



# THE DANISH AIR QUALITY MONITORING PROGRAMME

Annual Summary for 2015

Scientific Report from DCE – Danish Centre for Environment and Energy

No. 201

2016



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DCE – DANISH CENTRE FOR ENVIRONMENT AND ENERGY

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Thomas Ellermann  
Jesper Nygaard  
Jacob Klenø Nøjgaard  
Claus Nordstrøm  
Jørgen Brandt  
Jesper Christensen  
Matthias Ketzel  
Andreas Massling  
Steen Solvang Jensen

Aarhus University, Department of Environmental Science  
Updated version pr. 22 nov. 2016



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# Data sheet

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Abstract:	The air quality in Danish cities has been monitored continuously since 1981 within the Danish Air Quality Monitoring network. The aim is to follow the concentration levels of toxic pollutants in the urban atmosphere and to provide the necessary knowledge to assess the trends, to perform source apportionment, and to understand the governing processes that determine the level of air pollution in Denmark. In 2015 the air quality was measured in four Danish cities and at two background sites. In addition model calculations were carried out to supplement the measurements. At one street station (H.C. Andersens Boulevard) in Copenhagen NO <sub>2</sub> was found in concentrations above the EU limit value for the annual average, while NO <sub>2</sub> levels in Odense, Aarhus and Aalborg were below the limit value. Model calculations indicate exceedances of the NO <sub>2</sub> limit value at several streets in Copenhagen. Annual averages of PM <sub>10</sub> and PM <sub>2.5</sub> were below limit values at all stations and the average exposure indicator (PM <sub>2.5</sub> in urban background) has decreased with about 20% since 2010. The concentrations for most pollutants have been decreasing during the last decades.
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# Contents

<b>Summary and Conclusion</b>	<b>5</b>
<b>Danish summary - Dansk resumé</b>	<b>7</b>
<b>1 Introduction</b>	<b>9</b>
<b>2 Measurements and model calculations</b>	<b>11</b>
2.1 Measurements	11
2.2 Model calculations	14
<b>3 Nitrogen oxides</b>	<b>18</b>
3.1 Annual statistics	18
3.2 Trends	19
3.3 Results from model calculations	21
<b>4 Ozone</b>	<b>28</b>
4.1 Annual statistics	28
4.2 Trends	29
4.3 Results from model calculations	30
<b>5 Carbon monoxide</b>	<b>33</b>
5.1 Annual statistics	33
5.2 Trends	34
<b>6 Benzene and other Volatile Organic Compounds</b>	<b>35</b>
6.1 Annual statistics and trends	35
<b>7 Particles (TSP, PM<sub>10</sub>, PM<sub>2.5</sub> and particle number)</b>	<b>39</b>
7.1 Annual statistics	40
7.2 Trends	43
7.3 Impact of salt from winter salting and sea	46
<b>8 Heavy Metals</b>	<b>48</b>
8.1 Annual statistics	48
8.2 Trends	49
<b>9 Sulphur dioxide</b>	<b>51</b>
9.1 Annual statistics	51
9.2 Trends	52
<b>10 Polyaromatic Hydrocarbons (PAHs)</b>	<b>53</b>
10.1 Annual Statistics	53
10.2 Trends	55
<b>11 Organic carbon and elemental carbon</b>	<b>56</b>
11.1 Annual statistics and trends	56
<b>12 Chemical composition of PM<sub>2.5</sub></b>	<b>58</b>
<b>13 References</b>	<b>61</b>
<b>Appendix 1</b>	<b>64</b>
Pollutants measured in the network	64



## Summary and Conclusion

This report presents the result from the Danish Air Quality Monitoring Programme in 2015. The monitoring programme is carried out by the DCE - Danish Centre for Environment and Energy (DCE) at Aarhus University. The core part of this programme consists of continuous measurements at eleven monitoring stations; nine stations situated in the four largest cities, two stations located in background areas and a minor station in a suburban area. These measurements are supplemented with model calculations using DCE's air quality models.

The aim of the program is to monitor air pollutants relevant to human health in accordance with the EU air quality directives. The programme includes measurements of sulphur dioxide ( $\text{SO}_2$ ), nitrogen oxides ( $\text{NO}_x/\text{NO}_2$ ), particulate mass ( $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ ), particle number, benzene ( $\text{C}_6\text{H}_6$ ), toluene ( $\text{C}_7\text{H}_8$ ), carbon monoxide (CO), ozone ( $\text{O}_3$ ), polycyclic aromatic hydrocarbons (PAHs) and a number of heavy metals including lead (Pb), arsenic (As), cadmium (Cd), mercury (Hg) nickel (Ni), and a number of volatile organic compounds (VOCs) that are precursors for formation of ozone. The measurements and model calculations are used to evaluate the Danish air quality in relation to limit values as well as to follow trends. Further, the program serves as basis for determination of sources of the air pollutants, basis for evaluation of the impact of regulations of emissions and as basis for various research projects related to air quality.

The permitted number of exceedances in a year of the diurnal limit value of  $50 \mu\text{g}/\text{m}^3$  for  $\text{PM}_{10}$  was not exceeded at any station in the measuring network. Likewise, there were no exceedances of the annual limit values for  $\text{PM}_{10}$  ( $40 \mu\text{g}/\text{m}^3$ ) and  $\text{PM}_{2.5}$  ( $25 \mu\text{g}/\text{m}^3$  from 2015). The average exposure indicator (AEI) has decreased with 25% since 2010.

The number of particles in ambient air was about 14,000 particles per  $\text{cm}^3$  as annual average at the street station H.C. Andersens Boulevard. This is roughly a factor of 3 and 5 higher than in urban and rural background, respectively. A significant reduction in particle number has been observed since 2002.

The sodium content in  $\text{PM}_{10}$  on street stations was about  $1.6 \mu\text{g}/\text{m}^3$  corresponding to an estimated annual salt content ( $\text{NaCl}$ ) of about  $4.0 \mu\text{g}/\text{m}^3$ . High diurnal values of salt were observed during periods with winter salting of roads.

The annual limit value for  $\text{NO}_2$  ( $40 \mu\text{g}/\text{m}^3$ ) was exceeded at one street station in Copenhagen (H.C. Andersens Boulevard), whereas no exceedances were observed in Odense, Aalborg and Aarhus. The  $\text{NO}_2$  concentrations have decreased from 2014 to 2015 for most of the stations. At H.C. Andersens Boulevard (HCAB/1103) there were still elevated concentrations of  $\text{NO}_2$  compared to the situation before 2010 due to a permanent change in the traffic lanes at the street segment in front of the measurement station. Additionally, there has been a gradually decrease in the concentrations during the last years in parallel to the decrease observed at Jagtvej.

Model calculations at selected streets in Copenhagen and Aalborg indicate that the limit value was exceeded at 9 out of 98 calculated streets in Copenhagen but not at any streets in Aalborg in 2015. The number of street segments with model calculated exceedances has decreased to one third of the value in 2010.

The ozone levels in 2015 were on the same level as in 2014. No clear trend is observed for the average ozone concentration. The information threshold of 180 µg/m<sup>3</sup> was not exceeded in 2015. The target value for the maximum daily 8 hours mean ozone concentration of 120 µg/m<sup>3</sup> was not exceeded, but the long-term objective for this parameter was exceeded at all Danish stations. The long term objective has not entered into force.

Measurements of volatile organic compounds (VOCs) at the urban background in Copenhagen showed concentration levels between 0.01 µg/m<sup>3</sup> and 0.75 µg/m<sup>3</sup> for the selected 17 different compounds. VOCs can act as ozone precursors, and the aim of these measurements is to improve the general understanding of the ozone formation on a European level. The formation of ozone in Denmark is in general small due to moderate solar radiation. The ozone pollution in Denmark is to a large extent the result of long distance transport of pollutants from other European countries south of Denmark.

The levels of SO<sub>2</sub> and heavy metals have decreased for more than two decades and are now far below the limit values. The limit values for benzene and CO are not exceeded and the levels have decreased for the last decade.

Measurements of concentrations of particle bound PAH were performed at H.C. Andersens Boulevard, Copenhagen and at the suburban measurement station at Hvidovre. The average concentration of benzo[a]pyrene was 0.29 ng/m<sup>3</sup> and 0.25 ng/m<sup>3</sup> at H.C. Andersens Boulevard and Hvidovre, respectively. The target value for benzo[a] pyrene (1 ng/m<sup>3</sup>) was not exceeded in 2015.

Measurements of the chemical content in PM<sub>2.5</sub> showed that the annual average concentrations of NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> are very similar at the street station at H.C. Andersens Boulevard and at the rural station at Risø. The main difference between the two stations are for elemental carbon (EC), organic matter (OM) and Ca<sup>2+</sup> where the concentrations are higher at the street station compared to the rural background station. This is mainly due to emissions of these compounds from the traffic in Copenhagen.

Actual data, annual and multi-annual summaries are available at the website of DCE (<http://dce.au.dk/en/authorities/air/>), in Danish (<http://dce.au.dk/myndigheder/luft/>).

## Danish summary - Dansk resumé

Rapporten præsenterer resultater for 2015 fra Overvågningsprogrammet for luftkvalitet i danske byer. Programmet, som udføres af DCE - Nationalt Center for Miljø og Energi (DCE) ved Aarhus Universitet, er baseret på målinger ved ni målestationer placeret i de fire største danske byer samt ved to baggrundsmålestationer udenfor byerne og en mindre station i et forstadsområde. Disse måleresultater suppleres med resultater fra modelberegninger udført med DCE's luftkvalitetsmodeller.

Formålet med programmet er at overvåge luftforurening af betydning for sundhed i overensstemmelse med EU's luftkvalitetsdirektiver. I henhold til disse og øvrige danske behov måles koncentrationer af svovldioxid ( $\text{SO}_2$ ), nitrogenoxider ( $\text{NO}_x/\text{NO}_2$ ), partikelmasse ( $\text{PM}_{10}$  og  $\text{PM}_{2.5}$ ), partikel antal, benzen ( $\text{C}_6\text{H}_6$ ) og toluen ( $\text{C}_7\text{H}_8$ ), carbonmonoxid (CO), ozon ( $\text{O}_3$ ), udvalgte tungmetaller (fx bly (Pb), arsen (As), cadmium (Cd), kviksølv (Hg), nikkel (Ni)) og polyaromatiske kulbrinter (PAH'er) samt udvalgte flygtige kulbrinter (VOC'er), der kan føre til dannelse af ozon. Målingerne og modelberegningerne anvendes til at vurdere om EU's grænseværdier for luftkvalitet er overholdt. Rapporten beskriver endvidere udviklingen i koncentrationerne. Samtidigt tjener måleprogrammets resultater som grundlag for vurdering af kilderne til luftforureningen, vurdering af effekt af reduktionstiltag og som grundlag for en række videnskabelige undersøgelser, fx vurdering af små partiklers effekt på sundheden.

Der er fastsat grænse- og målværdier for flere af de målte stoffer. Grænseværdierne skal være overholdt fra 2005, 2010 eller 2015 alt efter, hvilke stoffer det drejer sig om. En detaljeret beskrivelse af gældende mål- og grænseværdier og deres gennemførelse findes i en bekendtgørelse fra Miljøministeriet (Miljøministeriet 2010). Bekendtgørelsen er baseret på det 4. datterdirektiv om tungmetaller og PAH'er (EC 2005) samt EU's luftkvalitetsdirektiv fra 2008 (EC 2008). En af de væsentligste ændringer i direktivet fra 2008 i forhold til de tre første datterdirektiver (1999, 2000 og 2002) er, at der stilles krav om målinger af de fine partikler ( $\text{PM}_{2.5}$ ), og at der er indført en grænseværdi for  $\text{PM}_{2.5}$ , som skal være overholdt i 2015.

De væsentligste konklusioner fra overvågningsprogrammet i 2015 er følgende:

- I 2015 blev grænseværdien for  $\text{NO}_2$  som årsmiddelværdi overskredet på én (H.C. Andersens Boulevard) af de to gademålestationer i København. I Odense, Aarhus og Aalborg var der ingen overskridelser. Koncentrationerne af  $\text{NO}_2$  i 2015 var for hovedparten af målestationerne stort set på niveau med koncentrationerne målt i 2014. På H. C. Andersens Boulevard blev der i 2010 indført en permanent ændring af vejbanerne ud for målestasjonen, hvilket førte til en forøgelse i koncentrationerne på omkring  $8 \mu\text{g}/\text{m}^3$  set i forhold til tidligere. Denne forøgelse i koncentrationerne ses fortsat om end koncentrationerne på H.C. Andersens Boulevard i gennem de seneste år er faldet parallelt med det generelle fald i koncentrationerne f.eks. som observeret på Jagtvej.
- Modelberegninger indikerer, at grænseværdien i 2015 var overskredet på 9 ud af 98 beregnede gadestrækninger i København, men ikke på udvalgte gadestrækninger i Aalborg. Siden 2010 er antallet af gadestræk-

ninger med beregnede overskridelser af grænseværdien blevet reduceret til en tredjedel.

- Der var ingen målestationer i måleprogrammet, hvor det tilladte antal af overskridelser af den daglige middelværdi for PM<sub>10</sub> (50 µg/m<sup>3</sup> må ikke overskrides mere end 35 gange årligt) blev overskredet.
- Indholdet af partikler mindre end 2,5 µm (PM<sub>2,5</sub>) overholdt ikke grænseværdien på 25 µg/m<sup>3</sup> som årsmiddelværdi, der skal være overholdt fra 2015. AEI-værdien (average exposure indikator) er faldet med omkring 25% siden 2010.
- Antallet af partikler mellem 6 og 700 nm var omkring 12.300 partikler per cm<sup>3</sup> på gademålestasjonen H.C. Andersens Boulevard, mens det var betydeligt mindre i by- og landbaggrund samt i forstadskvarter. Antallet af partikler på H. C. Andersens Boulevard faldt ca. 10% fra 2014 til 2015 og følger dermed det overordnede billede, hvor antallet af partikler er faldet med ca. 50% siden 2002.
- Indholdet af natrium i PM<sub>10</sub> på gademålestationerne var omkring 1,5 µg/m<sup>3</sup> svarende til et estimeret saltindhold (NaCl) på omkring 3,7 µg/m<sup>3</sup>.
- Ozonkoncentrationerne i 2015 var på niveau med 2014. Der er ikke fastsat egentlige grænseværdier for ozon (O<sub>3</sub>), men kun "målværdier" og "langsigtede mål" (hensigtsværdier). Der var i 2015 ingen overskridelser af målværdierne for beskyttelse af sundhed, mens de langsigtede mål (120 µg/m<sup>3</sup>) blev overskredet på alle bybaggrunds- og landstationerne. Den langsigtede målsætning er ikke trådt i kraft endnu. Tærsklen for information af befolkningen om høje ozonniveauer (timemiddel 180 µg/m<sup>3</sup>) blev heller ikke overskredet i 2015.
- De øvrige målte stoffer findes i koncentrationer under grænseværdierne, og for flere stoffer (fx svovldioxid og bly) er koncentrationerne faldet meget markant siden målingernes start i 1981.
- Målinger af partikelbundet PAH blev fortaget på H.C. Andersens Boulevard i København. Middelværdien for benz[a]pyren var 0,29 ng/m<sup>3</sup> og 0,25 ng/m<sup>3</sup> på henholdsvis H.C. Andersens Boulevard og ved målestasjonen i Hvidovre. Målværdien på 1 ng/m<sup>3</sup> var således ikke overskredet i 2015.
- Målinger af 17 udvalgte flygtige organiske kulbrinter (VOC'er) i bybaggrund i København viser koncentrationsniveauer, som spænder fra 0,01 µg/m<sup>3</sup> til 0,75 µg/m<sup>3</sup> i 2015. Disse VOC'er bidrager til den kemiske dannelse af ozon på europæisk plan, og målingerne skal først og fremmest understøtte den generelle forståelse af ozondannelsen i Europa. I Danmark skyldes størstedelen af ozon langtransport af luftforurening fra centrale og sydlige dele af Europa.
- Målinger af det kemiske indhold i PM<sub>2,5</sub> ved gademålestasjonen ved H. C. Andersens Boulevard og ved landbaggrundsmålestationen på Risø viser ligesom i 2011-2014, at de årlige gennemsnitskoncentrationer for NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup> og SO<sub>4</sub><sup>2-</sup> er stort set ens på de to stationer. Dette skyldes, at stofferne for en stor del stammer fra partikler transporteret til målestationer langvejs fra. De væsentligste forskelle mellem de to målestationer ses for elementært carbon (EC), organiske forbindelser (OM) og Ca<sup>2+</sup>, hvor koncentrationerne er højere på gadestationen som følge af uddelninger relateret til trafikken i København.

