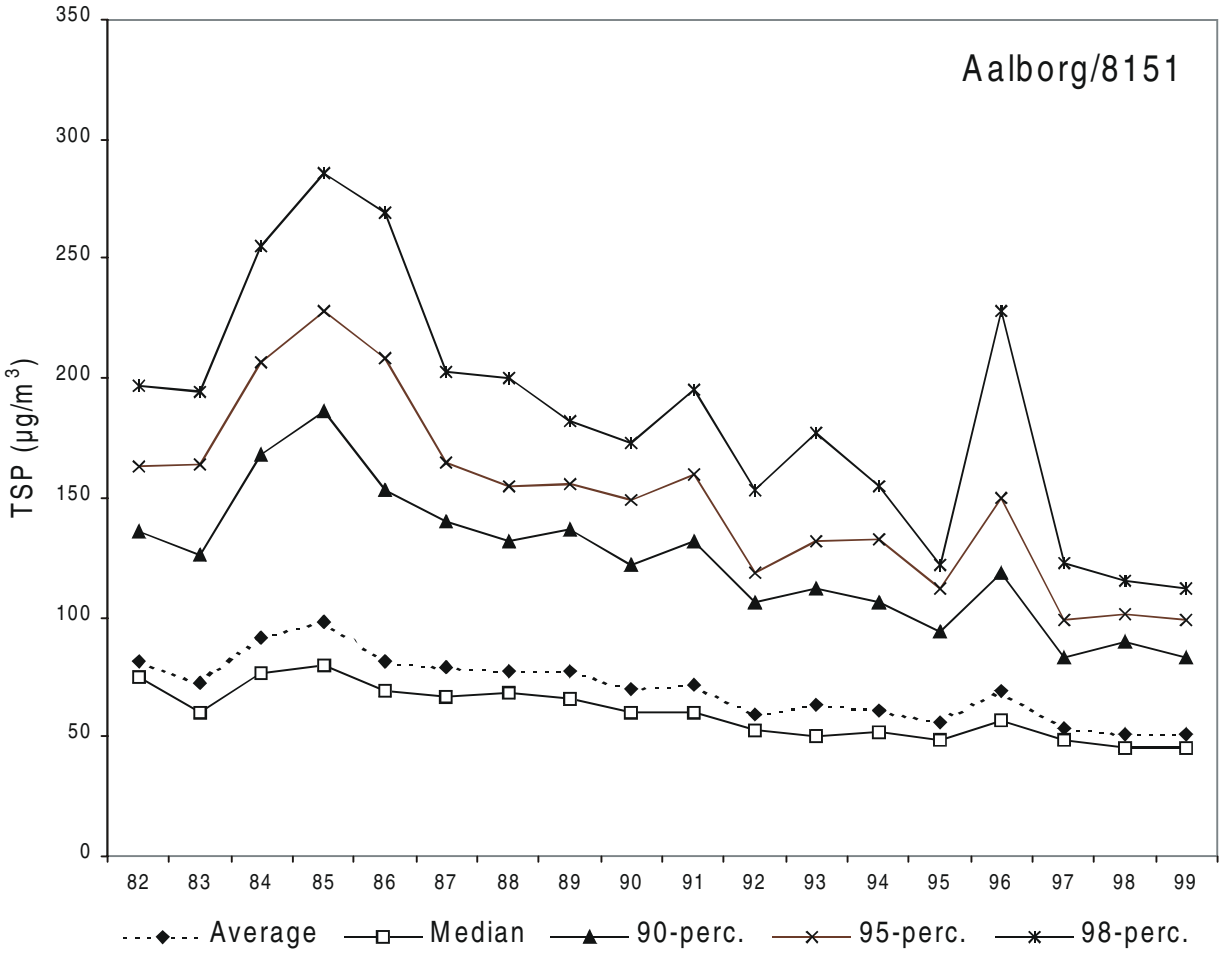


Figure 5.3 Trends for annual averages, median, 90-, 95 and 98-percentiles for TSP measured at Aalborg/8151.

Annual average

After relatively high concentrations from 1984-1986 there has been a continuous decrease since 1986 (figure 5.4). This may be a result of better emission control at power plants and other large combustion installations and the substitution of oil with natural gas for domestic heating. Other factors like the obligatory demand for "winter crops" from 1987 may, as discussed in the annual report for 1993 (Kemp, Palmgren, Manscher 1994), also play a role.

Annual variation

The highest concentrations were found in the early spring and in the autumn. The low summer concentrations were a result of generally lower wind velocities during the summer and a lower level of activity in the city areas in the holiday period around July, while occasional snow periods kept the dust grounded in the winter months.

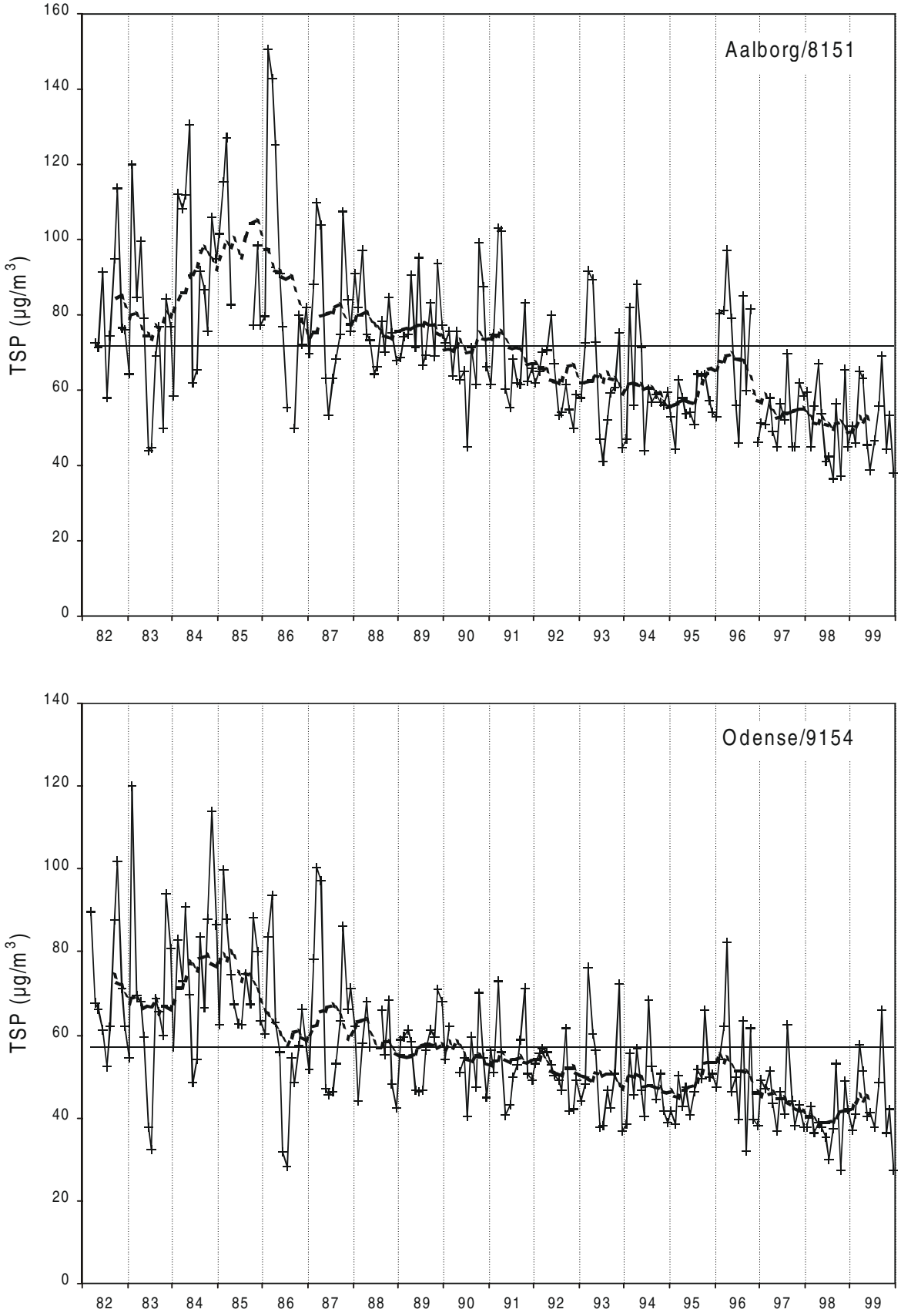


Figure 5.4 Trend for TSP measured at Aalborg/8151 and Odense/9154. . The points are measured monthly averages, the dotted curve is a moving average over 13 month and the straight line represents the average over the whole period.

5.4 PM_{10}

New limit values

The EU Council has adopted a Directive giving new limit values based on PM_{10}, i.e. particles having an aerodynamical diameter <10 µm (EC, 1999). The limit values shall be met in two stages (see table 5.2). It is allowable that the limit values are exceeded if:

- It is owing to concentrations in ambient air due to natural events, which result in concentrations significantly in excess of normal background levels from natural sources. In such cases member states shall provide necessary justification to demonstrate that such exceedances are due to natural events.

- Member states have designated zones or agglomerations within which the limit values are exceeded due to resuspension of particles following winter sanding of roads. Member states shall provide the necessary justification to demonstrate that exceedances are due to resuspension and that reasonable measures have been taken to lower the concentrations.

The directive also imposes the member states to perform measurements of $PM_{2.5}$ at representative places and report the results of these measurements to the Commission.

Table 5.2 EU limit values for PM_{10} (EC, 1999). All values are for protection of human health.

		Averaging	Limit value	Date by which limit value is to be met
Stage 1	24 hour limit value	24 hours	50 µg/m^3 not to be exceeded more than 35 times per year (90.4 percentile)	January 1, 2005
	Annual Limit value	calendar year	40 µg/m^3	January 1, 2005
Stage 2([1])	24 hour limit value	24 hours	50 µg/m^3 not to be exceeded more than 7 times per year (98 percentile)	January 1, 2010
	Annual Limit value	calendar year	20 µg/m^3	January 1, 2010

([1]) Indicative limit values to be reviewed in the light of further information on health and environmental effects, technical feasibility and experience in the application of stage 1 limit values in the member states.

These PM_{10} values are not directly comparable with the LMP results, which are based on TSP. Preliminary measurements indicate that the PM_{10} values are about 75% of the measured TSP concentrations (see below). Having that in mind it seems like the concentrations at the traffic sites are somewhat higher than the new limit values.