# 1 Introduction

The Danish Air Quality Monitoring Program (LMP) originates back to 1981. Today the programme is part of the National Monitoring Programme for the aquatic and terrestrial environment (NOVANA). The program consists of an urban monitoring network with stations in the four largest Danish cities and two background stations in rural areas (figure 2.1) which is supplemented by model calculations. The results are used for assessment of the air pollution in Denmark with special focus on Danish urban areas. The programme is carried out in co-operation between the DCE - Danish Centre for Environment and Energy (DCE), the Danish Environmental Protection Agency, and the Municipalities of Copenhagen, Aarhus, Aalborg and Odense. DCE is responsible for operating and maintaining the programme. Statistical parameters and actual data are accessible at the website: <http://dce.au.dk/en/authorities/air/>, (in Danish <http://dce.au.dk/myndigheder/luft/>). Selected near real-time data are also available at tele-text, Danish National Television. In addition, this report presents results from model calculations of air quality in Denmark carried out as supplement to the measurements.

The monitoring programme is carried out in accordance with the Danish Statutory Order No. 851 of 30 June 2010 from the Ministry of Environment (Miljøministeriet 2010) that implements the EU directives on air quality in Denmark (EC, 2005; EC, 2008).

One of the main objectives for the monitoring programme is to assess the air quality in relation to various air quality criteria (i.e. limit values, margin of tolerance, target values, long term objectives and alert thresholds) of which the limit values are the legally most important. The Danish air quality criteria are identical to those laid down in the EU directives described above.

The program was last revised in 2010 and the main changes due to this revision have been described in a previous report (Ellermann et al., 2013). Since 2012 there has only been minor changes were the most important are:

- Starting in August 2012 low volume samplers (LVS) for gravimetric determination of particle mass based on the reference method were introduced into the regular measuring programme and gradually installed at the PM-stations in the network to replace some of the older SM200 instruments that needed to be renewed. See introduction to Chapter 7 for an overview.
- A non-permanent measurement station at a suburban area in Hvidovre was started in the beginning of 2013 with measurements of PAHs in relation to use of wood burning as house hold warming. In June 2015 the measurement program in Hvidovre was supplemented with measurements of PM<sub>2.5</sub> by LVS, particle number and nitrogenoxides (NO and NO<sub>2</sub>).
- The urban background measurement station in Aarhus was in January 2015 moved to another position (Chapter 2.1).
- The street station in Aalborg had to be temporarily closed down from September 2014 and onwards due to nearby construction work (Chapter 2.1).

- At the street station in Albanigade in Odense there was a large decrease in daily traffic intensity starting from late June 2014 and the street was closed down for traffic in spring 2015. This change was due to major changes in the traffic patterns in Odense (section 2.1).

In the following chapters the results from measurements and model calculations for 2015 are presented and compared to limit and threshold values. Please refer to the EU Directives (EC, 2005; EC, 2008) for a detailed description of the exact definitions of the limit values, margin of tolerance, target values and alert thresholds.

## 2 Measurements and model calculations

### 2.1 Measurements

The core of measurement stations in the Danish air quality monitoring network originates back to the 1980s and the stations have therefore been positioned before the development of the EU directives on air quality. Despite this, the network still gives a comprehensive fulfilment of the requirements laid down in the directives.

Originally, the Danish measuring strategy was in short to place one or more pairs of stations in each of the four largest Danish cities. In each city one of the stations is located close to a street lane with a high traffic density. The other is located within a few hundred meters from the street station, and is placed so that it is representative for the urban background pollution; meaning that it is placed so that it is not influenced by pollutants from a single or a few streets or other nearby sources. In most cases the background stations are placed on rooftops. The short distance between street station and urban background station makes it possible to determine directly the traffic contribution as the difference between the two stations. In addition, two rural stations measure the pollution outside city areas. Further information about the program and results is found at the website: <http://dce.au.dk/en-authorities/air/>, in Danish (<http://dce.au.dk/myndigheder/luft/>). Although this strategy is still valid, it has been necessary to relax the criteria on the distance between the street station and urban background station, since it has not been possible to fulfil these criteria when new sites had to be found for example in Aarhus.

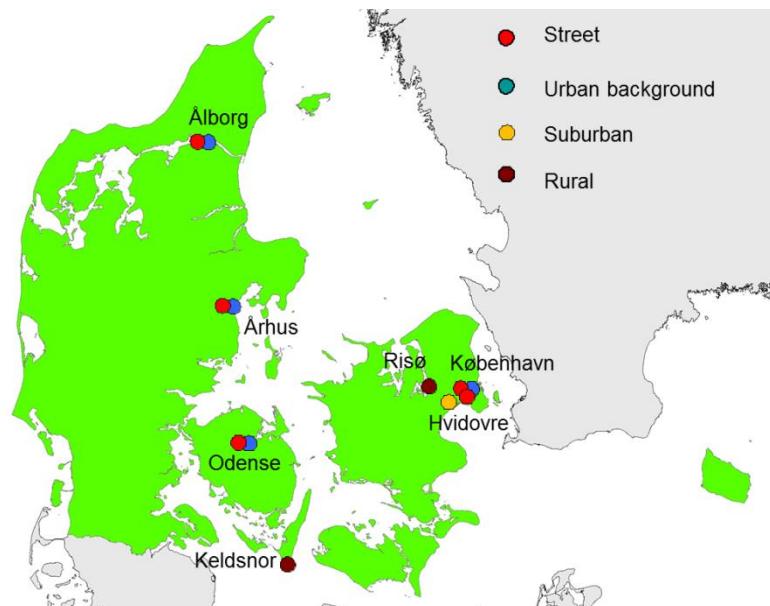


Figure 2.1. Main stations used for monitoring of air quality in relation to health.

**Table 2.1.** Main stations used for monitoring of air quality in relation to health

Location	Station type	Station number
<b>Copenhagen</b>		
H.C. Andersens Boulevard (HCAB)	Street	1103
Jagtvej	Street	1257
H.C. Ørsted Institute (HCØ)	Urban background	1259
Hvidovre, Fjeldstedvej 2650	Suburban	2650
<b>Odense</b>		
Albanigade	Street	9155
Town hall in Odense	Urban background	9159
<b>Aarhus</b>		
Banegårdsgade	Street	6153
Botanical Garden	Urban Background	6160
<b>Aalborg</b>		
Vesterbro, Limfjordsbroen	Street	8151
Aalborg/8158	Urban background	8150
<b>Rural</b>		
Lille Valby/Risø*	Rural background	2090I
Keldsnor/	Rural background	9055

\*The rural station at Lille Valby was in the middle of 2010 moved about 2 km west to Risø and is now situated close to DCE

In 2014-2015 there were three major changes regarding the stations:

- The measurement station on Vesterbro at Limfjordsbroen in Aalborg was closed down temporarily on September 8<sup>th</sup> 2014 due to a major construction work at the nearby house. The results for 2014 represent therefore only data for 250 days (70%) in 2014 and the station was closed down temporarily in 2015.
- In Odense a traffic plan has been adopted by the municipality for the entire city centre and the implementation of this plan began in late June 2014. This resulted in a major decrease in the traffic intensity at Albanigade, where the street station is situated. In spring 2015 Albanigade was closed for traffic. The station was shut down on 16 June 2015 and has been moved to a new position in summer 2016.
- In January 2014 the urban background station in Aarhus was moved to a new site since the municipality sold the house that the measurements station was placed upon. The new site is situated in the south easterly part of the Botanical Garden that belongs to Aarhus University (figure 2.2).



**Figur 2.2.** The new urban background measurement station at the south east end of the Botanical Garden in Aarhus. The map shows the position of the old site (red dot) and new site (blue dot). The distance between the two sites is about 900 m.

The following compounds were measured in 2015:

- Nitrogen oxides (NO, NO<sub>2</sub> and NO<sub>x</sub> (= NO + NO<sub>2</sub>)) were measured at all stations.
- Particle mass (PM<sub>10</sub> and/or PM<sub>2.5</sub>) were measured as 24 hour averages at all stations. At the following stations PM was measured throughout the year using low volume samplers (LVS) for gravimetric determination of particle mass based on the reference method (EN 12341: 2014): HCAB (PM<sub>10</sub> and PM<sub>2.5</sub>); HCØ (PM<sub>10</sub> and PM<sub>2.5</sub>); Jagtvej (PM<sub>10</sub> and PM<sub>2.5</sub>); Risø (PM<sub>10</sub> and PM<sub>2.5</sub>); Aarhus/street (PM<sub>10</sub> and PM<sub>2.5</sub>); Aarhus/urban background (PM<sub>2.5</sub>); Aalborg/ urban background (PM<sub>2.5</sub>). At Aalborg/street (PM<sub>2.5</sub>) no data were measured in 2015 due to relocation of the measuring station and problems finding a new site. At the remaining two stations (Keldsnor (PM<sub>10</sub>) and Odense/street (PM<sub>10</sub>)) PM was determined solely by using SM200 β-gauges measurements
- Elements (heavy metals) in PM<sub>10</sub> were measured at Copenhagen/street (HCAB), Copenhagen/urban background, Aarhus/street, Odense/street and the rural site Risø.
- Additionally, PM<sub>10</sub> and PM<sub>2.5</sub> were measured at both Copenhagen/street (HCAB) and Risø by means of TEOM that measures on a half hourly basis making it possible to resolve the diurnal variation. Part of these measurements was carried out in a research project funded separately by the Danish EPA.
- Particle number was measured at Copenhagen/street (HCAB), Copenhagen/urban background and Risø in cooperation with a particle research project funded separately by the Danish EPA. Additionally, measurements were started at a suburban site in Hvidovre in autumn 2015.
- Ozone (O<sub>3</sub>) was measured at all urban background and rural stations, and at the street stations Copenhagen/street (HCAB).
- Carbon monoxide (CO) was measured at all street stations except Jagtvej as well as at the urban background station, Copenhagen/urban background and the rural site Risø.
- Benzene and Toluene were measured at Copenhagen/street (HCAB) and Copenhagen/urban background using passive sampling on a weekly basis.
- PAHs were measured at Copenhagen/street (HCAB) and at the suburban site in Hvidovre.
- SO<sub>2</sub> was measured at Copenhagen/street (HCAB). The main purpose was to monitor episodic high concentrations.
- Elemental carbon (EC) and organic carbon (OC) were measured at Copenhagen/street (HCAB) and the rural site Risø.
- The meteorological parameters – air temperature, wind speed and direction, relative humidity and global radiation - were measured in Copenhagen, Odense, Aarhus and Aalborg at the urban background stations or at a location, which is representative for the meteorology at the urban background station.

The pollutants are described in more detail in Appendix 1.

Measurements of gasses (NO, NO<sub>x</sub>, NO<sub>2</sub>, O<sub>3</sub>, CO, SO<sub>2</sub>) and particle number were recorded as ½-hour averages. Particle mass (PM<sub>10</sub> and PM<sub>2.5</sub>) were measured both as 24 hour averages using beta measurements and low volume sampling (gravimetric method) and at ½-hour averages using TEOM (only part of particle mass). Elements in the particles as well as PAH were measured as 24 hour averages. EC and OC were measured as 24 hour averages. Benzene and toluene were measured weekly by passive sampling. Furthermore, volatile organic compounds were sampled as 24 hour averages.

## 2.2 Model calculations

In the monitoring programme the measurements at the permanent measuring stations are supplemented with model calculations using the THOR modelling system. In the present report model results are presented for NO<sub>2</sub> in streets and for ozone at a national level.

The THOR system is an integrated model system, capable of performing model calculations at regional scale to urban background scale and further down to individual street canyons in cities – on both sides of the streets. The system is driven by global meteorological analysed data from National Centres for Environmental Prediction, United States, which is used as input to the meteorological model MM5v7 (Grell et al., 1995).

The meteorological data for 2015 from MM5v7 is subsequently used to drive the air pollution models, including the Danish Eulerian Hemispheric Model, DEHM (Christensen, 1997; Brandt et al., 2012), the Urban Background Model, UBM (Berkowicz, 2000b; Brandt et al., 2001) and the Operational Street Pollution Model, OSPM® (Berkowicz 2000a; Ketzel et al., 2012). DEHM is providing air pollution input data for UBM which again is providing air pollution input data to OSPM. Further details about the integrated THOR system can be found in Brandt et al. (2000; 2001 and 2003 or under <http://www.au.dk/thor>).

Model calculations of air quality on national scale is carried out using DEHM (version 5.0), which is an Eulerian model where emissions, atmospheric transport, chemical reactions, and dry and wet depositions of air pollutants are calculated in a 3D grid covering the northern hemisphere with a resolution of 150 km x 150 km. The model includes a two-way nesting capability, which makes it possible to obtain higher resolution over limited areas. Three nested domains are used in the model runs under NOVANA, where the first domain is covering Europe with a resolution of 50 km x 50 km. The second domain is covering Northern Europe with a resolution of 16.7 km x 16.7 km. The calculations of air quality in Denmark are carried out in a third domain with a horizontal resolution of 5.6 km x 5.6 km. In the vertical direction the model is divided into 29 layers covering the lowest 15 km of the atmosphere. Of these the lowest layers are relatively thin (20 m) while the upper layers are relatively thick (2000 m). The model includes a comprehensive chemical scheme designed for calculation of the chemical reactions in the lower part of the atmosphere. The emission inventories used in DEHM have a geographical resolution of 1 km x 1 km for Denmark transformed into the 5.6 km x 5.6 km resolution domain and 16.7 km x 16.7 km for the remaining part of Europe. The emissions are based on Danish national emission inventories for the year 2014 compiled by DCE (<http://envs.au.dk/en/knowledge/air/emissions/>) and international

emission inventories for the year 2013 collected and distributed by EMEP ([www.emep.int](http://www.emep.int)).

The Urban Background Model, UBM, calculates the urban background air pollution based on emission inventories with a spatial resolution of 1 km x 1 km and based on input data from DEHM concerning the regional background. UBM is suitable for calculations of urban or rural background concentrations on high resolution (1 km x 1 km). The model includes a Gaussian plume approximation for calculation of the dispersion and transport of the air pollutants to every receptor point and a simple chemical model accounting for the photochemical reactions of NO<sub>x</sub> and ozone. The basic principles of the model are described in Berkowicz (2000b). In the recent years UBM has undergone many improvements in the formulation of physical processes and now treats both area and point sources in a more physically correct manner compared to earlier versions of the model. This has improved the overall performance of the model in comparison with measurements, and provides a more realistic spatial distribution of concentrations around large point sources. The emissions used in the UBM model are based on the SPREAD model that spatially distributes national emissions from 2014 from all sectors on a 1 km x 1 km grid for Denmark (Plejdrup & Gyldenkærne 2011). UBM has been calibrated against 2015 measurements at all four urban background stations in order to ensure good correspondence between measured and modelled NO<sub>2</sub>.

Finally, the street canyon model OSPM® ([www.au.dk/ospm](http://www.au.dk/ospm)) is used to calculate the air pollution at 2 m height at the sidewalks of selected streets. Meteorological data from the meteorological model MM5v7 and air pollution concentrations from UBM are used as input to the model. The model includes emissions from traffic, simple chemical reactions describing the reactions of air pollutants in the street canyons and the dispersion of the air pollution in the street canyon (due to meteorological conditions, turbulence induced by traffic and influence of the street geometry).

The input data for the OSPM on traffic data and street configurations for the selected urban streets are generated using the AirGIS system based on a GIS road network, GIS foot-prints of buildings and GIS calculation points (Jensen et al., 2001; 2009  
<http://envs.au.dk/videnudveksling/luft/model/airgis/>).

The model calculations for 2015 for Copenhagen and Aalborg have been carried out using the full model calculation system based on the THOR system, including MM5v7, DEHM, UBM, and OSPM. The calculations were carried out in order to determine the NO<sub>2</sub> concentration in 98 streets in Copenhagen and 31 streets in Aalborg.

### **2.2.1 Improved input data and re-calibration of OSPM**

In the assessment for 2013 the model calculations with OSPM were improved through major revisions. These included changes related to the general building height, revision of NO<sub>x</sub> emission factors for Euro 5 and 6 for passenger cars, and use of new travel speeds for the traffic based on GPS data (SpeedMap, speedmap.dk/portal/) and subsequent recalibration. Appendix 2 in Ellermann et al. (2014) describes the changes and presents documentation for the impact of the improved input data for the model calculations.

The model setup for the assessment for 2015 is similar to that of 2013 and also 2014. OSPM has been calibrated against 2015 measurements at the street stations of Jagtvej and Aarhus in order to ensure good correspondence between measured and modelled NO<sub>2</sub>. The street station of H.C. Andersens Boulevard has not been used in the calibration due to the about 8 µg/m<sup>3</sup> jump in concentrations since a change in street layout moved traffic closer to the station in 2010. It has been decided to move the station during 2016 to compensate for the change in street layout.

The traffic data used as input for the calculations with OSPM is updated annually for average daily traffic and vehicle distribution for the selected streets based on information obtained from the municipalities of Copenhagen and Aalborg. Traffic data are estimated at the location of the calculation points. For Copenhagen traffic data is based on manual counts performed annually or in 5-year intervals. Aalborg does not have a systematic traffic counting program similar to Copenhagen, and traffic data is based on available traffic data from manual and automatic counts together with data from a traffic model. Based on information from Copenhagen and Aalborg municipalities the Average Daily Traffic (ADT) and vehicle distribution on all streets have been updated with the most recent available traffic data. The vehicle distribution includes passenger cars, vans, trucks<32t, trucks>32t, and buses. In Copenhagen 33 out of the 98 calculation points had updated traffic data for 2015. For Aalborg 8 out of 31 streets had updated traffic data.

Manual traffic counts are carried out annually for the street segments in front of the measuring stations of H.C. Andersens Boulevard and Jagtvej in Copenhagen and Banegårdsgade in Aarhus. Manual counts for the 2015 assessment originate from September in Copenhagen and April in Aarhus. In Odense the street (Albanigade) with the measuring station was closed in May due to construction work and traffic has in recent years decreased considerably due to major changes in the overall traffic plan for Odense City. In Aalborg (Vesterbro) the measuring station was not in operation during 2015 due to nearby building construction work.

Calculations with the full model chain of DEHM-UBM-OSPM have been compared to measured NO<sub>2</sub> concentrations in 2015 for the fixed street measuring stations in Copenhagen and Aarhus. The model system slightly underestimates annual NO<sub>2</sub> concentrations with 1% for Jagtvej (Copenhagen) and slightly overestimates by 1% for Banegårdsgade (Aarhus). The model underestimates by 6% for H.C. Andersens Boulevard (Copenhagen) when comparing to measurements, and overestimates by 12% when taking into account the jump of about 8 µg/m<sup>3</sup> due to a change in street layout that has moved traffic closer to the measuring station. Calculations with the coupled DEHM-UBM models have also been compared to the fixed urban background measuring stations in Copenhagen, Aarhus, Odense and Aalborg. Here the model system predicts observations within -8% to 4%. DEHM overestimates observations by 3-41%.

The comparison of the modelled NO<sub>2</sub> concentrations presented in this report for 2015 with measurements at the 3 street locations shows a good overall agreement within -6 to 1% (table 2.2).

**Table 2.2.** Comparison of modelled and measured annual means of NO<sub>2</sub> concentrations in 2015

Unit: µg/m <sup>3</sup>	Measurements	Model results	Difference %	Models used
<b>Street:</b>				
Copenhagen/HCAB/1103	49/41*	46	-6%/12%*	DEHM/UBM/OSPM
Copenhagen/Jagtvej/1257	33	32	-1%	DEHM/UBM/OSPM
Aarhus/6153	31	31	1%	DEHM/UBM/OSPM
<b>Urban Background:</b>				
Copenhagen/1259	16	16	0%	DEHM/UBM
Aarhus/6159	13	13	0%	DEHM/UBM
Odense/9159	11	12	4%	DEHM/UBM
Aalborg/8159	12	11	-8%	DEHM/UBM
<b>Rural:</b>				
Risø/2090	7.6	11	41%	DEHM/UBM
Keldsnor/9055	7.4	8.4	13%	DEHM/UBM
Anholt/6001	4.4	5.6	27%	DEHM/UBM
Ulborg/7005	3.9	4.0	3%	DEHM/UBM

\* 49 µg/m<sup>3</sup> is measured at the measuring station at HCAB, but because of a change in street layout traffic has moved closer to the measuring station. Based on parallel measurements this rearrangement is estimated to have led to a jump of about 8 µg/m<sup>3</sup>. Without the change in street layout, about 41 µg/m<sup>3</sup> is expected. OSPM calculations are more representative of the measurements without the jump as OSPM calculations reflect concentration levels in front of the building facade.

## 2.2.2 Further development of the model

The detailed investigation on the impact of the changes in road lanes layout on HCAB (Ellermann et al., 2014) showed that the current OSPM version has some shortcomings in reproducing measurements at the location/re-location of the measuring station. The setting at HCAB is complex with multiple road lanes, inhomogeneous distribution of emissions and a measurement point right next to the nearest road lane, and at the same time a relatively long distance to the façade of the buildings. There is a need for further development of OSPM in order to describe the complex distribution of emissions in streets, definition of location of calculation point etc. A recently finished PhD study (Ottosen et al., 2015) has implemented the capability to handle inhomogeneous distribution of emissions in OSPM describing emissions according to each lane of the street layout. These improvements together with future supporting developments are expected to improve model calculations for complex street layouts, e.g. for HCAB, and will result in a new version of the model in years to come.

### 3 Nitrogen oxides

The nitrogen oxides (NO, NO<sub>2</sub>, NO<sub>x</sub>) are measured at eleven monitoring sites using gas monitors based on chemiluminescence. The concentrations are measured continuously throughout the year with a time resolution on minute scale that is aggregated to hourly averages for this reporting.

#### 3.1 Annual statistics

The annual statistics for 2015 for nitrogen dioxide and nitrogen oxides are shown in table 3.1 and 3.2. There was only exceedance of the annual limit value for NO<sub>2</sub> of 40 µg/m<sup>3</sup> (EC, 2008) at H.C. Andersens Boulevard (Copenhagen/1103). There were no exceedances of the hourly limit value for NO<sub>2</sub> of 200 µg/m<sup>3</sup>. This value must not be exceeded more than 18 times in a calendar year (see 19th highest hourly concentration in table 3.1). In 2015 there was no information to the public triggered by exceedance of the information threshold for NO<sub>2</sub> (three hours average must not exceed 400 µg/m<sup>3</sup>).

**Table 3.1.** Nitrogen dioxide (NO<sub>2</sub>) in 2015. All parameters are based on hourly averages.

Unit: µg/m <sup>3</sup>	Number	Average	Median	98. percentile	19. highest
<b>Street:</b>					
Copenhagen/1257	8105	33	28	83	108
Copenhagen/1103	8202	49*	47	108	135
Aarhus/6153	7930	31	27	80	103
Odense/9155 §§	3718	16	12	55	73
Aalborg/8151 §	0	-	-	-	-
<b>Urban Background:</b>					
Copenhagen/1259	7965	16	12	48	68
Aarhus/6160	7965	13	10	46	69
Odense/9159	8067	11	9	36	55
Aalborg/8159	8023	12	8	45	71
<b>Suburban:</b>					
Hvidovre/2650	4287	12	9	43	58
<b>Rural:</b>					
Risø	8075	8	5	32	52
Keldsnor/9055	7227	7	5	27	39
Limit value 2010	>7467**	40			200

\*) Limit value exceeded.

\*\*) 90% data capture of number of hourly measurements in relation to total number of hourly measurements in 2015 excluding hours used for calibration.

§) Aalborg/8151 (street) there is no data since the station has been shut down due to construction work at the site. It has not yet been possible to reinitiate the measurements in Aalborg (traffic).

§§) The site in Odense/9155 (Albanigade) was affected by a major permanent rearrangement of the roads in Odense. The station changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on 28 June 2014. The station was shut down on 16 June 2015 and has been moved to a new position in summer 2016.

**Table 3.2.** Nitrogen oxides ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ) in 2015. All parameters are based on hourly averages.

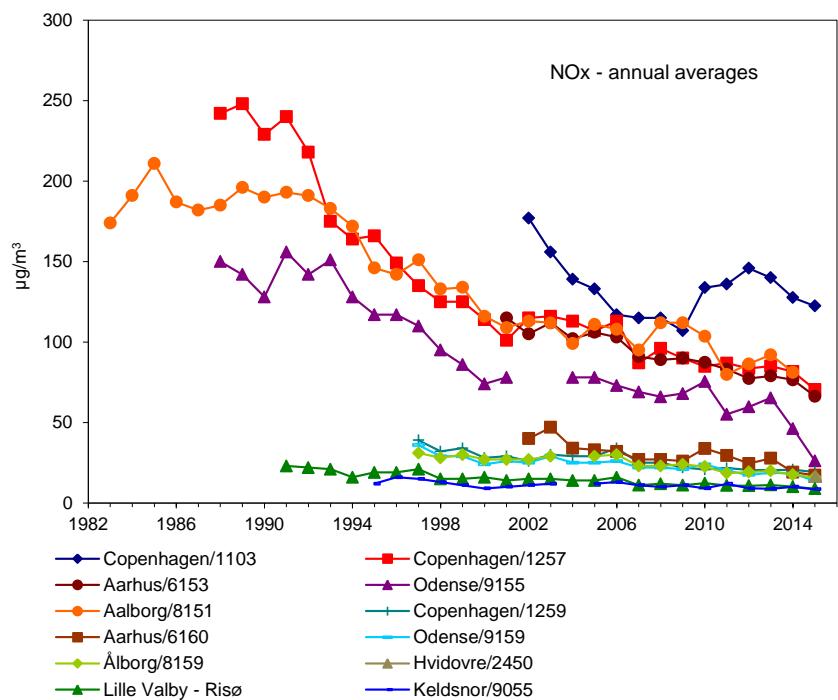
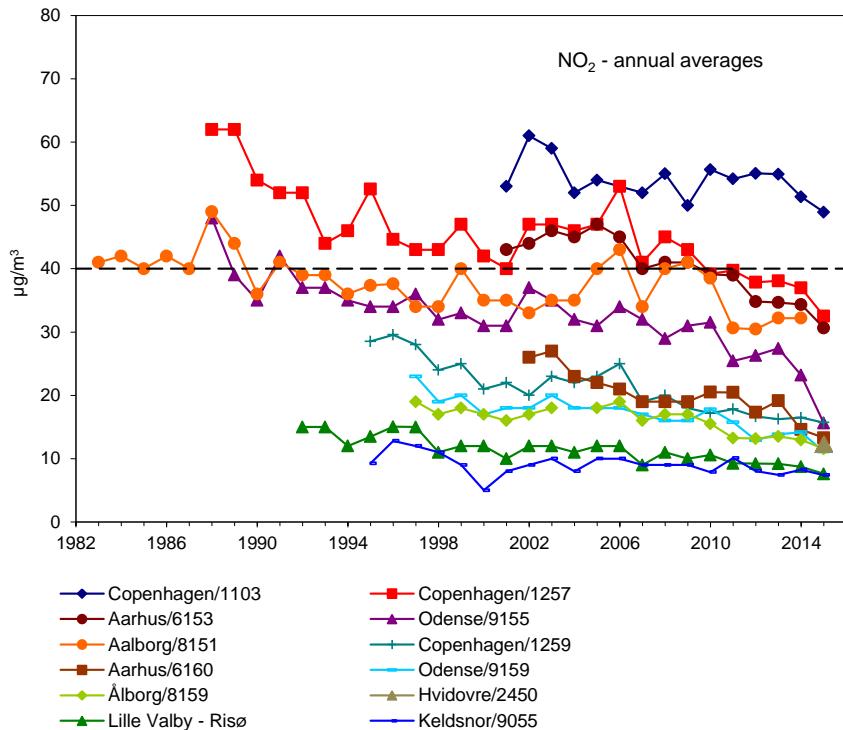
<b>Unit: <math>\mu\text{g}/\text{m}^3</math> (as <math>\text{NO}_2</math>)</b>	<b>Number</b>	<b>Average</b>	<b>Median</b>	<b>98. percentile</b>	<b>19. highest</b>
<b>Street:</b>					
Copenhagen/1257	8105	70	51	248	429
Copenhagen/1103	8202	122	102	377	590
Aarhus/6153	7930	66	48	238	450
Odense/9155	3718	26	16	143	205
Aalborg/8151	0	-	-	-	-
<b>Urban Background:</b>					
Copenhagen/1259	7965	19	14	68	118
Aarhus/6160	7965	17	12	71	195
Odense/9159	8067	14	10	52	120
Aalborg/8159	8023	16	10	72	218
<b>Suburban:</b>					
Hvidovre/2650	4287	17	10	92	208
<b>Rural:</b>					
Risø	8075	9	6	41	81
Keldsnor/9055	7227	9	6	32	50

### 3.2 Trends

The long term trends for  $\text{NO}_2$  and  $\text{NO}_x$  are shown in figure 3.1. For  $\text{NO}_x$  there are clear downward trends at all stations. The decreases in the concentrations of nitrogen oxides are due to the national and international regulations of the emissions. The large emission reductions in the cities are achieved by improvement of the vehicles, for example mandatory use of catalytic converters.

For many years the long term trend for nitrogen dioxide has decreased much slower than observed for  $\text{NO}_x$ . However, since around 2006  $\text{NO}_2$  has decreased with about the same rate as  $\text{NO}_x$ . The slow decrease before 2006 was mainly due to an increase in the share of diesel cars and increase in share of diesel cars with oxidative catalysts where up to about half of the emissions of  $\text{NO}_x$  consist of  $\text{NO}_2$  (called direct  $\text{NO}_2$ ). This increase of the direct emissions of  $\text{NO}_2$  counteracted the decrease in the traffic emissions from vehicles. The amount of directly emitted  $\text{NO}_2$  reached a maximum in around 2009-2011 and has slightly decreased after this. This change in the amount of directly emitted  $\text{NO}_2$  is believed to be one of the main reasons why  $\text{NO}_2$  now decreases at a similar pace as  $\text{NO}_x$ . At Odense street station and Aarhus urban background station there have been large decreases since 2013.

In Odense there was a major permanent rearrangement of the roads in Odense Centre that changed the traffic at the street station in Albanigade in two steps from a street with relatively high traffic intensity to a street with much reduced traffic intensity. Finally, the street was closed for traffic in 2015. These changes began on 28 June 2014. This is the reason for the large decrease of the  $\text{NO}_2$  and  $\text{NO}_x$  values for Odense/9155 in 2014 and 2015. The station was shut down on 16 June 2015 and was relocated to another position in summer 2016. The large change at Aarhus/background from 2013 to 2014 is due to the relocation of the measurement site in January 2014 (Chapter 2.1).