PM_{10} and health

The PM_{10} was chosen as basis for the limit values because there exists a comprehensive collection of measurements. The only large epidemiological studies on health related effects of particles in outdoor air are based on PM_{10} measurements. These studies indicate a correlation between the concentration of PM_{10} in the atmosphere and the frequency of respiratory diseases. It has not been possible to derive a threshold below which no effects occur and the exposure-response curve fit probably a straight line reasonably well at

concentrations below 100 µg/m^3. (WHO, 2000). The relative risk is estimated to 1.043 per 10µg PM_{10}/m^3 for long-term exposure. This leads to an estimate of around 400 premature deaths per year per one million inhabitants older than 30 years. It is, however, not clear which effects are important for the health. The fine particles are the potential most harmful, because they penetrates deep into the lungs. The ultra fine particles (0.01-0.1 µm) from diesel vehicles, which are emitted in a great number but carries little mass, and the secondary aerosols, which are formed by oxidation of e.g. SO_2 and $NO_{x,}$ may be the most harmful. It is expected that the knowledge in this field will be extended very much within the next few years. The PM_{10} based limit values may be considered as interim limit values until we have gained the necessary knowledge (cf. footnote to table 5.2).

PM_{10} measurements

PM_{10} measurements were started in July 1998 at Copenhagen/1257 in parallel to the ongoing TSP sampling. The PM_{10} particle concentration is determined both gravimetrically and with a β-gauge and the filters were analysed with PIXE, for the elemental composition of the dust, like the TSP filters. Some results of the parallel measurements are presented in the following.

The β-gauge is an indirect way of measuring the dust weight. The β absorption depends on the mass-thickness of the dust as well as on the elemental composition of the dust. Assuming that the composition does not vary very much, it is possible (with a suitable calibration) to obtain a good estimate of the thickness by measuring the difference between the absorption in the filter before and after exposure.

Annual statistics

The annual results in table 5.3 show that the average is below the stage 1 limit value, while the 24 hour limit value is exceeded. Both stage 2 limit values are exceeded. If the slightly downward going trend for TSP is continued and it is applicable for PM_{10} as well, there might be a chance that the stage 1 values are met by 2005, while it seems very unlikely that the stage 2 values will ever be met. The difference between the gravimetric and the β-gauge results is around 10%.

Table 5.3 Statistical parameters for PM_{10} measurements at Copenhagen/1257 during 1999.

	PM_{10} (µg/m^3)				
	Num-ber	Average - whole year	90-percentile (≈ 36th highest value)	98-percentile (≈ 8th highest value)	Max concentration
Gravimetric	319	33	57	71	98
β-gauge	319	36	62	81	115

Figure 5.5 shows a regression plot between the β-gauge and the gravimetric results. In average the results are very well correlated. A slope of 1.1 is to some extend compensated by a zero shift of 1.3 µg/m^3 on the x-axis. This may be a result of the conditioning of the

filters at 52% RH at room temperature prior to weighing, while the β-gauge measurement is carried out at 40^0 C under ambient humidity.

The comparison between simultaneous measurements of PM_{10} and TSP (fig. 5.6) shows a fairly good correlation. The PM_{10} results are in average around 65 % of the TSP.

The difference between PM_{10} and TSP is of course the particles >10 µm. These particles are expected to be mainly windblown dust and, at the street stations, coarse particles raised by the traffic. They will mainly be composed of soil minerals. There is no measurable difference between the S content in PM_{10} and TSP. The S is mainly present in submicron particles as SO_4^{--}, which is formed in the atmosphere by oxidation of SO_2. Typical soil elements as Si, Ti and Ca are found in almost around twice as high concentrations in TSP as in PM_{10} (Kemp and Palmgren, 1999).

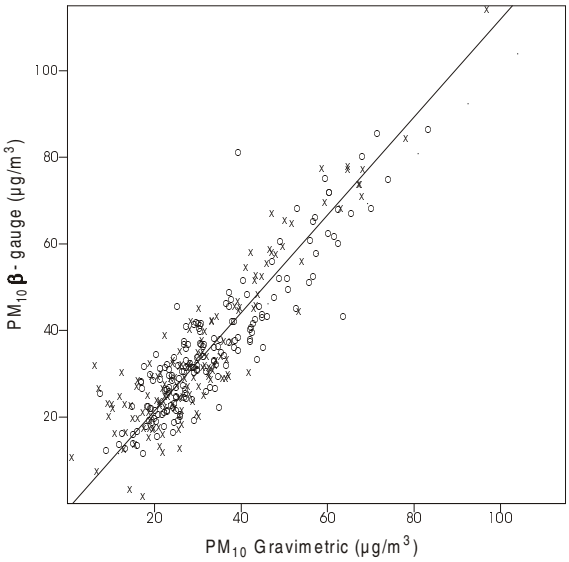


Figure 5.5 Comparison between β-gauge and gravimetric determination of the PM_{10} concentration. The dotted line is 1:1 and the drawn line is the orthogonal regression line: y = 1.13 x – 1.3 µg/m^3 (ρ=0.90).

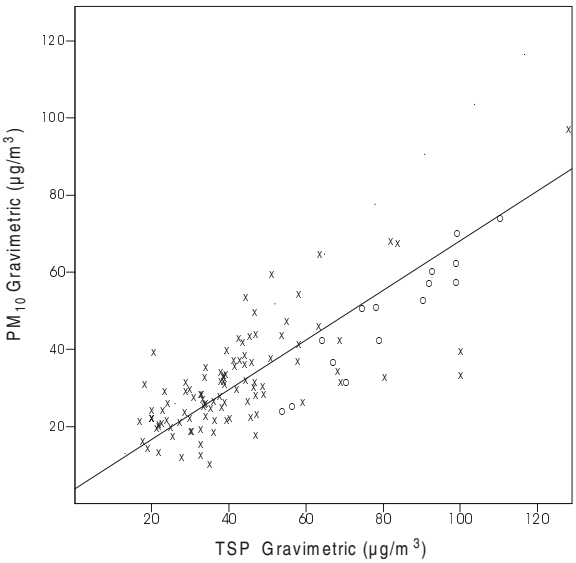


Figure 5.6 Comparison between TSP and PM_{10}. The dotted line is 1:1 and the drawn line is the orthogonal regression line: y = 0.64 x – 3.8 µg/m^3 (ρ=0.79).

5.5 Ultra-fine particles

Health effects of ultra-fine particles

It has been shown that the correlation between particle concentration and health effect increases with decreasing particle diameter (Dockery et al., 1993, Pope et al., 1995). WHO has not recommended a limit value for particulate matter, because no lower adverse effect level has been identified and more research is necessary.

The particle size distribution is thus an important factor that needs to be addressed whenever the PM pollution is concerned. Important properties - in addition to size - are state (liquid/solid), volatility,

hygroscopicity, chemical composition (content of organics, metals, salts, acids etc.), morphology, and density. These properties are also important for selection of methods for regulation and control of emissions. The ultra-fine particles in urban air are mainly emitted from diesel and petrol fuelled vehicles and the test cycles on the emission from the engines can be based on measurements on dynamometers using dilutions systems, which, however, not always are comparable with the real emission conditions in a street. It is therefore important to measure and determine the particle emission under normal driving conditions and in ambient air, in order to be able to establish the relationship between the sources and the exposure of the population. Results from studies of ultra-fine particles in urban streets are presented in the following.

Ultra-fine particles emitted from petrol as well as diesel engines are formed at high temperature in the engines, in the exhaust pipe, or immediately after emission to the atmosphere. Some of these particles may be in the so-called nucleation mode (nano-particles < 50 nm). The dominating ultra-fine particle mode has a number concentration peak in the range 50 nm - 100 nm.

Measurements of size distribution

The measurements were performed by a Differential Mobility Analyser (DMA) at Copenhagen/1257 and Odense/9155 (Wåhlin et al. 2001). The DMA is able to determine the size distribution of particles in the size range 10-700 nm.

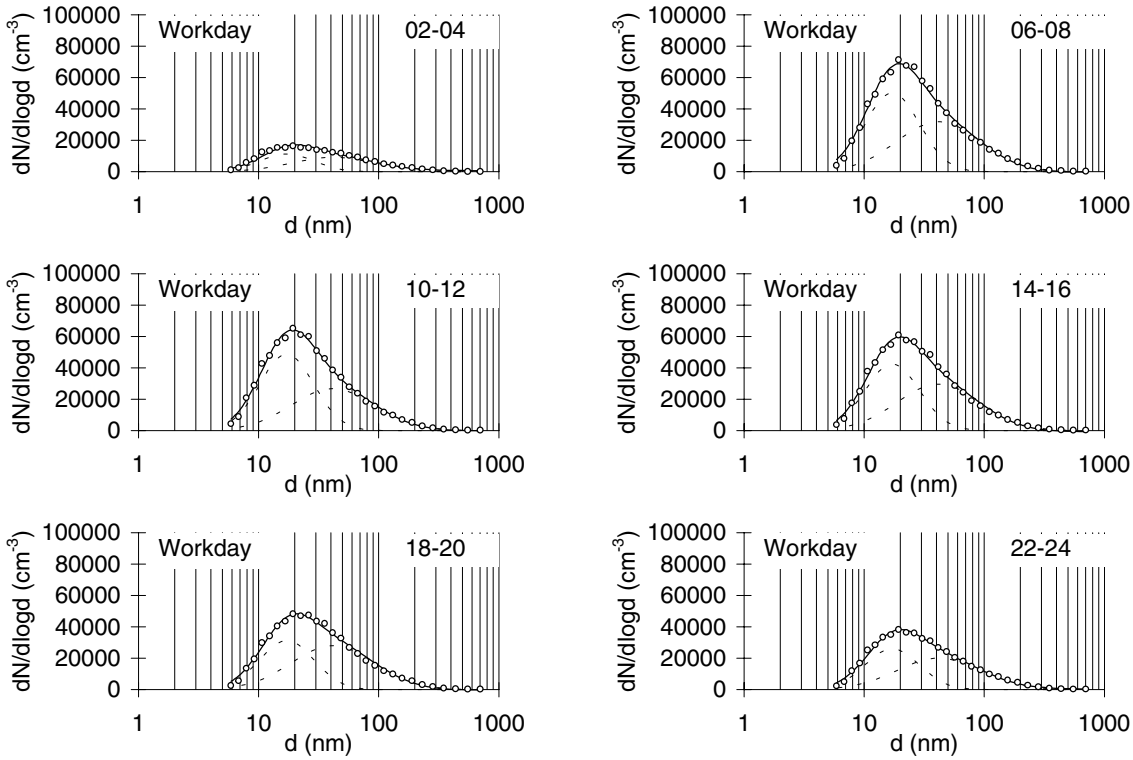


Figure 5.7. Average particle size distributions at Copenhagen/1257 during specific workday two-hour periods. The distribution fitted with the same two log-normal distributions to elucidate the differences in shape.