**Figure 3.1.** The graphs show the time series for the annual average values of NO<sub>2</sub> and NO<sub>x</sub>. The dashed line on the upper graph shows the limit value that entered into force in 2010. Results from the previous (6159) and the new background station (6160) in Aarhus are shown on the same curve.

In 2010 the driving lanes were changed at the section of H.C. Andersens Boulevard where the measurement station (Copenhagen/1103) is sited. This change moved the traffic closer to the measurement station and resulted in an increase of the annual average concentrations of NO<sub>2</sub> of about 8 µg/m<sup>3</sup> in comparison to the levels measured before the introduction of the new driving lanes. This increase in the concentrations of NO<sub>2</sub> is still measured, although there has been a general reduction of concentrations in line with the observations at other stations e.g. at Jagtvej.

### 3.3 Results from model calculations

Model calculations of NO<sub>2</sub> have been performed for selected streets in Copenhagen (capital) and Aalborg (fourth largest city).

The selected streets represent busy streets and are mainly so-called street canyons. Concentrations are elevated in this type of streets due to the high emissions and restricted dispersion conditions. 98 streets were selected in Copenhagen and 31 in Aalborg.

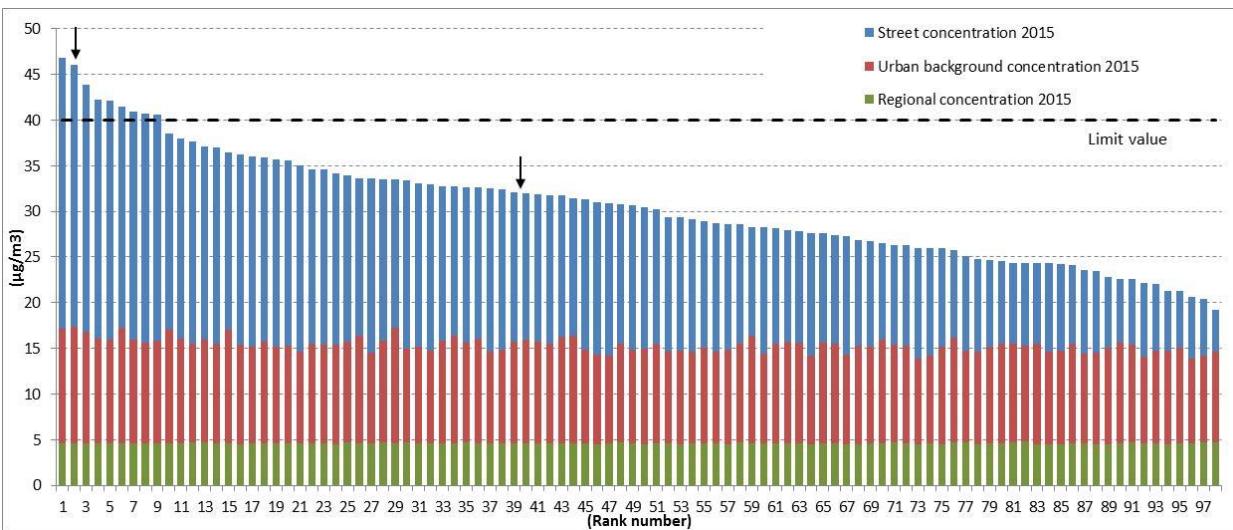
ADT (Average Daily Traffic) was between 5,400 and 67,600 vehicles/day in Copenhagen and between 2,700 and 28,300 vehicles/day in Aalborg.

Model calculations have been carried out in order to determine the annual concentrations of NO<sub>2</sub> for comparison with the limit values. The air quality limit value for the annual mean is 40 µg/m<sup>3</sup>. The number of streets with exceedances is one of the parameters discussed in the next section. An exceedance is registered if the calculated concentration is higher than 40.5 µg/m<sup>3</sup> since the limit value is given as an integer.

#### 3.3.1 NO<sub>2</sub> model calculations for Copenhagen

The annual mean concentrations of NO<sub>2</sub> for streets in Copenhagen in 2015 are shown in figure 3.2 (bar chart) and figure 3.3 (map).

The average of NO<sub>2</sub> concentrations at all 98 streets has decreased by 3% from 2014 to 2015 corresponding to 1 µg/m<sup>3</sup>. The general decrease is a result of a combination of changes in traffic, emission factors, urban background and meteorology. Modelled urban background concentrations decreased 4% or about 0.7 µg/m<sup>3</sup> which accounts for more than half of the reduction in street concentrations. Vehicle emission factors show a decrease due to the general replacement of the car fleet and leads to a decrease in modelled concentrations. There has been marginal changes in traffic as ADT increased 0.2%, heavy-duty share decreased 0.7% and travel speeds are assumed to be the same as in 2014. In 2015 the limit value for the annual mean concentration was exceeded in 9 out of the 98 selected streets in Copenhagen according to the model results (figure 3.2). This is slightly less than in 2014 where the number of streets exceeding the limit value was 11 out of 98. The number of streets exceeding the limit value is very sensitive to small changes in concentrations and uncertainties in the assumptions as can be seen from figure 3.2 where a number of streets are close to the limit value.



**Figure 3.2.** Annual mean concentrations of NO<sub>2</sub> in 2015 for 98 streets in Copenhagen according to model calculations. The contribution from traffic in the street canyons is based on the street canyon model OSPM® (blue colour). The urban background (reddish colour) is obtained from calculations with the urban background model UBM with input from the regional scale model DEHM (green colour). The value for a street segment is for the side of the street with the highest annual mean concentration of the two sides. However, for streets with a measuring station it is the side where the station is located. The names of the streets can be seen in table 3.3. Arrows indicate street segments with a measuring station.

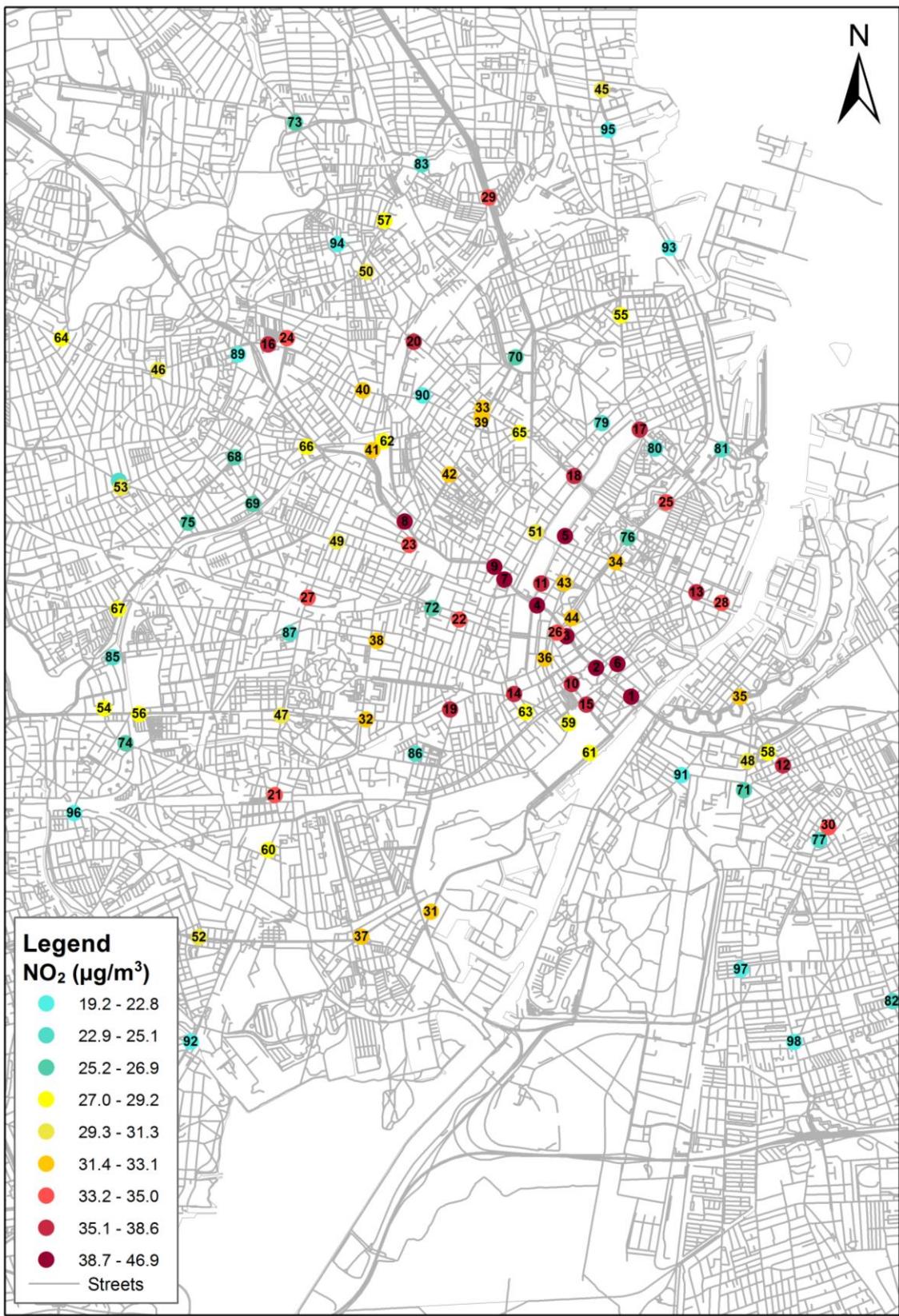
The streets where the limit value were exceeded all have average daily traffic in the range of 12,800 to 67,600 vehicles per day. However, it is not only the traffic intensity which determines the concentration of NO<sub>2</sub>. Also the travel speed, vehicle distribution, and street geometry like the width of the streets, the height of the surrounding buildings, openings in the building facade, orientation of the street and background concentrations and meteorology have impacts on the concentration of NO<sub>2</sub> in a street.

The names of the 98 streets are given in table 3.3 and the locations of the streets together with the annual NO<sub>2</sub> concentration levels are shown in figure 3.3. It is seen that the exceedances are concentrated in the central part of the city and at the main arterial roads from H.C. Andersens Boulevard to Ågade, and also Stormgade and Øster Søgade.

There have been minor changes in the ranking of streets according to NO<sub>2</sub> concentrations from 2014 to 2015 due to mainly small changes in traffic inputs. The highest modelled NO<sub>2</sub> concentration in 2015 is at H.C. Andersens Boulevard (2) (47 μg/m<sup>3</sup>). The second highest (46 μg/m<sup>3</sup>) is where the measuring station is located (H.C. Andersens Boulevard (1)). Observed concentrations are 49/41 μg/m<sup>3</sup> (explanation in table 2.2). This location was ranked number one in 2014.

**Table 3.3.** Number and names for the street segments that are shown in figure 3.2 and 3.3. The streets are numbered (1-98) according to NO<sub>2</sub> levels in 2015 (1 = highest, 98 = lowest). The numbers in parentheses refer to different segments of the same street that has more than one model calculation. An asterisk (\*) indicates a street segment with a measurement station. Note that Vester Voldgade is not included, as the road segment, where the calculation point is, has been closed due to construction of new Metro line.

No.	Street name	No.	Street name	No.	Street name
1	H C Andersens Boulevard(2)	34	Gothersgade(1)	67	Grøndals Parkvej
2	H C Andersens Boulevard(1)*	35	Torvegade	68	Hulgårdsvej(2)
3	H C Andersens Boulevard(3)	36	Vester Farimagsgade	69	Godthåbsvej(2)
4	Gyldenløvesgade	37	P Knudsens Gade(2)	70	Jagtvej(2)
5	Øster Søgade	38	Gammel Kongevej(1)	71	Røde Mellemvej(1)
6	Stormgade	39	Jagtvej(1)*	72	Bülowsvæj(2)
7	Åboulevard(1)	40	Frederikssundsvej(1)	73	Frederiksborgvej(1)
8	Ågade	41	Nordre Fasanvej(3)	74	Ålholmvej(2)
9	Åboulevard(3)	42	Jagtvej(3)	75	Rebildvej
10	Bernstorffsgade(1)	43	Nørre Farimagsgade	76	Øster Voldgade(2)
11	Nørre Søgade	44	Nørre Voldgade(2)	77	Englandsvej(1)
12	Amagerbrogade(2)	45	Strandvejen(1)	78	Frederikssundsvej(2)
13	Bredgade	46	Frederikssundsvej(8)	79	Blegdamsvej
14	Vesterbrogade(1)	47	Søndre Fasanvej(2)	80	Dag Hammarskjölds Allé
15	Bernstorffsgade(2)	48	Amagerfælledvej	81	Folke Bernadottes Allé
16	Frederikssundsvej(3)	49	Godthåbsvej(3)	82	Amagerbrogade(3)
17	Østerbrogade(4)	50	Tagensvej(1)	83	Tuborgvej(1)
18	Fredensgade	51	Nørrebrogade	84	Slotsherrensvej(2)
19	Enghavevej	52	Folehaven(1)	85	Peter Bangs Vej(1)
20	Tagensvej(2)	53	Jyllingevej(1)	86	Vesterfælledvej
21	Toftegård's Allé(1)	54	Ålholmvej(1)	87	Peter Bangs Vej(2)
22	H.C. Ørsted's Vej(2)	55	Østerbrogade(1)	88	Slotsherrensvej(1)
23	Falkoner Alle(2)	56	Roskildevej(1)	89	Bellahøjvej
24	Tomsgårdsvæj(2)	57	Tuborgvej(2)	90	Halmetgade
25	Øster Voldgade(1)	58	Amager Boulevard	91	Artillerivej
26	Hammerichsgade	59	Ingerslevsgade	92	Gammel Køge Landevej(2)
27	Nordre Fasanvej(1)	60	Gammel Køge Landevej(1)	93	Strandvænget(2)
28	Toldbodgade	61	Kalvebod Brygge	94	Frederiksborgvej(2)
29	Lyngbyvej(2)	62	Hillerødsgade(1)	95	Strandvejen(2)
30	Amagerbrogade(1)	63	Istedgade	96	Vigerslevvej(2)
31	Scandiagade	64	Frederikssundsvej(5)	97	Røde Mellemvej(2)
32	Vesterbrogade(3)	65	Tagensvej(4)	98	Englandsvej(2)
33	Tagensvej(3)	66	Hillerødsgade(3)		

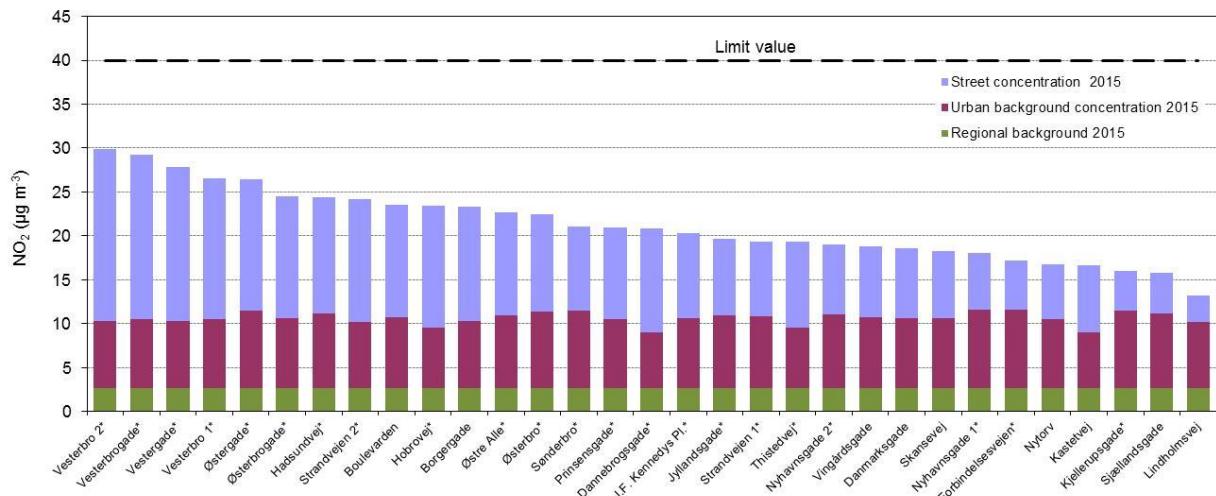


**Figure 3.3.** Map showing the locations of the selected streets in Copenhagen and the annual mean concentrations of NO<sub>2</sub> for 2015. The contribution from traffic in the street canyons is based on the street canyon model OSPM®. The urban background is obtained from calculations with the urban background model UBM with input from the regional scale model DEHM. The value for a street segment is for the side of the street with the highest annual mean concentration of the two sides. However, for streets with a measurement station it is the side where the station is located. The names and numbers for the streets are shown in table 3.3. The map can be viewed at a webGIS service, see <http://arcg.is/1Z5u0r6>.