Two-hour average particle spectra representing all workdays at Jagtvej are shown in Figure 5.7. Although the shapes seem very similar with a peak of the size distribution at approx. 20 nm, small

differences exist with shifts to a finer average size in the spectra collected during working hours when diesel traffic is relatively dense. Similar data are available for Odense/9155 and the urban background station Copenhagen/1259. Figure 5.8 shows the different average size distributions during the respective sampling periods at the three locations.

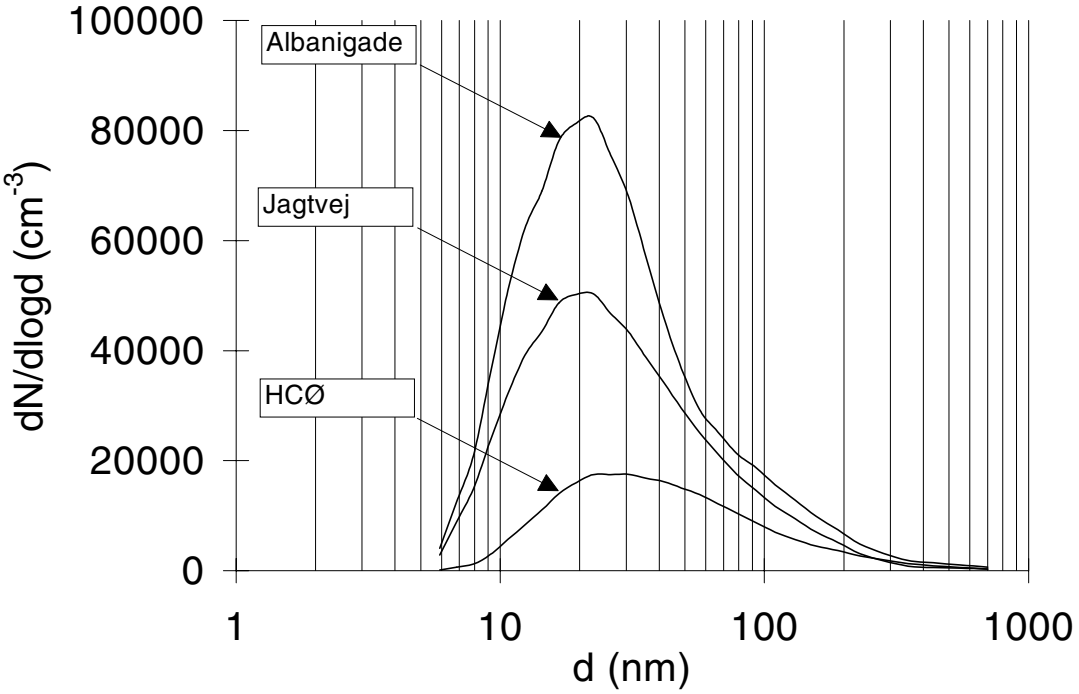


Figure 5.8. The average particle number distributions for Odense/9155 (Albanigade), Copenhagen/1257 (Jagtvej), and Copenhagen/1259.

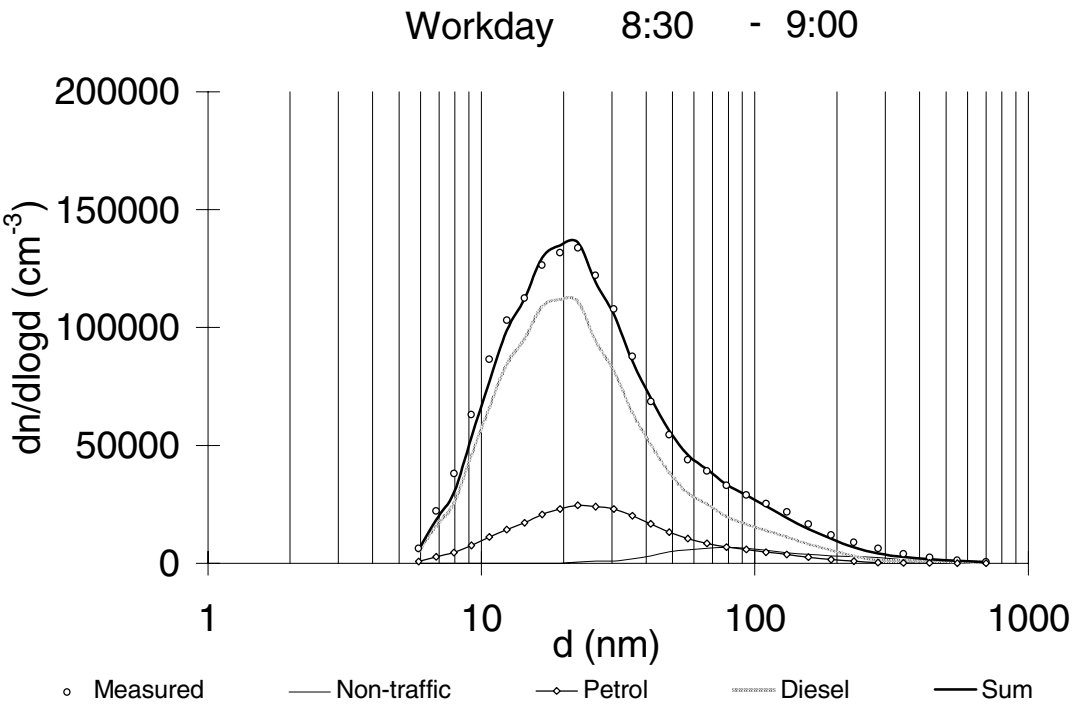


Figure 5.9. The average particle size spectrum at Odense/9155 during morning rush hours resolved as the sum of spectra from different sources.

Average weekly cycles of the entire measurement periods at the street stations of particles, NO_x and CO concentrations were used for the analysis (the cycle of total particles for Odense/9155 is shown in Figure 5.10). A clear diurnal variation of the three parameters with a sharp rush hour peak in the morning and another rush hour peak during the afternoon was observed on workdays. The pattern is different on Saturdays and Sundays. Although the correlations between particles, NO_x and CO are generally good, some deviations were observed. The differences can be related to differences in the traffic patterns of petrol and diesel vehicles (diesel taxies compared with other traffic culminate at night, petrol cars during rush hours).

A technique developed at NERI (Wåhlin et al. 2001) has been used to determine the contribution from different sources, i.e. petrol vehicles, diesel vehicles and non-traffic sources, to the particle pollution at different particle sizes. A particle size spectrum with the contributions from the different sources at Albanigade is shown in Figure 5.9. The average size of the particles emitted by the diesel vehicles was considerably smaller than the size of particles emitted by the petrol vehicles. The contributions from the different sources during the average week at Odense/9155 are also shown at Figure 5.10.

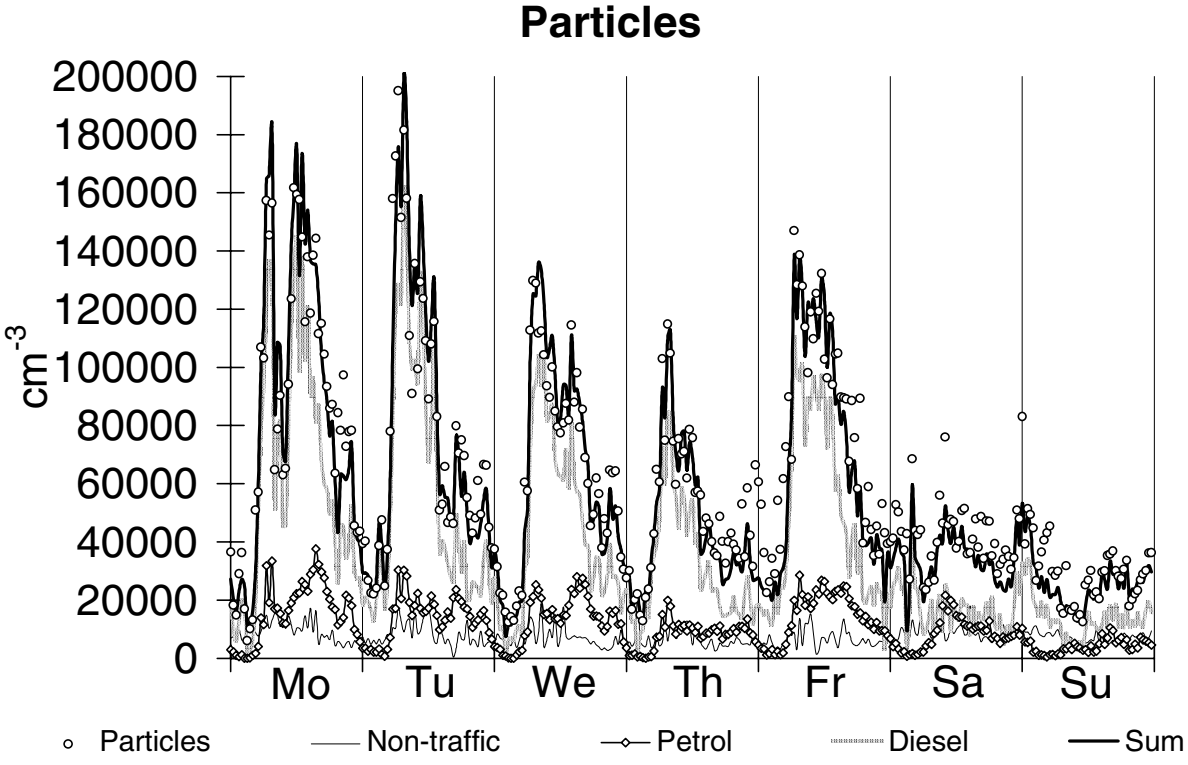


Figure 5.10. The average weekly cycle of particle concentrations measured at Odense/9155, 03.05.99-20.05.99, and the fitted contributions from the different sources.

6 Elements

The aerosol samples are analysed by means of a true multi-element method (PIXE) (Kemp 1993). About 20 elements are found in concentrations above the detection limit in almost all samples, while about 5 more elements are found frequently. The elemental composition of the aerosols was in 1999 measured at all five stations in operation. In addition to monitoring purposes the measurement of the elements may be used for evaluation of the different

Table 6.1 Average values for 1999. All concentrations are given as ng/m^3 (1 µg/m^3 = 1000 ng/m^3). N_{tot} is the number of measurements in 1999. N_0 is the number of measurements above the detection limit. The arithmetic mean values are calculated for the measured concentrations, if more than 90% of the measurements were above the detection limit. If less than 90% of the measurements were above the detection limit, a fit to a log-normal distribution is calculated based on the values above the detection limit. The values in the tables represent in these cases the arithmetic mean value for the fitted distribution. The method is under normal conditions reliable if less than half of the measurements are below the detection limit and may in any case give an impression of the average values even if it is based on a few values only.

	Copenhagen/1257		Odense/9155		Odense/9154		Aalborg/8151		Lille Valby/2090	
Element	N_o	Average	N_o	Average	N_o	Average	N_o	Average	N_o	Average
Al	354	512.0	355	537.0	355	420.0	358	492.0	326	189.0*
Si	356	944.0	352	1080.0	357	733.0	356	926.0	287	324.0*
S	356	1220.0	360	1210.0	362	1210.0	358	1180.0	363	991.0
Cl	356	1560.0	360	2310.0	362	1520.0	358	2790.0	352	837.0
K	356	321.0	360	357.0	362	304.0	358	314.0	363	196.0
Ca	356	1070.0	360	1090.0	362	780.0	358	1020.0	362	341.0
Ti	356	49.1	360	54.6	361	43.2	358	45.1	352	11.4
V	354	6.6	353	4.8	356	4.9	342	4.4	355	4.3
Cr	353	7.3	353	6.0	356	7.4	357	5.2	192	1.0*
Mn	356	18.2	360	23.0	362	26.2	358	12.7	358	4.0
Fe	356	1120.0	360	910.0	362	675.0	358	930.0	363	134.0
Ni	355	3.5	360	2.9	361	2.6	358	2.9	361	1.7
Cu	356	57.2	360	33.6	362	22.8	358	41.2	354	3.9
Zn	356	54.3	360	60.7	362	46.2	358	55.0	362	21.0
As	226	1.3*	226	1.2*	237	1.0*	208	1.0*	223	0.8*
Se	187	0.4*	225	0.5*	266	0.5*	196	0.4*	292	0.5*
Br	356	4.6	360	4.7	362	4.7	358	5.4	363	3.7
Sr	356	4.3	358	4.0	357	3.1	358	4.5	315	1.4*
Zr	337	3.8	312	3.2*	326	2.4	343	3.1	63	0.4*
Mo	283	3.2*	207	2.2*	192	1.5*	276	2.5*	9	0.3*
Cd	20	0.7*	18	0.5*	20	0.7*	16	0.7*	17	0.6*
Sb	310	16.0*	241	9.7*	207	6.0*	290	10.3*	26	1.3*
Ba	294	29.2*	225	24.9*	237	16.7*	282	25.0*	16	3.3*
Pb	356	16.6	360	13.6	361	10.7	358	12.5	353	7.2
Ntot	356		360		362		358		363	

* Calculated from a fit to a log-normal distribution.

source types that contribute to the pollution, because many of the elements mainly come from a single source type. For instance combustion of heavy oil is the main source of V and Ni, wind blown dust of Si, and particulate S is mainly caused by long range transport.

6.1 Annual statistics

The average values for the elements are listed in table 6.1.

New Years day

New years days is an exception from the general trend for Pb due to the traditional launching of fireworks and occasional bonfires in the streets. A comparison between the annual averages and the concentrations New Years day shows that the concentration New Years day apparently have been increasing the last few years, probably as consequence of increased import of fireworks. In year 2000 it literally exploded. Pb is only present in imported, i.e. mainly Chinese, fireworks, as it is illegal to use Pb in fireworks produced in Denmark (Barfod, 1998). The contribution this one day may influence the annual average with some 5-10% the previous years and almost 50% in 2000. A legally implemented reduction of Pb in fireworks is in preparation.

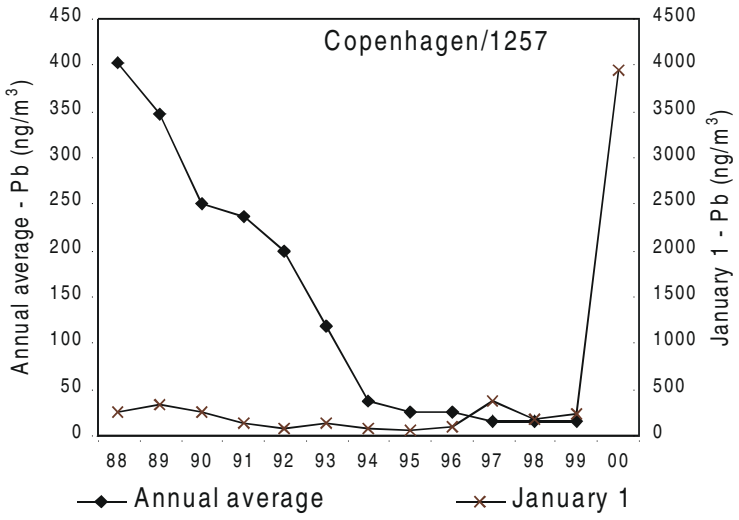


Figure 6.2 Measured concentration New Years day and annual averages at Copenhagen/1257.

6.2 Heavy metals in urban and rural areas

Several of the elements are toxic heavy metals. WHO has assessed the toxic effects of some heavy metals (WHO 1987, WHO 2000). These are Cr, V, Mn, Ni, As, Cd, Hg and Pb. Guidelines are suggested for V, Mn, Cd, Hg and Pb. Cr, Ni, and As are carcinogenic in certain compounds and only life time cancer risks are estimated. A daughter directive under the EU framework directive (EU 1999) is under preparation. Limit values for Ni, As and Cd will be introduced in the Directive, which is expected to be adapted in 2001 or 2002.

Heavy metals from the traffic

Increased concentrations of Cr and Cu are observed in 1999 at the traffic sites (fig. 6.3). Wind direction analysis of the measurements shows that traffic is the main source for these elements at street level. Brake pads contain 10-20% Cu and other metals as Cr, Zn and Pb in smaller amounts (Westerlund, 1998). The relation between the traffic and Cu and Cr is illustrated on fig. 6.4. NO_x is used as indicator for the traffic and the close correlation between NO_x and Cu therefore indicates that the traffic is the main source for Cu. Further the close correlation between Cu and Cr indicates that the two elements have the same sources. It can be seen from fig. 5.4 that the ratio between NO_x and Cu in general is higher in winter than in summer. This may be a result of lower fuel efficiency in the winter.

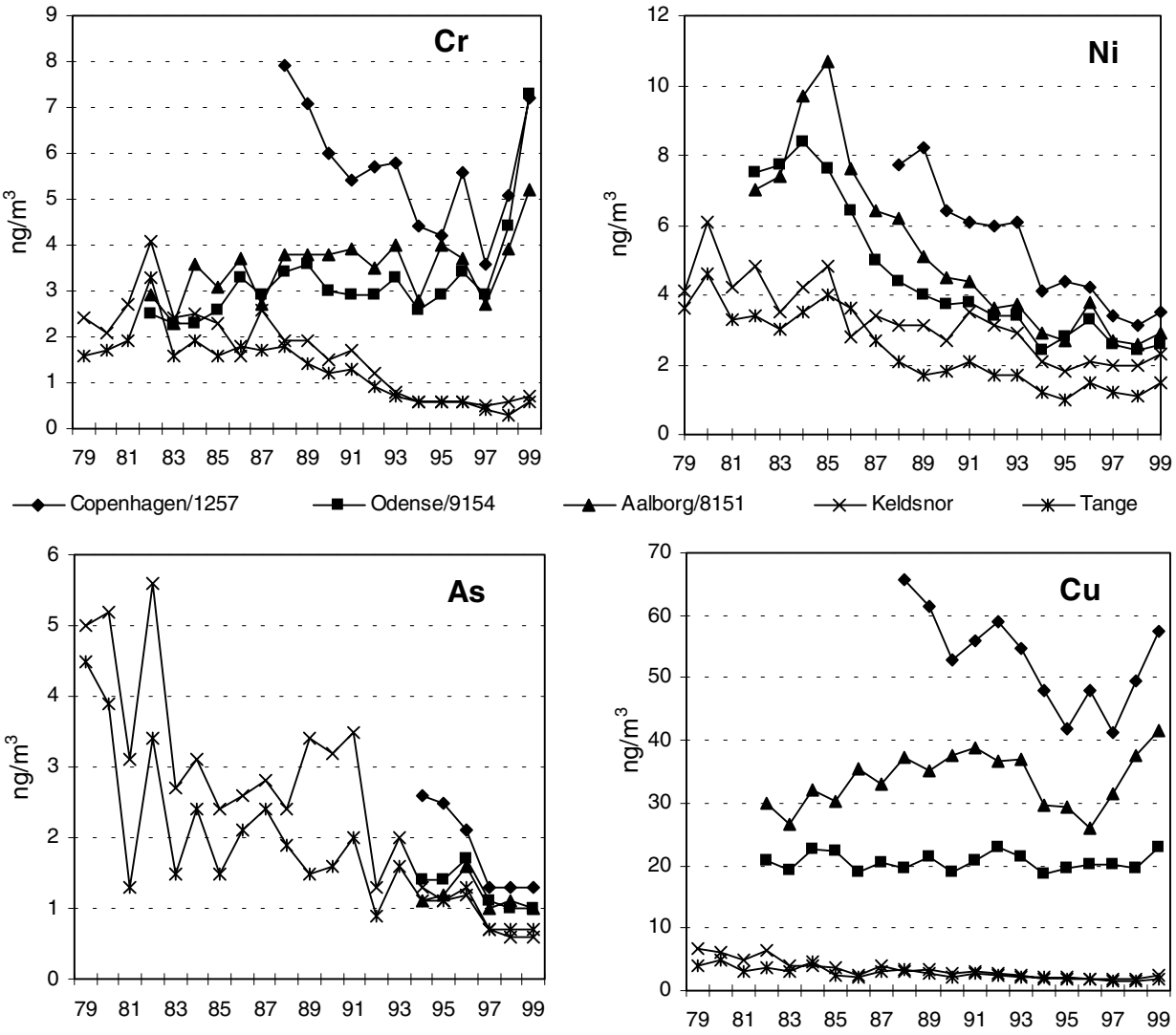


Figure 6.3 Yearly average values some heavy metals. The stations in Copenhagen, Odense and Aalborg are placed in busy streets.Results from the stations in the Danish Background Monitoring Program at Tange and Keldsnor are included for the comparison.