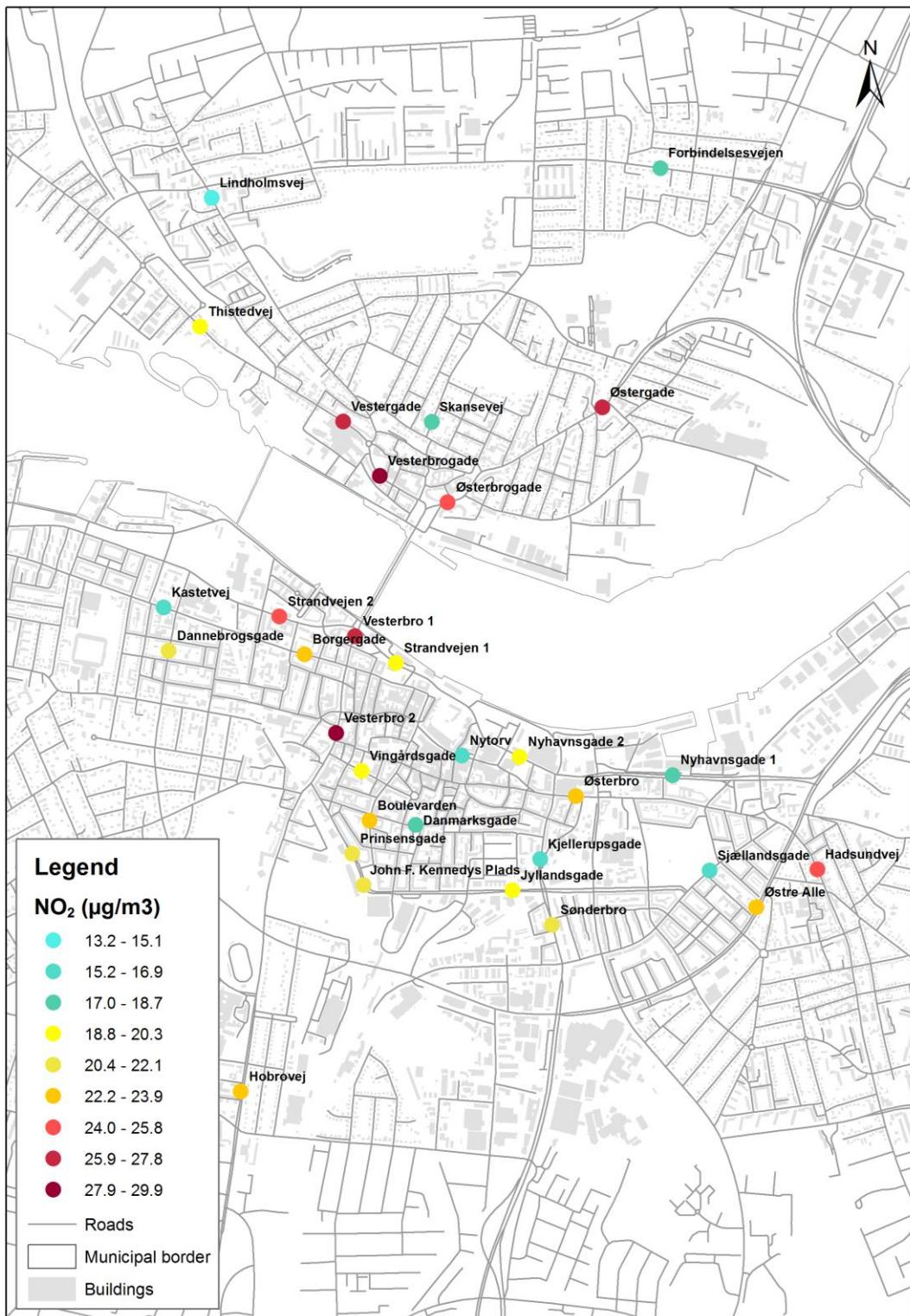
### 3.3.2 NO<sub>2</sub> model calculations for Aalborg

For Aalborg the modelled street concentrations show an average decrease of 10% for NO<sub>2</sub> compared to 2014 when considering all 31 street segments, corresponding to a decrease of about 2 µg/m<sup>3</sup>. The general decrease is a result of a combination of several factors. The contribution from regional and urban background is almost entirely responsible for the modelled decrease. On average ADT increased about 0.6% whereas the heavy-duty share of vehicles was unchanged, and travel speeds were assumed to be unchanged. This would - all other things equal - slightly increase concentrations due to these changes in traffic inputs. Reduced emissions due to replacement of the car fleet should lead to lower concentrations.

According to the model calculations the limit value for the annual mean concentration in 2015 was not exceeded at any of the 31 selected streets which was also the case in 2014 (figure 3.4 and figure 3.5). The order of some of the streets has changed slightly due to changes in traffic data.



**Figure 3.4.** Modelled annual mean concentrations of NO<sub>2</sub> in 2015 for 31 streets in Aalborg. The contribution from traffic in the street canyons is based on the street canyon model OSPM® (blue colour). The urban background (dark red colour) is obtained from calculations with the urban background model UBM (reddish colour) with input from the regional scale model DEHM (green colour). The value for a street segment is for the side of the street with the highest annual mean concentration of the two sides. However, for streets with a measurement station it is the side where the station is located. Vesterbro 1 is the street segment where the measurement station is located. However, the station was not operational during 2015 due to nearby building construction works. An asterisk (\*) at the end of the street name indicates that calculations were based on travel speed data from SpeedMap, whereas an average of these travel speeds were used for the rest of the streets.



**Figure 3.5.** Map showing the location of the selected streets in Aalborg and the annual mean concentrations of NO<sub>2</sub> for 2015. The contribution from traffic in the street canyons is based on the street canyon model OSPM®. The urban background is obtained from calculations with the urban background model UBM with input from the regional scale model DEHM. The value for a street segment is for the side of the street with the highest annual mean concentration of the two sides. However, for streets with a measurement station it is the side where the station is located. Vesterbro 1 is the street segment with the measurement station, however, not operating in 2015 due to nearby building construction work. Map can be viewed at a webGIS service, see <http://arcg.is/1RvRuzS>

### 3.3.3 Trends in modelled exceedances of NO<sub>2</sub>

In Figure 3.6 modelled trends in exceedances of annual mean of NO<sub>2</sub> are shown for Copenhagen and Aalborg. The limit value of 40 µg/m<sup>3</sup> for annual mean of NO<sub>2</sub> had to be met in 2010 and in previous years the limit value plus a margin of tolerance depending on the year in question had to be met.

For Copenhagen the number of exceedances has decreased from 58 in 2008 to 9 in 2015.

The main reason for the increase in number of exceedance in Copenhagen from 32 in 2007 to 58 in 2008 is the following. The limit value plus margin of tolerance for the annual mean concentration of NO<sub>2</sub> decreased from 46 µg/m<sup>3</sup> in 2007 to 44 µg/m<sup>3</sup> in 2008 (EC, 2008). This decrease naturally lead to a higher number of streets exceeding the limit value plus margin of tolerance in 2008 compared to 2007. If the limit value plus margin of tolerance had been 44 µg/m<sup>3</sup> in 2007, then the number of streets exceeding the limit value plus margin of tolerance would have been 53. Roughly the same level as in 2008.

In Copenhagen the analysis includes 138 streets during 2007 to 2010 and 98-99 the following years. The reduction of included streets from 2011 and onwards was implemented to better match locations of selected streets with locations with manual traffic counts.

For Aalborg 3-4 exceedances were modelled in 2007-2009 and none since 2010. Here the analysis includes 32 streets from 2007 to 2010, and 31 streets from 2011 to 2015.

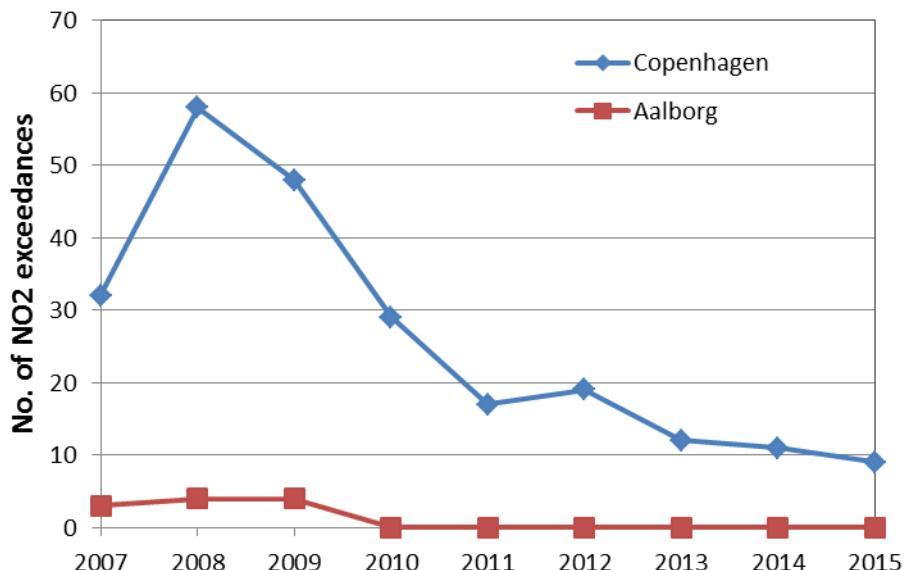


Figure 3.6. Trends in modelled exceedances of annual mean of NO<sub>2</sub> in Copenhagen and Aalborg.

## 4 Ozone

Ozone is measured at seven monitoring sites using gas monitors based on ultraviolet photometry. The concentrations are measured continuously throughout the year with a time resolution on minute scale that is aggregated to hourly averages for the present reporting.

### 4.1 Annual statistics

The annual statistics for 2015 for ozone are shown in table 4.1. The maximum 8 hours daily mean value must not exceed 120 µg/m<sup>3</sup> more than 25 days per calendar year averaged over three years (EC, 2008). This target value was not exceeded for 2013-2015 at any of the stations. The long term objective (maximum 8 hours daily mean value must not exceed 120 µg/m<sup>3</sup>; table 4.1 column 5) was exceeded at six of the stations. However, the long term objective has not entered into force.

In 2015 there was no exceedance of the information threshold (hourly average 180 µg/m<sup>3</sup>) and no exceedance of the alert threshold (hourly average 240 µg/m<sup>3</sup>) for ozone.

**Table 4.1.** Ozone (O<sub>3</sub>) in 2015. All parameters are based on one-hour average values. The 8 hour values are calculated as a moving average based on hourly measurements. Days above target value is the number of days that the maximum running 8 hour average exceeds 120 µg/m<sup>3</sup> averaged over 2013-2015.

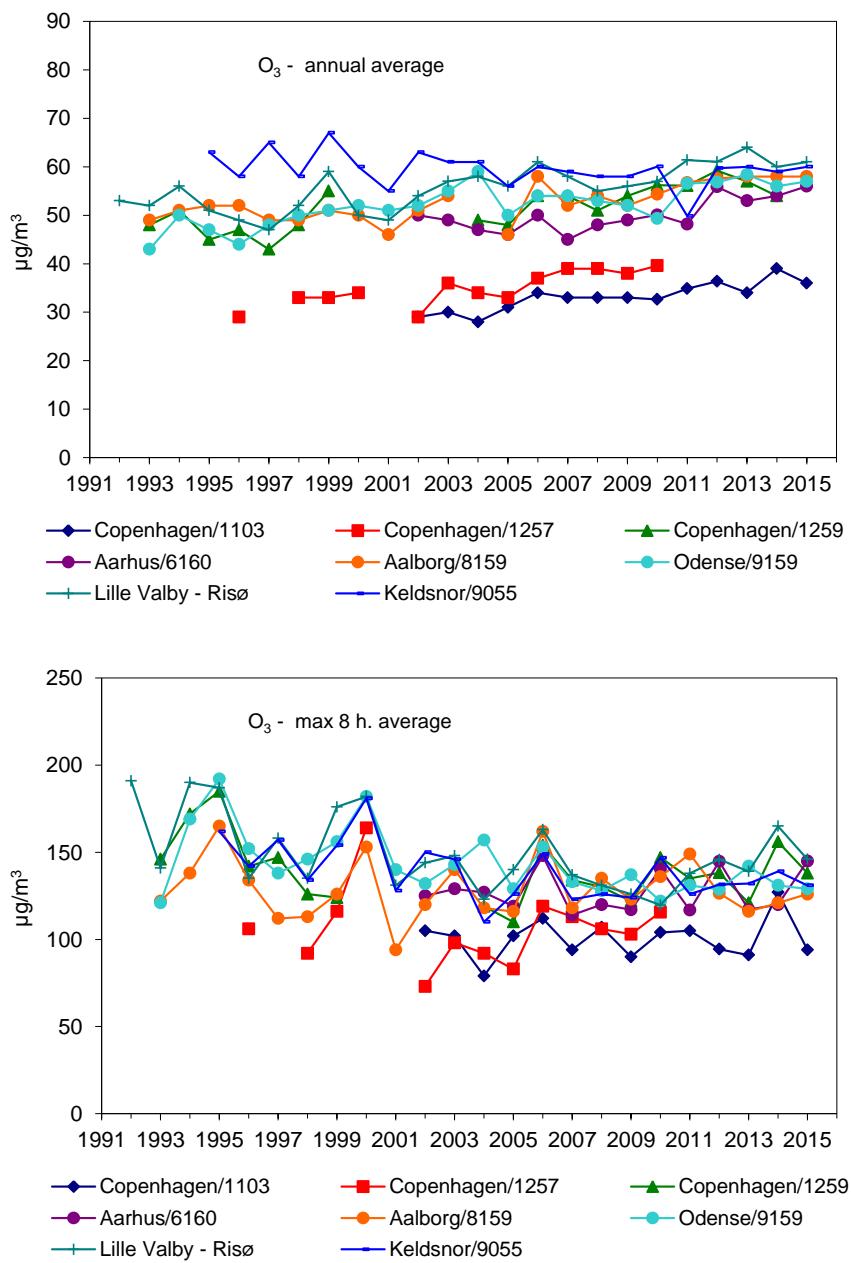
Unit: µg/m <sup>3</sup>	Number of results	Average	Median	Max 8 hours	Days above target value 8 hours	Max 1 hour
<b>Urban Background:</b>						
Copenhagen/1259	7603	59	61	138	4	153
Aarhus/6160	7233	56	59	145	1	162
Odense/9159	7735	57	59	129	4	146
Aalborg/8158	7124	58	61	126	1	147
<b>Rural</b>						
Risø/2090	7941	61	63	146	7	167
Keldsnor/9055	7698	60	62	131	3	155
<b>Traffic</b>						
Copenhagen/1103	7728	36	37	94	0	114
Target value <sup>1</sup>	-	-	-	-	25	-
Long term objective	-	-	-	120	-	-
Information threshold	-	-	-	-	-	180
Data capture*	>7154	-	-	-	-	-

<sup>1</sup> As average over 3 years.