Non-carcinogenic heavy metals

Table 6.3 shows the WHO guidelines for yearly averages for the non-carcinogenic elements together with the corresponding measured values at Aalborg in 1982 and 1999 and at Copenhagen/1257, where in most cases the highest values are measured. Apart from Pb, 10 years ago, all measured values are more than a factor of 10 below the

guideline values. Hg is not measured within the LMP programme, but estimates of the Hg pollution in the air in Europe indicates that the concentrations are between 0.001 and 20 ng/m^3 (Lahmann et al. 1986). The amount of V in urban air has decreased by a factor of 3-5 since 1982. It followed the reduction in the SO_2 concentrations and it is expected to decrease further in the future. Mn does only show a slightly decreasing trend.

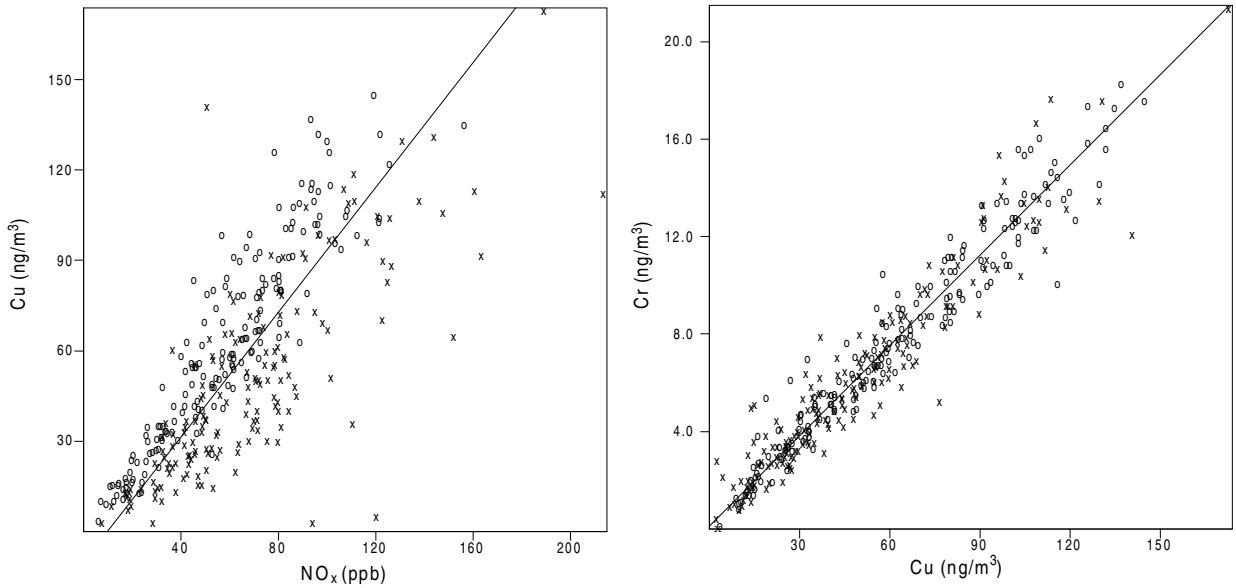


Figure 6.4 The correlation coefficient (ρ) between NO_x and Cu is 0.79, while it is 0.96 between Cu and Cr. The drawn lines are the orthogonal regression lines. The points represent 24 hour averages in 1999 measured at Copenhagen/1257. o = summer and x = winter results.

Table 6.3 WHO guidelines (WHO 1987 and 2000) for non-carcinogenic heavy metals compared to measured annual average concentrations at street level. N.M. = not measured.

ng/m^3	WHO guideline	Aalborg/8151	Aalborg/8151	Copenhagen/1257
		1982	1999	1999
V	1000	22	4.4	6.6
Mn	150	20	13	18
Cd	5	<2	0.7	0.7
Hg	~1000	N.M.	N.M.	N.M
Pb	500	1100	13	16

Carcinogenic heavy metals

The estimated human lifetime risks are estimated for air concentrations of 1 µg/m^3. These values and the corresponding urban concentrations are shown in table 6.4. The evaluation of the lifetime risks is very uncertain and assessment of acceptable risks is debatable. The risks of concentrations at the measured levels are calculated assuming the dose-response curve can be extrapolated linearly towards zero (see below). Fig. 6.3 shows the trends for the LMP stations and the stations in the Danish Background Monitoring Program with long time records.

Table 6.4 Estimated lifetime risks (WHO 2000) for carcinogenic heavy metals compared to measured concentrations.

	Lifetime risk at 1 µg/m^3	Aalborg/8151 1982 µg/m^3	Aalborg/8151 1999 µg/m^3	Copenhagen/1257 1999 µg/m^3
Cr	1)	0.0029	0.0052	0.0073
Ni	3.8×10^{-4}	0.0070	0.0029	0.0035
As	1.5×10^{-3}	<0.02	0.0010	0.0013

1) the WHO estimated life time risk for Cr(VI) is 4×10^{-2}, while the measurements are total Cr (see text).

Chromium

Only hexavalent Chromium (Cr(VI)) is carcinogenic, while the most abundant trivalent Chromium (Cr(III)) is relatively harmless. Little is known about the fraction of Cr(VI) in the ambient atmosphere, but it is expected to amount to a very small part of the total Cr because Cr(VI) is easily reduced to Cr(III). A downward trend at the background stations and an almost constant level at the urban stations indicate that the sources for Cr are mainly urban. As discussed above there was a marked increase in 1999 compared to the previous years at the urban stations.

Nickel

A linear extrapolation indicates that the measured Ni concentrations correspond to a life time risk above 10^{-6}. The estimate becomes however further uncertain because the carcinogenic effect of various Ni compounds is very different and the partitioning between the different compounds in the air is not known. The WHO estimate is based on epidemiological data for workers at Ni refineries. The Ni pollution has been decreasing at all stations and it is expected to be further reduced. The major part of the Ni is emitted from oil burning and the trend is following that of SO_2 (see chapter 4). The difference between the concentrations at urban and background stations is decreasing, which indicates that the major part is long range transport.

Arsenic

The detection limit for determining As has unfortunately previously been too high to give reliable estimates of the yearly average values at the urban stations. However, data from 1994 and on are available due to the decreasing Pb concentrations (Pb interferes with the determination of As) and an improved data treatment method. The records go back to 1979 at the background stations and shows that the concentration has been reduced by a factor of 3 since 1979. The values for 1999 are almost equal at the background stations and the street stations in Odense and Aalborg, while the values in Copenhagen are somewhat higher. At present the concentrations correspond to a lifetime risk above 10^{-6}. It can, as for Ni, be concluded that a major part of As is long range transported.

Summary

If the risks for the three elements are added the total lifetime risk may be estimated to more than 10^{-6}. The variation of the concentration from station to station indicates that the occurrence of Cr is widespread over urban areas and Ni and As over the whole country.

7 CO and VOC

Continuous measurements of CO have been performed since the beginning of 1996 at the three traffic stations Copenhagen/1257, Odense/9155 and Aalborg/8151 together with the urban background station Copenhagen/1259. Measurements of the VOC's benzene, toluene and xylene (BTX) have been performed at the traffic stations Copenhagen/1257 and Odense/9155 for a couple of years. The measurements have been possible by integration of the LMP and TOV (Berkowicz et al. 1996) measurements partly funded by the Ministry of Traffic. In addition the BTX measurements are partly funded by the Danish Environmental Protection Agency. The behaviour of the three VOC's is very much alike. Only benzene and toluene will be discussed in the following. Benzene is carcinogenic, while the most severe effect of toluene is damage to the central nervous system.

Sources

The main source of CO, benzene and toluene is petrol vehicles. The CO is emitted due to incomplete combustion of the petrol. The TWC will remove the major part of the CO. There is practically no CO in the exhaust from diesel vehicles. It has been estimated that of the total hydrocarbon emission from motor vehicles in Denmark only about 9% came from diesel vehicles while petrol fuelled vehicles accounted for the remaining 91%; of the latter, about 59% came from the exhaust while 41% was released by evaporative emission (Danmarks Statistik 1993). The hydrocarbon content of vehicle exhausts are influenced by several factors related to driving mode and speed, ambient temperature, vehicle conditions (e.g. age and performance), fuel to air ratio and fuel type. A limit of 5% content of benzene has been introduced in 1986 in all EU-countries. The petrol in Denmark was produced with a benzene content between 3 and 5% depending on the content in the crude oil and the production processes. From 1995 petrol with approx. 2 % of benzene has been sold in the eastern part of Denmark (east of the Great Belt). The total content of aromatic compounds is 40-45 %. The emission of benzene depends on the content in petrol, but the emission is also linked to the total content of aromatic compounds in petrol, because benzene may be formed during the combustion process. The benzene content is now below 1%.

7.1 Annual statistics

Statistics

A number of statistical parameters are shown in table 7.1 and 7.2. Compared with 1997 (Kemp, Palmgren and Manscher, 1998) the CO levels were in 1998 generally slightly lower and the benzene and toluene concentrations were reduced by around 25% for the average and median and 10-20 % for the higher percentiles. For benzene the risk of cancer can be estimated to about $3 \cdot 10^{-5}$ at an life time exposure to the average value. The concentration of toluene seems not to be critical compared to the WHO guideline value. A EU Directive for CO and benzene has been adopted (EC, 2000). The limit values are set to 10 mg/m^3 as the maximum daily 8 hour average based on

moving hourly measurements for CO and 5 µg/m^3 as the average over one calendar year for benzene. The value for CO shall be met in 2005, while 2010 is set for benzene. The 1999 values for CO were around half the expected new limit value (table 7.1). The measured yearly average in 1999 were close to the expected limit values; but the concentrations in 2010 will be well below the limit value if the present trend continues as expected.

Table 7.1 Statistical parameters based on hourly results for CO in 1999. All values are given in µg/m^3. *) The 8 hours values are calculated as a moving average based on hourly results.

Station	Average	Median (hour)	98-perc. (hour)	99.9-perc. (hour)	Max. 24 hour	Max. 8 hour *)	Max. 1 hour
Copenhagen/1257	1290	1050	3900	6420	3325	5080	10600
Odense/9155	830	595	2920	5070	2590	4090	7800
Aalborg/8151	1150	895	3690	5720	2960	4380	7500
Copenhagen/1259	39	275	825	1780	1070	1610	2470
WHO guideline value	-	-	-	-	-	10000	30000
EU limit value (proposal)	-	-	-	-	-	10000	-

7.2 Trends

The aromatic compounds show very high correlation with CO (Kemp, Palmgren and Manscher, 1998), which confirms that they were emitted from petrol powered vehicles. This was expected since the contribution of CO from diesel vehicles was relatively low at Copenhagen/1257 and the emission of aromatic VOCs from diesel powered vehicles was expected to be unimportant. The main emission of aromatic compounds from diesel vehicles is expected to be heavier hydrocarbons and PAHs. The correlation between CO, benzene, toluene, ethylbenzene, n-, o- and p-xylene were generally high indicating that they have been emitted from the same type of sources.

Table 7.2 Statistical parameters based on hourly results for benzene and Toluene in 1999 measured at Copenhagen/1257 and Odense/9155. All values are given in µg/m^3. The values from Odense 9155 are based on only 8 months results. The lifetime risk and guideline value are from WHO, 1997. The 7 days maximum is calculated as a moving average based on 24 hour averages.

Specie	Station	Average	Median (hour)	98-perc. (hour)	99.9-perc. (hour)	Max. 7 days	Lifetime risk at 1 µg/m^3	Guideline for 7 days average
Benzene	Copenhagen/1257	5.0	4.2	15	28	-	$4.4\text{-}7.5 \cdot 10^{-6}$	-
	Odense/9155	3.5	2.4	13	24			
Toluene	Copenhagen/1257	23	19	75	134	39	-	260
	Odense/9155	15	9.6	63	118	36		

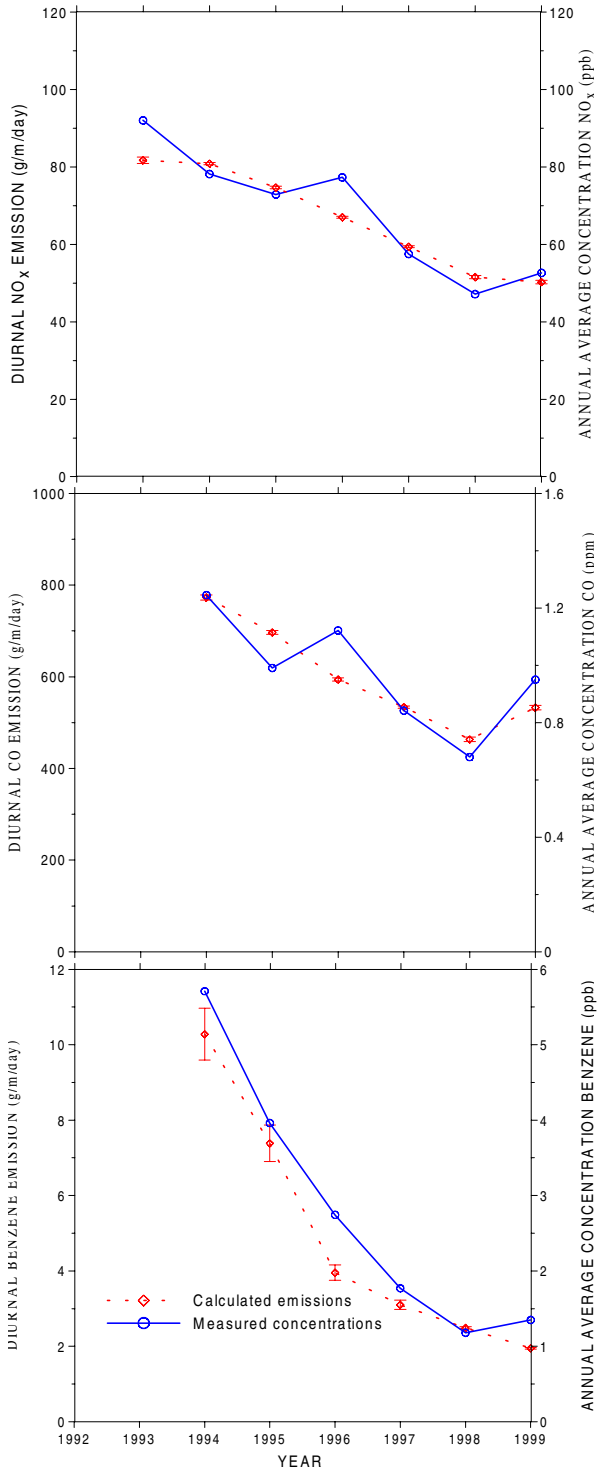


Figure 7.1a. Trends of annual averages of NOx, CO and benzene concentrations and calculated traffic emissions in Copenhagen (street canyon, Jagtvej). Urban background pollution has been subtracted from the measured concentrations.

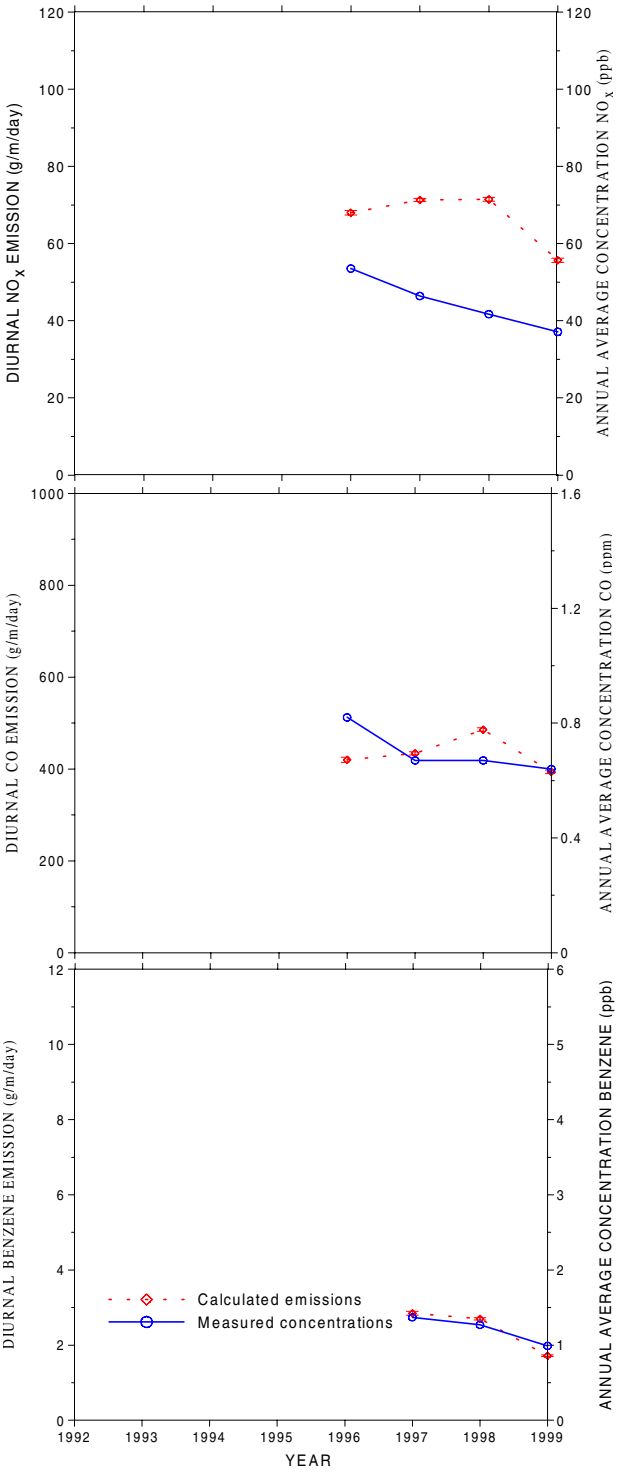


Figure 7.1b. Trends of annual averages of NOx, CO and benzene concentrations and calculated traffic emissions in Odense (street canyon, Albanigade). Urban background pollution has been subtracted from the measured concentrations.

7.3 Emission

The emissions and the meteorological conditions influence the trends of the air pollution concentrations. A technique to determine the

emission from road traffic has been developed at NERI (Palmgren et al. 1999) based on inverse model calculations by the OSPM model (Berkowicz et al., 1997). The technique has been applied on CO, benzene and NO_x data from Copenhagen/1257 and Odense/9155. In the figures 7.1a and 7.1b are shown the measured concentrations of NO_x, benzene and CO from 1994 to 1999. In addition, the calculated daily emissions are shown based on the inverse model calculation. Fluctuations in the concentrations of NO_x and CO were observed due to the meteorological conditions, e. g. the concentrations were higher in 1996 than in 1995. However, the emissions decreased continuously from 1994 to 1999 due to the increasing number of TWC cars and nearly constant traffic flow at Jagvej (Copenhagen/1257). For benzene more steep negative trends were observed for the concentration as well as for the emission. This is due to the reduced content of benzene in petrol from 1995 from approx. 3.5% to approx. 1% in 1998. This reduction dominated over the unfavourable dispersion conditions in 1996.

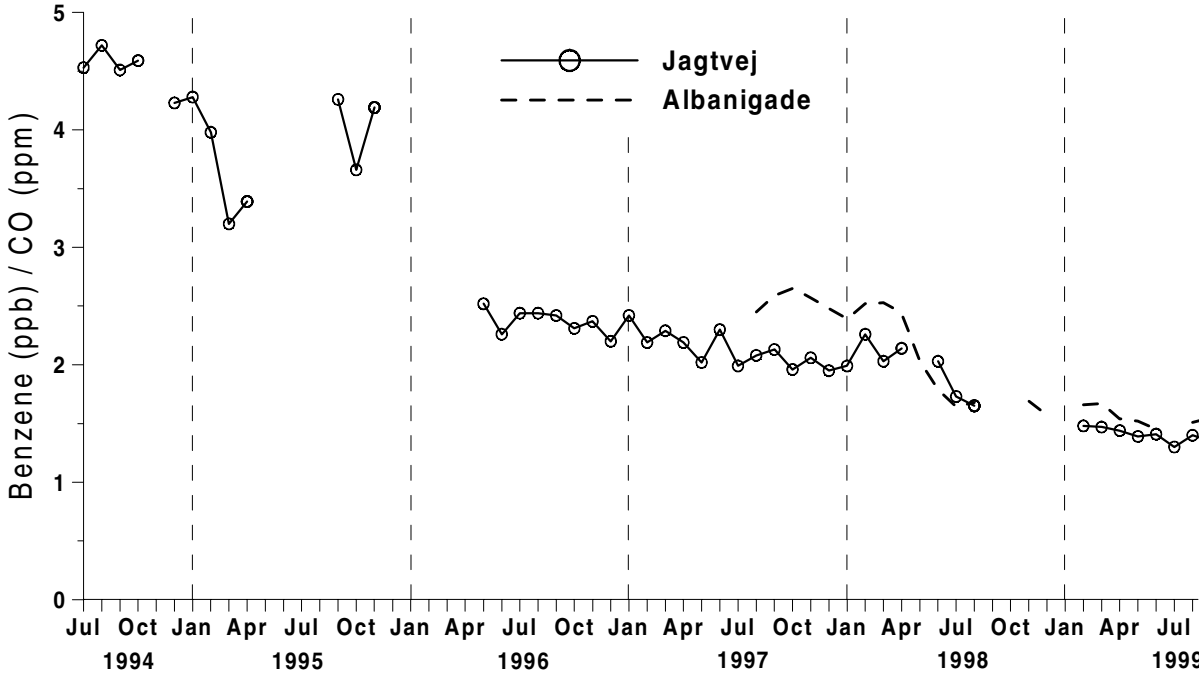


Figure 7.2 Monthly ratios of benzene/CO (ppb/ppm), determined as the slopes of the linear regression lines of measured hourly concentrations at Copenhagen/1257 (Jagtvej) and Odense/9155 (Albanigade). The steep decrease in this ratio in the periods 1995-1996 and during the summer 1998 coincides with the reduction of the benzene in petrol sold in Denmark.