\*) 90% data capture of number of hourly measurements in relation to total number of hourly measurements in 2015 excluding hours used for calibration.

## 4.2 Trends

The long term trends of ozone are shown in figure 4.1. The annual averages of ozone have been nearly constant since 1992. The Danish and European reductions of the precursors to ozone formation ( $\text{NO}_x$ , volatile organic compounds) have therefore not been sufficient to reduce the ozone concentration. However, the reductions of the precursors have decreased the maximum concentrations of ozone. This is illustrated by the decrease in the maximum eight hour average concentrations.



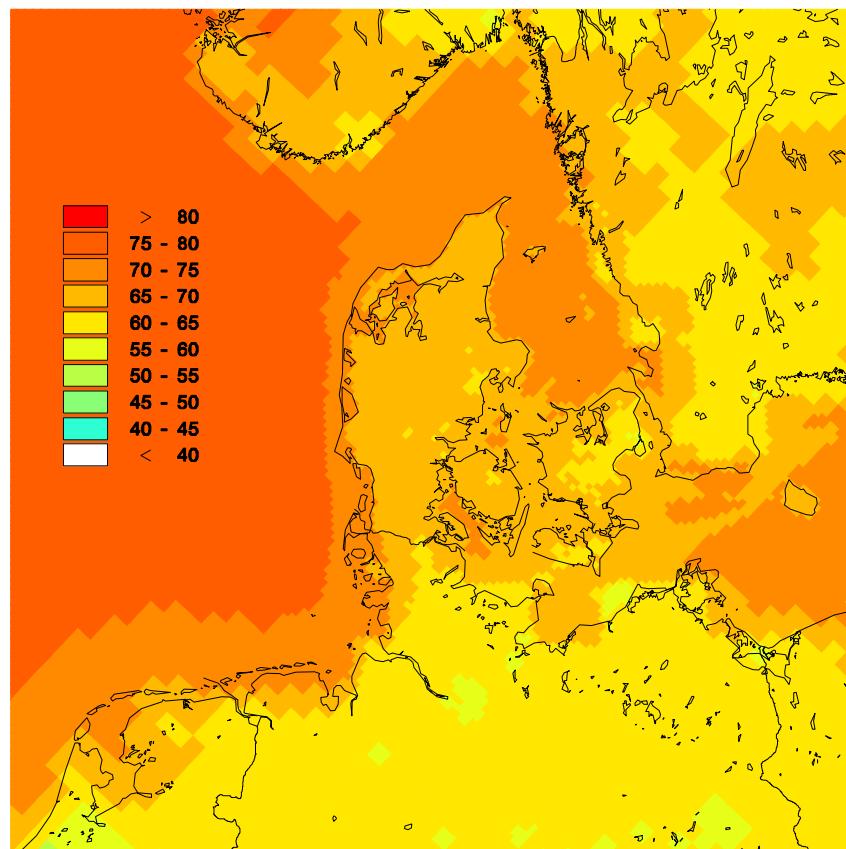
**Figure 4.1.** Annual average values and the max. 8 hour average value. The latter is calculated as 8 hourly running averages according to the provisions in the EU Directive (EC, 2008). Results from the previous (6159) and the new background station (6160) in Aarhus are shown on the same curve.

### **4.3 Results from model calculations**

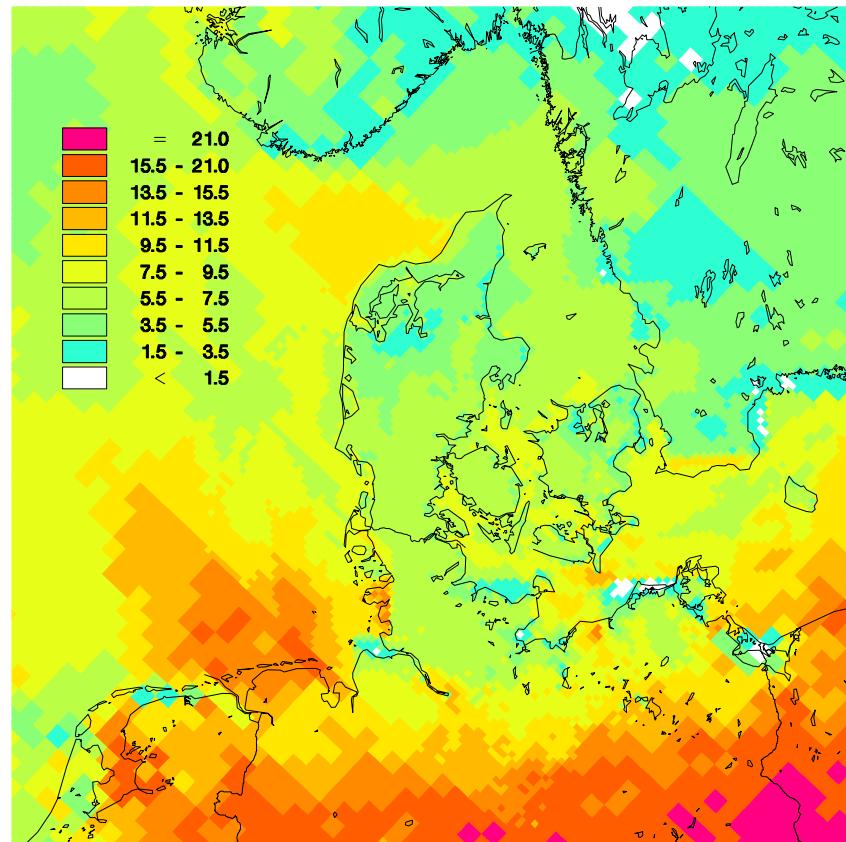
The annual mean concentration of ozone is fairly constant throughout Denmark (figure 4.2). This is because the main production of ozone takes part in the southern part of Europe and ozone is subsequently long range transported to Denmark. At the coasts the concentrations are slightly higher than over the remaining land areas, because ozone is deposited faster over land than over sea. In the cities the concentrations are lower than the average, because ozone is degraded by nitrogen oxide emitted from mainly traffic in the cities. This is clearly seen for Copenhagen. Both model results and measurements show slightly higher ozone concentrations in 2015 compared to the previous years. This is due to a slightly higher long range transport of ozone to Denmark than in 2014.

The target value for protection of human health is that the running 8 hour mean concentration of ozone must not exceed  $120 \mu\text{g}/\text{m}^3$  more than 25 times during a calendar year calculated as an average over three years. The long term objectives are that the running 8 hour mean concentration of ozone must not exceed  $120 \mu\text{g}/\text{m}^3$ . The target value and long term objective are given in the EU Directive (EC, 2008). Results from the model calculations for 2015 show that the number of days with maximum daily 8 hour mean value above  $120 \mu\text{g}/\text{m}^3$  was well below the target value for the entire country. Highest number of days was seen at coastal areas where the maximum number of days reached 13 days above  $120 \mu\text{g}/\text{m}^3$  (figure 4.3). The target value that is determined as an average over three years (2013-2015), was not exceeded. However, the long term objective was exceeded all over Denmark (figure 4.4). The highest 8 hour mean concentrations were observed at coastal areas due to slow deposition over sea and long range transport of ozone.

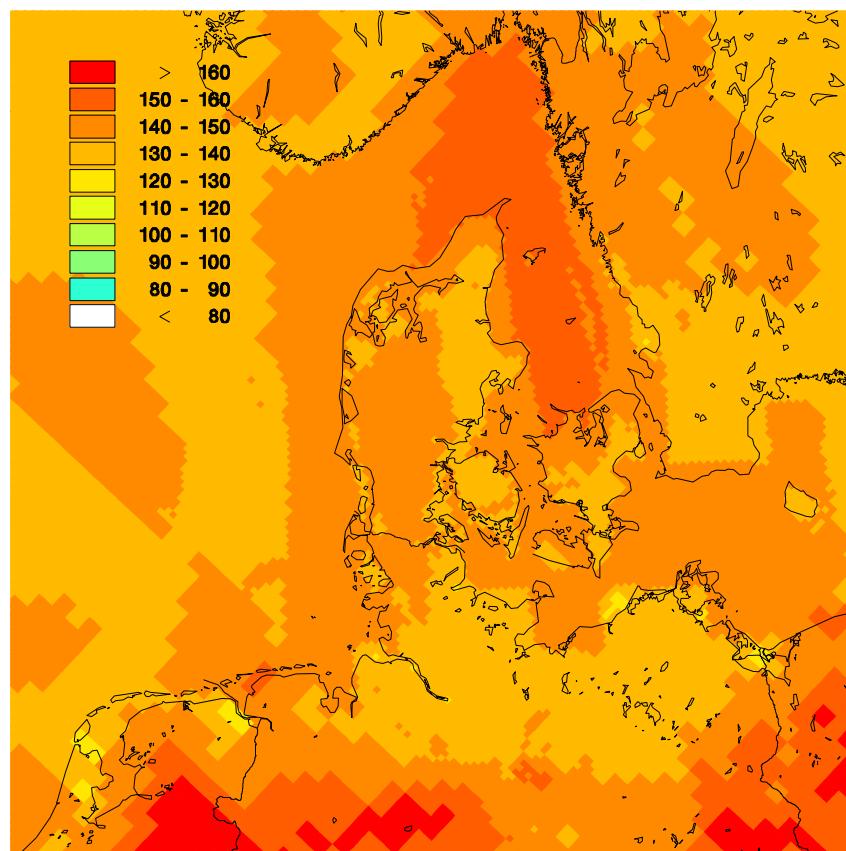
According to the directive (EC, 2008) the public has to be informed if the one hour average concentration exceeds the information threshold at  $180 \mu\text{g}/\text{m}^3$ . Both measurements and model calculations shows that this threshold was not exceeded in 2015 (figure 4.5). The model calculations underestimate the maximum one hour mean concentration with about 10-20%. One of the reasons for this discrepancy is most likely that the model does not include emissions of ozone precursors from wild fires that are known to increase episodic ozone concentrations.



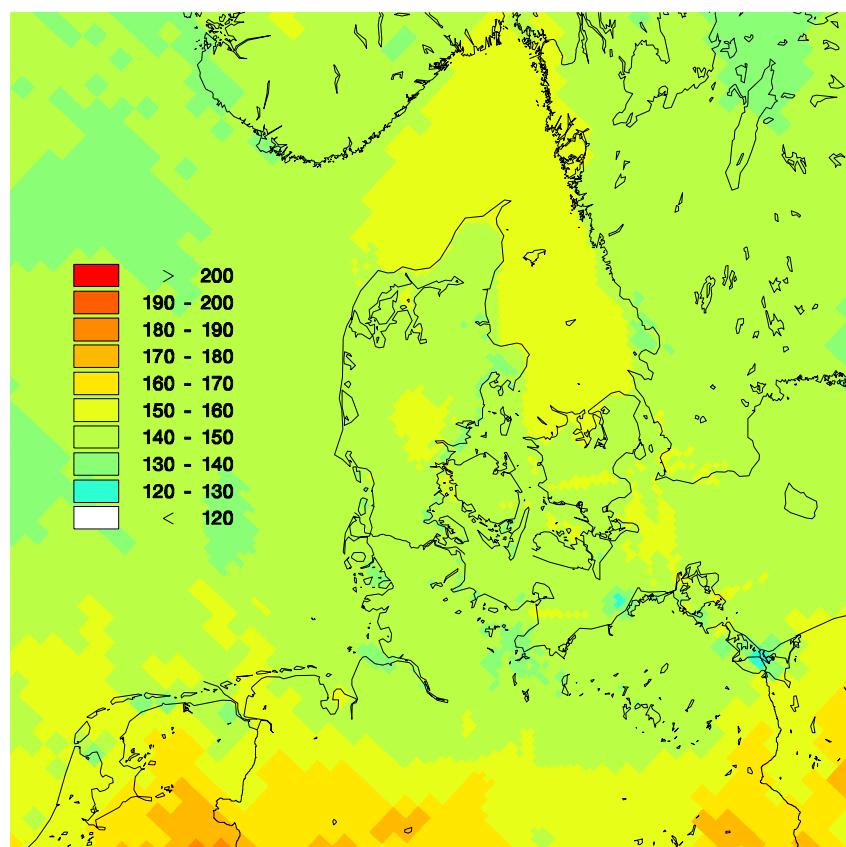
**Figure 4.2.** Annual mean concentrations of O<sub>3</sub> (µg/m<sup>3</sup>) for 2015 calculated using DEHM. The figure shows the average concentrations for the 6 km x 6 km grid cells used in the model.



**Figure 4.3.** Number of exceedances of 120 µg/m<sup>3</sup> for 8-hour running mean concentrations of ozone in 2015. The calculations were carried out using DEHM.



**Figure 4.4.** Maximum 8 hour running mean concentration ( $\mu\text{g}/\text{m}^3$ ) of ozone in 2015 calculated using DEHM.



**Figure 4.5.** Maximum one hour mean concentration of ozone ( $\mu\text{g}/\text{m}^3$ ) in 2015 calculated using DEHM.

## 5 Carbon monoxide

Carbon monoxide is at present measured at three traffic oriented monitoring sites (Aalborg street is temporarily closed down), at the urban background site in Copenhagen and at the rural site at Risø using gas monitors based on non-dispersive infrared spectroscopy. The concentrations are measured continuously throughout the year with a time resolution on minute scale that is aggregated to hourly averages for this reporting.

### 5.1 Annual statistics

The annual statistics for 2015 for carbon monoxide are shown in table 5.1. The limit value for carbon monoxide is based on the maximum daily 8 hour average concentration that must not exceed 10.000 µg/m<sup>3</sup> (EC, 2008). This limit value was not exceeded at any of the stations.

**Table 5.1.** Annual statistics for carbon monoxide (CO) in 2015. All parameters are based on hourly average. The 8-hour values are calculated as a moving average based on hourly results.

Unit: µg/m <sup>3</sup>	Number	Average	Median	98-percentile	99.9-percentile	Max. 8-hours	Max. hour
<b>Traffic:</b>							
Copenhagen/1103	7762	355	327	766	1045	942	1274
Århus/6153	8072	233	209	526	758	677	1095
Odense/9155 §	3724	255	255	591	897	816	997
Aalborg/8151 §§	0	-	-	-	-	-	-
<b>Urban Background:</b>							
Copenhagen/1259	7730	194	183	386	570	559	598
<b>Rural:</b>							
Risø	7988	173	162	322	465	473	604
Data capture*	>7467	-	-	-	-	-	-
EU Limit value	-	-	-	-	-	10 000	-
WHO Guideline values (WHO, 2000)	-	-	-	-	-	10 000	30 000

\*) 90% data capture of number of hourly measurements in relation to total number of hourly measurements in 2015 excluding hours used for calibration.