7.4 Ratio Benzene/CO

The different trends in CO and benzene emissions can be investigated by analysing the trends in the ratio of benzene to CO air concentrations. This ratio doesn't depend on the meteorological conditions and is directly related to the emissions. The annual averages of the ratios between benzene and CO (ppb/ppm), estimated as the slopes of the linear regression lines are shown in

Figure 7.2. A clear reduction of the benzene-CO ratio was observed between 1995 and 1996 for Copenhagen/1257. This coincides with the reduction of the benzene content in petrol from 3.5% to 2% at the Statoil refinery, which delivers approx. 80% of the petrol in Copenhagen. Another sharp decrease was observed at both sites during 1998, when both Danish refineries, which produce approx. 80% of the petrol used in Denmark, reduced their content of benzene in petrol to 1%. In Copenhagen, the ratio between benzene (ppb) and CO (ppm) was 4.3 in 1994-1995. In the period 1996-1997, the ratio was 2.4. The reduction in this ratio corresponds to the relative reduction in the benzene content of approx. 40%. A similar analysis of data from a street in London (Marylebone Road, www.aeat.co.uk/netcen/archive/my1.html) shows a ratio between benzene and CO of approx. 1.4. This is significantly lower than derived from the Danish data, indicating different fuel qualities. The toluene-CO ratio did not change so drastically (Palmgren et al., 2000), which indicates that benzene was removed specifically. A small year-to-year decrease of the toluene-CO ratio in the period 1994-1999 can be explained by reduced evaporation losses from newer cars.

Benzene in petrol and air

The clear correspondence between the reductions in benzene content in petrol and the concentration in air shows that the benzene content in petrol is a crucial factor. However, the available data do not allow quantification the benzene emission due to non-benzene aromatics, but the study indicates increasing relative importance of this part of the benzene emission.

8 Conclusion

Nitrogen dioxide

The measured 98-percentiles for NO_2 were about half the limit value (200 µg/m^3), whereas medians were approximately close to the guide value (50 µg/m^3). The measured values were in 1999 slightly higher than in the previous years, which probably is a result of higher O_3 concentrations. A trend analysis covering the last 17 years shows, however, that the NO_2 level has been slightly decreasing, while a more substantial decrease is observed for NO since 1993 as a result of the increasing number of cars with three way catalytic converters. The measured concentrations are close to the new EU limit.

Ozone

A set of threshold values for O_3 has been implemented in Denmark in 1994. The values are based on an EEC directive for protection of human health and vegetation. During 1999 some of the threshold values were exceeded frequently. The reduction of O_3 concentrations can only be obtained in an international co-operation, because the precursors to a large extent are emitted in other countries and because O_3 in the lower troposphere is a secondary pollutant. The precursors are NO_x and organic gases which may be of either natural or anthropogenic origin. The time series from 1992 to 1997 seems to indicate a downward going trend at a rate of around 0.5 ppb per year for the yearly averages. But the observed concentrations 1999 were higher than in the previous years. The meteorological conditions can to some extent explain the difference.

Sulphur dioxide

The actual measured values at all stations were more than a factor of 10 lower than the limit values for SO_2. The measured values were more than a factor of 5 below the EEC guide values and the new EU limit values. There is a marked decrease of the SO_2 levels since 1982. The most pronounced reduction was observed in 1986 as a result of the decrease of the sulphur content in the fossil fuel products used in Denmark. But even in the recent years the concentrations have been decreasing due to better emission control on larger installations and the introduction of natural gas for i.e. domestic heating. The negotiation of international protocols for a further reduction of the SO_2 emission in the European countries will probably lead to a continuation of the downward going trend. The concentrations have been considerably lower since 1997. The decrease is mainly due to lower concentrations under winds from southeast. The results reflect probably lower emissions in Eastern Europe.

Particles (TSP and PM_{10})

The collected TSP is a mixture of "natural" wind blown dust (including the particles generated by the traffic) in form of coarse particles and anthropogenic derived fine particles. The measured concentrations were lower than half the limit values. A slightly decreasing trend has been observed for the last 10 years. This may be a result of the "winter crops" during the winter and of better control with the combustion processes. The decrease can thus be expected to continue in the future, due to the introduction of catalysts on all new gasoline driven cars, and as expected limitations on the particle

emission from diesel vehicles become more stringent. The TSP values were slightly lower in 1999 than in the previous years. PM_{10} measurements are performed at the street station Copenhagen/1257. The results from 1999 show good correlation with TSP at a ratio of 3:4 between PM_{10} and TSP. The average value is slightly below the new 2005 EU limit value, while the 50 $\mu g/m^3$ limit was exceeded more frequent than the "allowed" 35 times.

Ultra-fine particles

The ultra-fine particles (< 0.1 µm) attract special attention due to their possible negative health effects. Measurements performed at some of the LMP sites show that the number concentration if the ultra-fine particles is very high at street level; but that the mass concentration is only a minor part of the total PM_{10} concentration. The ultra-fine particles are found in the exhaust from gasoline and especially diesel driven vehicles.

Elements (heavy metals)

The main source for lead is no longer the local traffic, but industrial and traffic sources outside the country. The level is about 1% of the limit value. Most of the other heavy metals (e.g. Ni, As and Cd) have shown a decreasing trend since 1982. An exception is Cr and Cu, which have been increasing for the last 2-3 year at the traffic sites. The main source is probably particles from brake pad wear. The levels are relatively low compared to WHO guidelines. For the potential carcinogenic heavy metals Cr, Ni and As the lifetime risk is estimated to around 1:10[6].

CO and benzene

CO was found in concentrations below the WHO guideline values; while the average concentration of benzene was approx. 4 $\mu g/m^3$, which is below the proposed new EU limit value for annual average. Petrol cars are the dominating source for both compounds at street level. The concentration of benzene decreased significantly from 1995 to 1999 mainly due to reduced benzene content in petrol.

Smog warning

A smog warning system was implemented from April 1994. It is for SO_2 and NO_2 a continuation of the provisional system, which was started in 1987. The population is warned through the public broadcast stations, if the concentration of either SO_2 or NO_2 exceeds 350 $\mu g/m^3$ in more than three consecutive hours and an immediate improvement is not expected. No warnings have been issued since the start in 1987 and, taking the measured concentrations and the expected development into account, it can with almost certainty be excluded that the warning limits will be exceeded in Denmark. O_3 has been included in the new system: The population is informed immediately if the hourly average concentration exceeds 180 $\mu g/m^3$ and if the hourly concentration exceeds 360 $\mu g/m^3$ a warning is issued to the population. The information threshold was exceeded during one episode in 1999. The information threshold will statistically be exceeded once every second year. It is not realistic to assume that the warning threshold will be exceeded.

The results from the Danish Air Quality Program in 1999 showed that the concentrations found in the Danish cities were below the existing limit values, but the new EU limit values for NO_2 and PM_{10} may be exceeded. The O_3 threshold values, which were implemented in 1994, are however frequently exceeded. The NO_2 concentrations

were around half the limit value and a clear downward going trend was observed. The level of O_3 seems also to be slightly decreasing. The measurements in LMP III program aims i.a. at a description of the O_3-NO_x interaction in order to reveal the effect of actions already taken to reduce the NO_x emissions (obligatory three way catalysts on new gasoline driven cars and reduction of the emission from power plants) and the effect of future reductions, which hopefully will be realised through international protocols within the ECE-LRTAP convention and the coming EU acidification strategy for the emission of the O_3 precursors.

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Danish Summary - Dansk resumé

Det Landsdækkende Luftkvalitets Måleprogram
Årsrapport for 1999

Faglig rapport fra DMU nr. xxx

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LMP

Luftkvaliteten i de danske byer overvåges gennem Det Landsdækkende Luftkvalitets Måleprogram (LMP). LMP startede i 1982 og er ændret med henblik på belysning af de aktuelle forureningsproblemer ved revisioner i 1987 og 1992. I det nuværende program (LMP III) foretages målinger i København, Odense og Aalborg i et samarbejde mellem Danmarks Miljøundersøgelser (DMU), Miljøstyrelsen, Hovedstadsregionens Luftovervågningsenhed (HLU), Fyns Amt og Ålborg kommune. Det praktiske arbejde udføres af DMU sammen med Miljøkontrollen i København, og Erhvervs- og Miljøforvaltningen i Aalborg.

Måleprogram

I hver af de tre byer er opstillet et par af målestationer. En basisstation i gadeniveau på en trafikeret gade og en tagstation nogle hundrede meter fra basisstationen (Kemp, K. 1993). Kampagnemålingerne er fra 1997 blevet udvidet så de foregår kontinuert gennem hele året. På basisstationen foretages således kontinuert måling af NO, NO_2, SO_2, CO, svævestøv samt grundstofindholdet i svævestøvet. På tagstationerne måles NO, NO_2 og O_3 koncentrationen i "taghøjde" samt følgende meteorologiske parametre: vindretning, vindhastighed, relativ fugtighed, temperatur og global stråling. I København måles desuden CO på tagstationen og O_3 i gadeniveau. Benzen og toluen måles i gadeniveau i København og Odense. Baggrundsforureningen måles på en station ved Lille Valby ca. 25 km vest for København og på Keldsnor på sydspidsen af Langeland. Ud over den generelle overvågning af luftkvaliteten er et væsentlig formål med LMP III programmet at give mulighed for at beskrive vekselvirkningen mellem og dannelsen af NO_x og O_3.

Nye grænseværdier

EU er i gang med at indføre luftkvalitetsnormer, som skal gælde i alle medlemslandende. Det sker i form at et "rammedirektiv", som fastlægger de generelle retningslinier og strategier for vurdering af luftkvaliteten på grundlag af målinger på udvalgte steder kombineret med vurderinger af luftkvaliteten i større områder, bl.a. ved brug af modelberegninger. Rammedirektivet suppleres med en række "datterdirektiver", som fastlægger grænseværdier for enkelte stoffer. Rammedirektivet blev vedtaget i 1996 og det første datterdirektiv,

som omfatter SO_2, NO_2, NO_x ($NO+NO_2$), partikler og Pb, er vedtaget i 1999. Der er desuden direktiver under udarbejdelse for O_3, CO, benzen og tungmetallerne Cr, As og Ni.

Specialprogrammer

LMP målingerne giver kontinuerte flerårige måleserier, som er nødvendige for at vurdere de systematiske variationer og sammenhængen mellem forekomsten af forskellige forurenende stoffer i atmosfæren. Flere mere forskningsorienterede projekter udnytter den umådelige fond af viden, som LMP programmerne har bragt til veje. Disse projekter har bl.a. til formål at beskrive omdannelsen af kvælstofoxider, som udsendes fra trafikken, forekomst og kilder til PAH (polycykliske aromatiske hydrocarboner) som er kræftfremkaldende stoffer, og VOC (flygtige organiske hydrocarboner) af hvilke nogle er kræftfremkaldende og som i øvrigt er af stor betydning ved dannelsen af O_3 i atmosfæren. For tiden tiltrækker de ultrafine partikler (dvs partikler mindre end 0,1 µm i diameter) sig særlig interesse p.gr.a. deres mulige sundhedsskadelige effekter. DMU er involveret i flere projekter til belysning af dannelse og egenskaber af de ultrafine partikler.

Det følgende resumé er inddelt i afsnit efter kapitlerne i selve rapporten. Relevante tabeller og figurer kan findes i de pågældende kapitler.

Nitrogenoxider

For NO_2 (kapitel 2) er de målte koncentrationer i alle tilfælde under den gældende grænseværdi på 200 µg/m^3 for 98-percentilen og de vejledende værdier på 135 µg/m^3 for 98-percentilen og 50 µg/m^3 for medianen. Afstanden til de vejledende værdier er ikke ret stor. WHO's vejledende værdi på 40 µg/m^3 for årgennemsnittet er overskredet i København. Emissionen af NO_x fra benzindrevne bliver reduceret efterhånden som der kommer katalysatorer på flere og flere biler. Det er nu tydeligt at niveauet for NO er blevet reduceret siden begyndelsen af 1990'erne. Der kan nu også konstateres en svagt faldende tendens for NO_2; men reduktionen er ikke så stor som for NO fordi dannelsen af NO_2 i gaderum i væsentlig grad er begrænset af manglen på O_3. Emissionen fra de øvrige hovedkilder, dieselbiler og kraftværker, vil ikke blive ændret ret meget de første par år. Der er vedtaget et nyt EU Direktiv som bl.a. giver nye, væsentlig strengere, grænseværdier for NO_2 og NO_x ($NO+NO_2$). De nye værdier er fastlagt udfra en vurdering af den skadelige effekt af kvælstofoxider. Grænseværdierne for NO_2 er fastlagt på grundlag af en helbredsmæssig vurdering. Grænseværdierne, 40 µg/m^3 for årsgennemsnittet og 200 µg/m^3, som højst må overskrides 18 gange på et år, er gældende fra 1. januar 2010. Den kommende grænseværdien for årsgennemsnittet var i 1999 overskredet på Jagtvej i København. Til beskyttelse af plantevækst skal årsgennemsnittet for NO_x i landområder være under 30 µg(ækvivalent NO_2)/m^3 fra 19. juli 2001. De målte koncentrationer er under halvdelen af denne værdi.

Ozon

De målte O_3 værdier (kapitel 3) var som tidligere næsten ens over hele landet. Med gennemførelsen af et EU direktiv om O_3 er der i 1994 fastsat en række tærskelværdier i Danmark i forbindelse med beskyttelse af både plantevæksten og sundheden. Flere af disse

tærskelværdier blev overskredet i løbet af 1999. O_3 i den nedre del af atmosfæren dannes ved fotokemiske reaktioner. VOC og kvælstofoxider er af stor betydning for dannelse af O_3. Da en stor del af den VOC der findes i luften i Danmark stammer fra andre lande, kan en effektiv nedsættelse af O_3 forureningen kun ske gennem et internationalt samarbejde. De største koncentrationer findes i sommerhalvåret i perioder med varmt og solrigt vejr. Der er indført en tærskelværdi på 180 µg/m^3. Hvis timemiddelværdien overskrider denne værdi, skal befolkningen underrettes. Tærskelværdien på 180 µg/m^3 blev overskredet én gang i 1999. Målingerne på Lille Valby/2090 viser en svagt faldende tendens for O_3 niveauerne i perioden 1991-1998. I 1999 var koncentrationerne noget højere end i de nærmest foregående år.

Svovldioxid

Forureningen med SO_2 (kapitel 4) er klart faldende i Danmark. De målte koncentrationer var mere end en faktor 10 under grænseværdierne og mere end en faktor 5 under den i EU gældende vejledende værdi. De målte koncentrationer ligger også langt under de nye grænseværdier, som findes i det nye EU datterdirektiv. Det største fald skete omkring 1985-86, hvor svovlindholdet i fossile brændsler blev begrænset som følge af et lovindgreb, men bedre røgrensning, indførelse af naturgas og en fortsat reduktion af svovlindholdet i bl.a. olieprodukter har fortsat den positive udvikling i svovlforureningen. Der kan dog fortsat episodisk findes høje koncentrationer. De største værdier findes oftest under de såkaldte hot-spot episoder, hvor røgfanen fra en nærliggende industri eller kraftværk "slår ned" ved målestationen. Antallet af hot-spot episoder er gået ned i de seneste år. Der der et markant fald i 98 percentilen for SO_2 og gennemsnitskoncentratioen af partikulært svovl fra 1997 og de følgende år. Begge dele kan bruges som indikatorer for forurening transporteret over lange afstande. Det er ikke klart i hvor høj grad nedgangen skyldes mindre emissioner i Østeuropa, idet forskellige meteorologiske faktorer også kan have været afgørende.

Svævestøv

Den totale partikelkoncentration i luften, TSP (=Total Suspended Particulate matter) (kapitel 5) findes i byerne i koncentrationer på mellem 1/2 og 1/4 af grænseværdierne. TSP består af en blanding af bidrag fra flere kilder, hvoraf ophvirvlet jordstøv er den væsentligste. Der er generelt en svagt nedadgående tendens for TSP. Den kan til dels forklares ved bedre kontrol med partikeludslippet ved forbrændingsprocesser (især kraftværker og trafik); men også de "grønne marker" om vinteren, som er blevet mere og mere udbredt, synes at have haft en virkning. De nye grænseværdier, som er vedtaget af EU, er baseret på PM_{10} (dvs partikler med en aerodynamisk diameter mindre end 10 µm) i stedet for TSP. De målte TSP værdier er derfor ikke umiddelbart sammenlignelige med de nye grænseværdier. I 1999 er der foretaget PM_{10} målinger på Jagtvej i København. Disse målinger viser, at årsgennemsnittet er tæt på grænseværdien på 40 µg/m^3, som er gældende fra 2005; mens døgnmiddelværdier på over 50 µg/m^3 forekom hyppigere end de tilladte 35 gange pr. år. Det ventes at grænseværdierne vil blive revideret i løbet af nogle år, da PM_{10} næppe er den mest relevante partikelfraktion til vurdering af partiklernes sundhedsskadelige

virkning. Efter alt at dømme spiller de ultrafine partikler (0.01 – 1 µm) en væsentlig rolle for den sundhedsskadelige virkning af svævestøvet. Der er i 1999 foretaget målinger af ultrafine partikler på flere LMP stationer. I gadeniveau udgør de ultrafine partikler antalsmæssigt den overvejende del af den totale partikelmængde; men massen er kun en mindre del af den totale PM_{10} koncentration. De ultrafine partikler i gadeniveau stammer fra benzin- og især dieseldrevne køretøjer.

Tungmetaller

For de fleste tungmetaller har der været en nedadgående tendens siden 1982 (kapitel 6). Den største nedgang er målt for bly. I takt med at blyet er fjernet fra benzinen er blyforureningen i de danske byer faldet til samme niveau som flere andre tungmetaller. Cr og Cu udgør en undtagelse for den generelt faldende tendens, idet der for disse to stoffer er observeret stigende koncentrationer på gadestationerne gennem de sidste par år. Cr og Cu i gadeniveau stammer sandsynligvis især fra slid på motorkøretøjernes bremseklodser. Cr, Ni og As kan findes i forbindelser, som er carcinogene. På grundlag af WHO's vurderinger kan det skønnes, at de målte koncentrationer udgør en livstidsrisiko på omkring $1:10^6$ for udvikling af kræft.

Kulmonoxid, benzen og toluen

Der foretages målinger af CO på alle gadestationer samt på tagstationen i København. Der er målt flere organiske forbindelser, bl.a. benzen og toluen på gadestationerne i København og Odense (kapitel 7). CO-koncentrationerne var i 1999 under WHO's vejledende værdier. Benzen koncentrationerne er faldet markant siden 1994 som følge af katalysatorerne på benzinbilerne og en nedsættelse af benzenindholdet i benzin. Målingerne bekræfter, at såvel CO som benzen hovedsageligt stammer fra benzin biler.

Resumé

LMP resultaterne fra 1999 viser, at den forurening, der findes i de danske byer, var under de gældende grænseværdier, men tærskelværdierne for O_3 blev overskredet jævnligt. Selv om NO_2 koncentrationerne var under grænseværdien, var de dog ret tæt på. LMP III programmet har bl.a. til hensigt at give grundlag for en beskrivelse af O_3-NO_x vekselvirkningen i byatmosfæren, så virkningen af emissionsbegrænsende foranstaltninger (katalysatorer på nye biler og reduktion af udslippet fra kraftværker) kan dokumenters. På længere sigt vil konsekvenserne af internationale protokoller for reduktion af udslippet af udgangsstofferne for O_3 forhåbentlig også slå igennem.

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