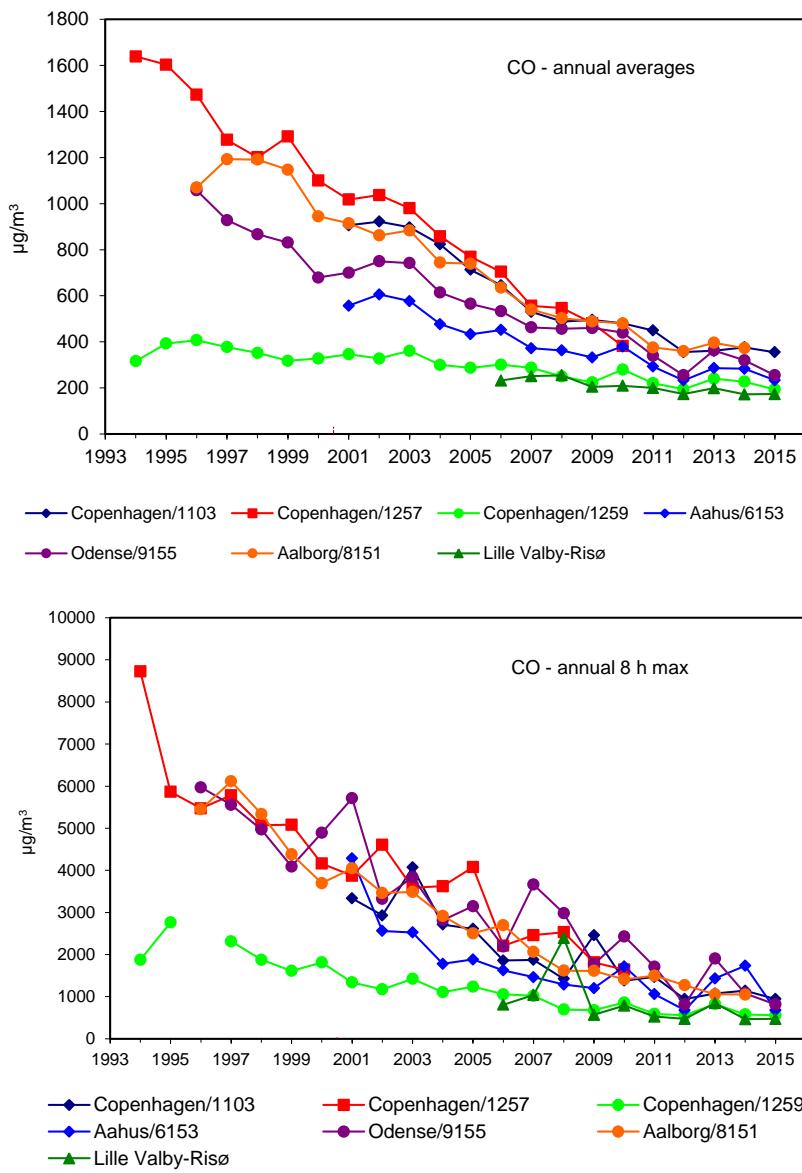
§) The site in Odense/9155 (Albanigade) was affected by a major permanent rearrangement of the roads in Odense. It changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on 28 June 2014. The station was shut down on 16 June 2015 and has been moved to a new position during summer 2016.

§§) Aalborg/8151 (traffic) there is no data since the station has been shut down due to construction work at the site. It has not yet been possible to reinitiate the measurements at the street station in Aalborg .

## 5.2 Trends

The long term trends for carbon monoxide are shown in figure 5.1. During the last two decades there has been a large decrease of both the annual concentrations and of the maximum daily eight hour average concentrations. The reductions are due to national and international regulation of the emissions, among others by requirement of catalytic converters on all vehicles.

At the street stations in Odense/9155 (Albanigade) there was a larger reduction in CO from 2013 to 2015 than at the other stations. This is due to a major permanent rearrangement of the roads in Odense that resulted in a large reduction in the traffic intensity in Albanigade.



**Figure 5.1.** Annual average values and highest 8-hour values calculated based on an hourly moving average. The site in Odense/9155 (Albanigade) was due to a major permanent rearrangement of the roads in Odense. It changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on 28 June 2014.

## **6 Benzene and other Volatile Organic Compounds**

This section merges previous years sections on *Ozone Precursors* and *Benzene and Toluene*, all of which are Volatile Organic Compounds (VOC).

Benzene, toluene, ethylbenzene and xylenes are monitored on two kerbside stations in Copenhagen with weekly resolution, i.e. Jagtvej/1257 and H.C. Andersen's Boulevard/1103. These anthropogenic VOCs are collected using passive sampling, and subsequently extracted and analysed by Gas Chromatography MS (GC-MS).

In addition to this benzene and toluene is also measured at the urban background station (Copenhagen/1259). Here a comprehensive set of 18 potential ozone precursor VOCs is measured with diurnal time resolution. The focus is VOCs of anthropogenic origin, though also isoprene is monitored, which is typically from deciduous trees. Atmospheric samples are preconcentrated on the adsorbent Carbopack X using active sampling, and analysed using Thermal Desorption Gas Chromatography Mass Spectrometry (TD-GC-MS).

### **6.1 Annual statistics and trends**

Annual averages of benzene and toluene for 2015 are listed in Table 6.1 and 6.2. Benzene is well below the EU-limit value of 5 µg/m<sup>3</sup> (EC, 2008), averaging 0.79 µg/m<sup>3</sup> and 0.37 µg/m<sup>3</sup> at the kerbside stations and urban background, respectively. The local input of benzene from traffic amounts to 54% of the ambient kerbside concentration. For toluene, the local input is as high as 64%, which indicates that other sources contribute to ambient benzene besides traffic. One source is residential wood combustion, and for this reason the summer concentrations of benzene are lower even at kerbside stations, which are highly influenced by traffic. Both kerbside stations in Copenhagen show similar concentrations of anthropogenic aromatic compounds, including toluene and benzene (Table 6.1), in spite of their differences with respect to traffic load and buildings close to the street. These VOCs decreased dramatically at the kerbside stations during 2004-2008 (Figure 6.1) and has continued to decrease, though at a slower yet comparable rate in the urban environment. In fact, benzene has decreased 36% and 39% at the kerbside station and urban background, respectively, from 2010 to 2015. With respect to toluene, the corresponding decays are 31% and 50%, respectively. Of the monitored VOCs at kerbside, toluene is by far the most abundant. Other aromatic compounds are comparable in abundance to benzene (Table 6.1).

**Table 6.1.** Annual statistics for benzene, toluene, ethylbenzene and xylenes in 2015 based on weekly average concentrations ( $\mu\text{g}/\text{m}^3$ ) at kerbside stations Jagtvej (1257) and H.C.Andersens Boulevard (1103) at 1 atm., 293 K. The limit value for benzene is 5  $\mu\text{g}/\text{m}^3$  (EU Directive 2008/50/EC).

Concentration $\mu\text{g}/\text{m}^3$	Copenhagen/1103	Copenhagen /1257	Number of results
<b>Benzene</b>	0.83	0.79	52, 52
<b>Toluene</b>	2.12	2.07	52, 52
Ethylbenzene	0.53	0.52	52, 52
m/p-Xylene	1.36	1.35	52, 52
o-Xylene	0.55	0.55	52, 52

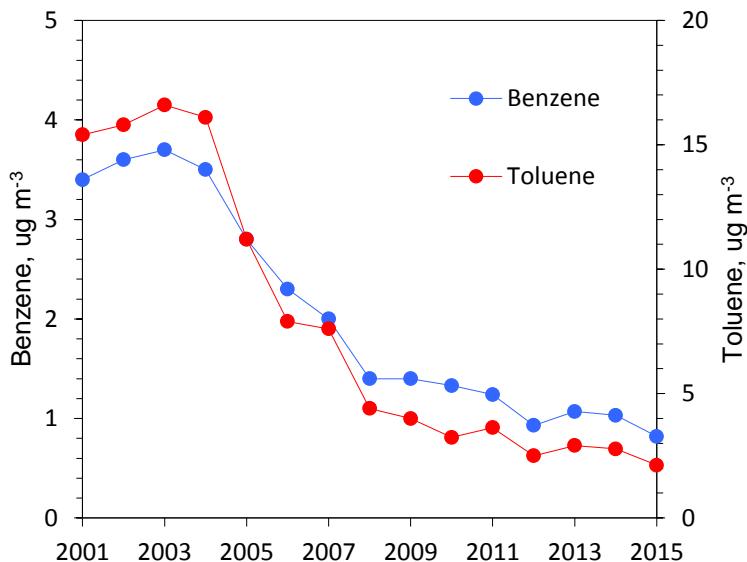
Benzene is not measured directly in Aarhus and Odense. However an objective estimate of the concentrations can be used to determine the concentration levels, since the concentrations are below the lower assessment threshold limit.

The objective estimate for benzene is based on the correlations between the average concentrations of benzene and CO. Ellermann et al. (2011) documented that the benzene concentrations can be estimated based on the simple empirical model:

$$\text{Benzene} = 0.0044 \cdot \text{CO} - 0.37$$

where benzene and CO are in units of  $\mu\text{g}/\text{m}^3$ .

Based on this and the concentrations of CO (table 5.1) the annual average concentrations of benzene is estimated to about 0.7  $\mu\text{g}/\text{m}^3$  for all the three street stations in Aarhus, Odense and Aalborg in 2015.



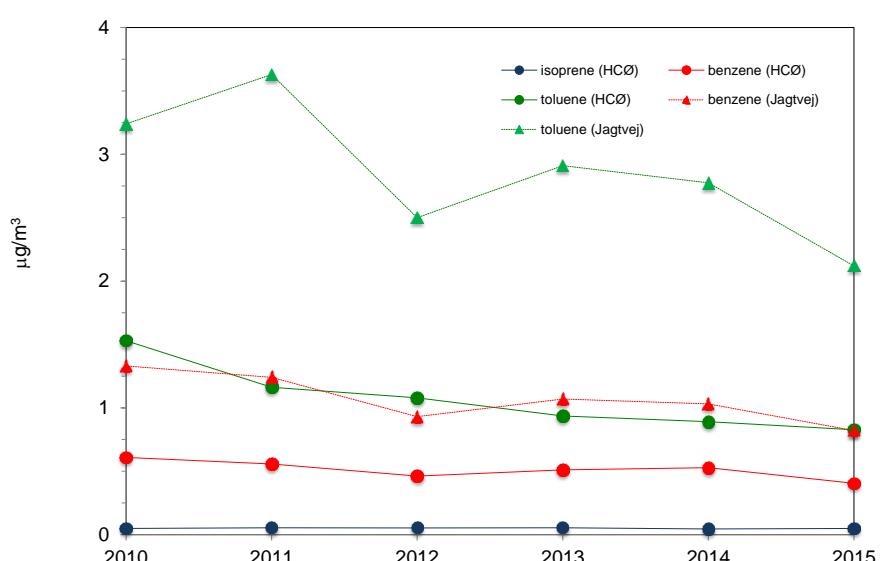
**Figure 6.1.** Trend in benzene and toluene (annual averages) on the kerbside station Jagtvej, Copenhagen/1257.

The main reasons for the significant decrease of benzene and toluene up to 2008 are reductions of the emissions from gasoline-fueled traffic due to increased use of catalysts and higher ratio of diesel cars.

**Table 6.2.** Annual statistics for VOCs in urban background in Copenhagen (1259) based on daily average concentrations (1 atm., 293 K).

Concentration $\mu\text{g}/\text{m}^3$	Average concentration	Annual trend 2010-2015	Number of results
1-Pentene	0.03	-0.3%	319
n-Pentane	0.44	+2%	323
Trans-2-pentene	0.01	-0.2%	324
Isoprene	0.04	-0.2%	322
2-Methylpentane	0.22	-2%	313
n-Hexane	0.11	-1%	280
<b>Benzene</b>	<b>0.37</b>	<b>-4%</b>	<b>324</b>
n-Heptane	0.14	-0.5%	324
2,2,2-Trimethylpentane	0.06	-0.3%	292
<b>Toluene</b>	<b>0.75</b>	<b>-14%</b>	<b>325</b>
n-Octane	0.04	-0.2%	325
Ethylbenzene	0.13	-2%	325
m,p-Xylene	0.40	-6%	325
o-Xylene	0.16	-2%	325
1,3,5-Trimethylbenzene	0.04	-0.3%	96
1,2,4-Trimethylbenzene	0.16	-1%	280
1,2,3-Trimethylbenzene	0.04	-0.3%	324
Sum of VOC's	3.14		

Measurements of mainly anthropogenic volatile organic compounds in urban background, which may act as ozone precursors, were initiated in 2010 in the urban background. The major ozone precursors are the aromatic compounds: benzene, toluene, ethylbenzene, xylenes and trimethylbenzenes (TMB), which are also measured at the kerbside stations in Copenhagen (1103 and 1257), and the C<sub>5</sub>-C<sub>7</sub> alkanes: pentane, 2-methylpentane hexane and heptane. The more reactive unsaturated compounds are less abundant (Table 6.2).



**Figure 6.2.** Annual average concentrations of benzene, toluene and isoprene at the kerbside station, Copenhagen/1257, and at urban background, Copenhagen 1259.

In contrast to the anthropogenic VOC's (Figure 6.2), the annual average concentration of isoprene has stayed constant throughout the monitoring period (Table 6.2). Isoprene origins mainly from terrestrial vegetation and peaks in the warmer summer months June, July and August with low concentrations in the winter months. Except for n-pentane, all anthropogenic VOC's either stayed constant during 2010-2015 or decreased (Table 6.2). Toluene has decreased 14% annually. A number of other VOCs has decreased more than 1% annually, i.e. 2-methylpentane, n-hexene, benzene, ethylbenzene, the isomers of xylene, and 1,2,4-trimethylbenzene.

The urban background ratio between toluene and benzene is somewhat smaller at both kerbside stations sites, i.e. 2.0 versus 2.6. This indicates different sources to benzene and toluene and faster atmospheric decomposition of toluene.

## **7 Particles (TSP, PM<sub>10</sub>, PM<sub>2.5</sub> and particle number)**

As part of an overall maintenance plan for the measuring programme it has been decided to substitute the old SM200 samplers with measurements of PM that follows the reference method for the determination of PM<sub>10</sub> and PM<sub>2.5</sub> (EN 12341: 2014, into which the previous standards for PM<sub>10</sub>, EN 12341: 1998, and for PM<sub>2.5</sub>, EN 14907:2005, have been merged). The basic measuring principle of the reference method uses Low Volume Sampling (LVS) i.e. a flow of 2,3 m<sup>3</sup>/hr with gravimetric determination of the sampled mass in the laboratory. Results from comparison of LVS gravimetric determination and the SM200 β-method for PM measurements have not documented any systematic deviation between the two measuring methods except for an improved reproducibility and data capture using the LVS instruments.

The SM200 sampler manufactured by OPSIS, Sweden, has been used in Denmark to measure PM<sub>10</sub> in accordance with the EU Directive (EC, 2008). Measurements with this instrument have from 2007 been extended to also include PM<sub>2.5</sub>. The sampler provides the possibility for online diurnal measurements of PM in combination with sampling of PM on filters. The filters can later be used for chemical analysis. The online measurements of PM are obtained immediately after the diurnal sampling period by means of absorption of β-rays in the particles sampled on the filters. This option provides the possibility of presenting "on-line" results via the internet.

Results indicate that the β-ray results from the SM200 sampler comply better with the reference method for PM<sub>10</sub> given in the EU Directive, than the results from weighing of the filters using the SM200 as a filter sampler for PM<sub>10</sub> (Harrison, 2006). For this reason we have decided from 2006 and onwards to report results from the β-method. Previously, only results from weighing of the filters collected by the SM200 were reported. The results from the two methods differ slightly. From 2002 to 2005, where comprehensive data sets are available, it is shown that the β-method in average yields results that are 1.08 times the weighing for the yearly average and 1.09 times the weighing for the 36th highest concentration.

Since August 2012 measurements of PM<sub>10</sub> and PM<sub>2.5</sub> based on the LVS reference method (EN 12341: 2014) were introduced into the regular measuring programme to replace some of the older SM200 instruments that needed to be renewed and LVS is now installed at most of the stations in the network. Data series from stations where a single measurement method (i.e. either SM200 or LVS) does not cover a full calendar year consists of a combination of SM200 and LVS measurements.

Initiation of LVS measurements:

**2012**

HCAB PM10 (Copenhagen street): 5 September 2012

HCAB PM2.5 (Copenhagen street): 12 September 2012

Jagtvej PM10 (Copenhagen street): 28 August 2012

HCØ PM2.5 (Copenhagen urban background): 2 August 2012

Risø PM2.5 (Rural): 29 August 2012

**2013**

HCØ PM10 (Copenhagen urban background): 26 June 2013

Jagtvej PM2.5 (Copenhagen traffic): 16 November 2013

**2014**

Risø PM10 (Rural): 4 January 2014

Aarhus PM10 (street): 10 September 2014

Aarhus PM2.5 (street): 10 September 2014

Aarhus PM2.5 (urban background): 1 January 2014

Aalborg PM2.5 (street): 9 April 2014

Aalborg PM2.5 (urban background): 9 April 2014

**2015**

Hvidovre PM2.5 (Copenhagen, suburban): 17 June 2015

Measurements of particle numbers have been carried out since 2002 in cooperation between the monitoring programme and research projects financed by the Danish Environmental Protection Agency. The measurements have been carried out using a Differential Mobility Particle Sizer (DMPS) that counts particles with mobility diameter between 6 and 700 nm. In 2015 additional measurements were started at the non-permanent measurement station in Hvidovre using a Scanning Mobility Particle Sizer (SMPS) with mobility diameter between 11 and 478 nm.

## **7.1 Annual statistics**

At all the PM<sub>10</sub>- and/or PM<sub>2.5</sub> stations particulate material was collected continuously on filters on a diurnal basis for subsequent mass detection either by gravimetric determination (LVS) or by β-absorption measurement using SM200-monitors depending on the method used on the particular station (table 7.1 and 7.2). Subsequently the particle samples were analysed in the laboratory. Additionally, PM is measured at the stations in the Copenhagen area using a TEOM (Tapered-Element Oscillating Microbalance) instrument. The TEOM measurements have a time resolution of 30 minutes (table 7.3). During sampling the collected particles are heated to 50°C. At that temperature some of the volatile compounds evaporate (mainly secondary aerosols). The loss will depend of the actual composition of the aerosols. The European Commission has accepted that TEOM measurements for PM can be used in relation to EU limit values if the measured values are multiplied with a factor 1.3. However, the correction factor depends e.g. on the specific measurement site and seasonality and correction of TEOM measurements of PM using a correction factor of 1.3 do therefore have considerable uncertainty.

In 2015 the permitted number of exceedances in a year of the diurnal limit value of 50 µg/m<sup>3</sup> for PM<sub>10</sub> was not exceeded at any stations in the measuring network, even at stations where exceedances previously have occurred (the two traffic stations in Copenhagen (HACB/1103 and Jagtvej/1257)). Likewise, there were no exceedances of the annual limit value for PM<sub>10</sub> (of 40 µg/m<sup>3</sup>) and PM<sub>2.5</sub> (of 25 µg/m<sup>3</sup>) at any measuring station.

The EU-directive on air quality (EC, 2008) prescribes that the national average exposure indicator (AEI) has to be determined based on three years average of the average urban background concentration of PM<sub>2.5</sub>. For the years 2013-2015 the AEI is determined to 11 µg/m<sup>3</sup>. In Denmark the average exposure indicator is measured in urban background at Copenhagen/1259, Aarhus/6159 and Aalborg/8158.

In 2015 the number of particles in ambient air was about 12.300 particles per cm<sup>3</sup> at the street station H.C. Andersens Boulevard (table 7.5). This is a factor of roughly about 3 and 4 higher than in urban and rural background, respectively.

**Table 7.1.** Annual statistics for PM<sub>10</sub> in 2015. All parameters are given as diurnal averages at ambient temperature and pressure.

Unit µg/m <sup>3</sup>	Number of results	Average (µg/m <sup>3</sup> )	Median	Days above 50 µg/m <sup>3</sup>	90 percentile	Max. day
<b>Street</b>						
Copenhagen/1103 <sup>1</sup>	358	29	26	26	43	75
Copenhagen/1257 <sup>1</sup>	360	23	21	13	38	76
Aarhus/6153 <sup>1</sup>	347	21	18	7	31	70
Odense/9155 <sup>2</sup> §	166	21	19	3	34	66
<b>Urban background</b>						
Copenhagen/1259 <sup>1</sup>	354	18	16	4	30	64
<b>Rural</b>						
Risø <sup>1</sup>	363	17	14	3	27	61
Keldsnor/9055 <sup>2</sup>	321	20	18	7	35	62
Limit value (2005)		40		35***		
90% data capture		>328**				

<sup>1</sup> Measurements based on low volume sampling with gravimetric determination of particle mass

<sup>2</sup> Measurements based on SM200 beta gauge determination of particle mass

\* Combination of low volume sampling and SM200 beta gauge determination of particle mass

\*\* 90% data capture of number of diurnal measurements in relation to number of days in 2015

\*\*\* Permitted number of exceedances in a year of the diurnal limit value of 50 µg/m<sup>3</sup>

§ The site in Odense/9155 (Albanigade) was from 28 June 2014 affected by a major permanent rearrangement of the roads in Odense, and the site changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic. On 15 June 2015 the measurements had to be stopped. The PM<sub>10</sub> measurements from Odense/9155 (Albanigade) for 2015 thus do not represent a traffic site but rather have character of an urban background site.

**Table 7.2.** Annual statistics for PM<sub>2.5</sub> in 2015. All parameters are given as diurnal averages at ambient temperature and pressure.

Unit µg/m <sup>3</sup>	Number of results	Average (µg/m <sup>3</sup> )	Median	90 percentile	Max. day
<b>Street</b>					
Copenhagen/1103	338	16	13	27	59
Copenhagen/1257	359	14	12	26	59
Aarhus/6153	357	12	9	23	57
Aalborg/8151*					
<b>Suburban</b>					
Hvidovre/2650	192	11	8	19	57
<b>Urban background</b>					
Copenhagen/1259	338	12	9	22	54
Aarhus/6159	361	10	7	18	56
Ålborg/8158	348	9	8	17	52
<b>Rural</b>					
Risø <sup>1</sup>	356	11	8	21	54
Limit value (2015) (parenthesis gives proposed value for 2020)		25(20)			
90% data capture		>328*			

Measurements at all stations in 2015 were based on low volume sampling (LVS) with gravimetric determination of particle mass

\* No data from Aalborg/8151 (traffic site) in 2015 because the station is closed temporarily due to construction work.

\*\* 90% data capture of number of diurnal measurements in relation to number of days in 2015

**Table 7.3.** Annual statistics for PM<sub>10</sub> measured in 2015 using TEOM. The values are based on ½-hourly averages. Total annual number of ½-hours is 17520.

Unit: µg/m <sup>3</sup>	Number of results	Average	Average x 1.3
<b>Street</b>			
Copenhagen/1103	16294	29	38
<b>Rural</b>			
Risø	13841	14	18
Limit value			40

**Table 7.4.** Annual statistics for PM<sub>2.5</sub> measured in 2015 using TEOM. The values are based on ½-hourly averages. Total annual number of ½-hours is 17520.

Unit: µg/m <sup>3</sup>	Number of results	Average	Average x 1.3
<b>Street</b>			
Copenhagen/1103	15067	11	15
<b>Rural</b>			
Risø	16469	8	11
Limit value (2015) (parenthesis gives proposed value for 2020)			25(20)

**Table 7.5.** Annual statistics for particle number measured in 2015. All values are based on ½-hourly averages. Total annual number of ½-hours is 17520.

Unit: particles per cm <sup>3</sup>	Number of results	Average
<b>Street</b>		
Copenhagen/1103*	11388	12310
<b>Urban Background</b>		
Copenhagen/1259*	11563	3990
Hvidovre/2650**	4030§	3570
<b>Rural</b>		
Risø*	12614	2960

\* Measured with DMPS (6nm – 700 nm)

\*\* Measured with SMPS (11nm – 478 nm)

§ SMPS measurements at Hvidovre began 1 October 2015.

DMPS and SMPS measure slightly different size ranges of particles. It is estimated that less than 10% of the particles are in the size range from 6nm – 10nm plus 479nm – 700nm. The underestimation of the SMPS measurements due to the slightly narrower size range compared to the DMPS measurements is therefore estimated to be less than 10%.

## 7.2 Trends

Up to the year 2000 the particulate matter was measured as Total Suspended Particulate matter (TSP) corresponding to particles with a diameter up to around 25 µm (figure 7.1). The exact cut-off depends strongly on the wind velocity. From 2001 most of the measurements of particulate matter were changed from TSP to PM<sub>10</sub> according to the EU directive adopted in 1999 (EC, 1999) and PM<sub>10</sub> measurements were started at all stations except Copenhagen/1103 where the TSP measurements were continued to the end of 2005. The TSP is on the average 30-80% higher than PM<sub>10</sub> at the street stations, while the difference is less at urban background and rural sites.

The measurements show a tendency for a decrease in PM<sub>10</sub> at all the measurement stations since 2001, where the measurements began (figure 7.2). Although the measurements at HCAB (Copenhagen/1103) began later, there is also a decrease in PM<sub>10</sub> at this station. However, this is mainly due to a major reduction (7 µg/m<sup>3</sup>) in PM<sub>10</sub> from 2008 to 2009. Detailed examination of all the measurements at HCAB showed that the main reason for this decrease from 2008 to 2009 was new asphalt surface on the road laid out during August and September 2008 (Ellermann et al., 2010) that significantly reduced dust generation from road abrasion.

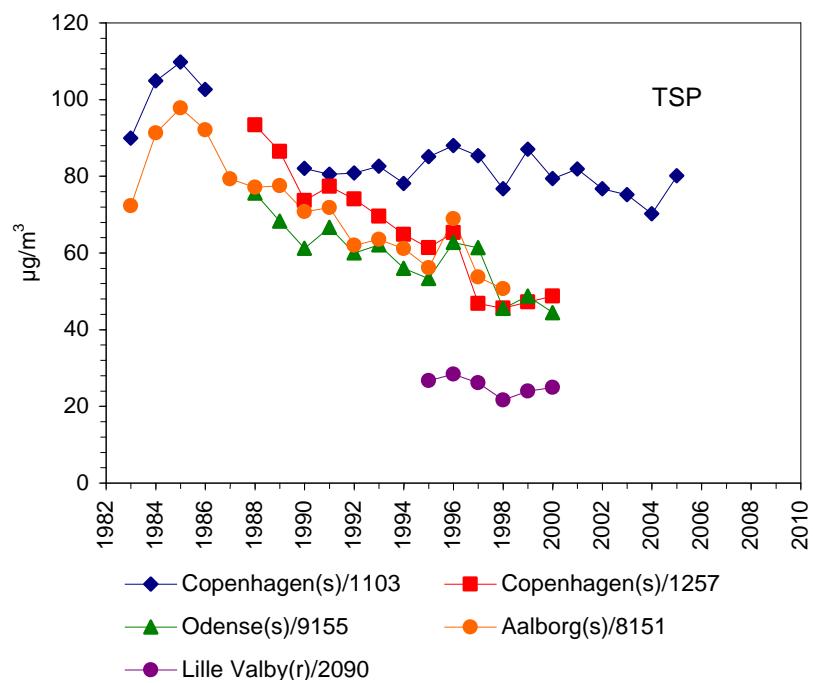
The site in Odense/9155 (Albanigade) was affected by a major permanent rearrangement of the roads in Odense. It changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on 28 June 2014. This has affected the measured PM<sub>10</sub> levels in the second half of 2014 and this is the reason why there is unchanged PM<sub>10</sub> value for Odense/9155 in 2014 while all the other traffic stations display an increase in 2014 compared to 2013. In 2015 the road next to the measuring station was closed for traffic. PM<sub>10</sub> measurements from Odense/9155 (Albanigade) for 2015 do not represent a traffic site but rather have character of an urban background site.

The measurements of PM<sub>2.5</sub> started in 2007 at Copenhagen/1103 and at the other stations in 2008. Figure 7.3 presents all the results from diurnal meas-

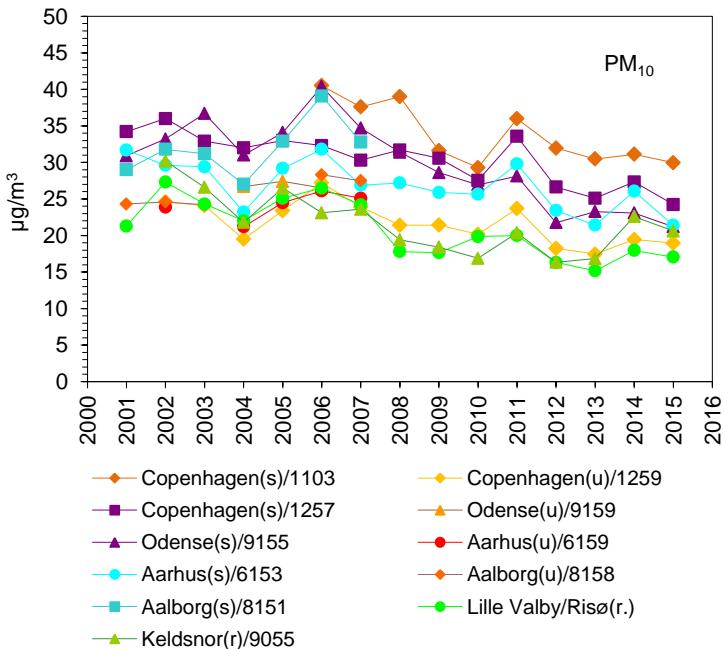
urements of PM<sub>2.5</sub> until now . There seems to be a tendency to a small reduction in PM<sub>2.5</sub>, although this tendency is uncertain due to the relatively short period with measurements.

The AEI (the average exposure index for PM<sub>2.5</sub>) is determined as the average PM<sub>2.5</sub> measured at urban background in Copenhagen, Aarhus and Aalborg averaged over a three year period. The trend for AEI is shown in figure 7.5 and as seen for PM<sub>2.5</sub> itself, there is a small reduction in the AEI, although this tendency is uncertain due to the relatively short period with measurements and the large interannually variation in PM<sub>2.5</sub> due to the natural variations in the meteorological conditions.

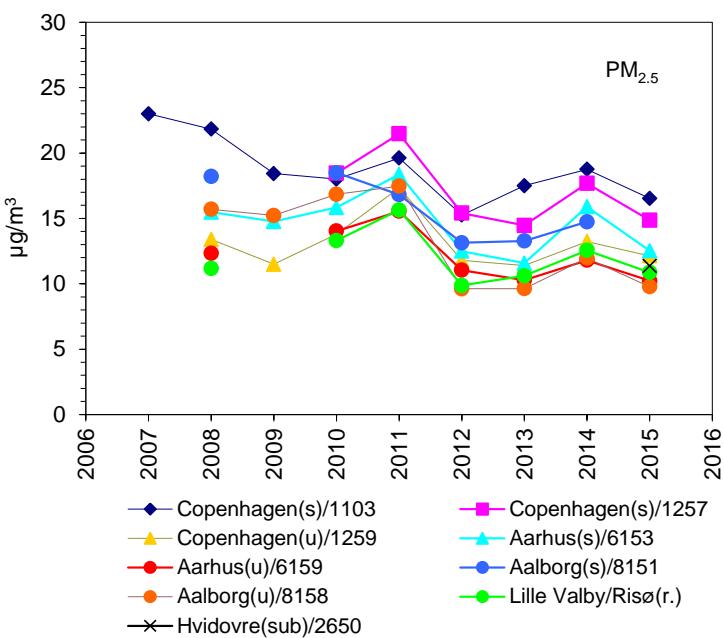
The measurements show a significant reduction of particle number concentration in ambient air over the entire measuring period (figure 7.5). On HCAB the number of particles has decreased about 50% during the period 2002-2015. At the urban background station (HCØ) particle numbers show a similar decrease of about 50% for the same period. At the rural background station (LVBY/Risø) over the period 2005-2015 a reduction in particle numbers was also observed, though the decrease is only about 30%.



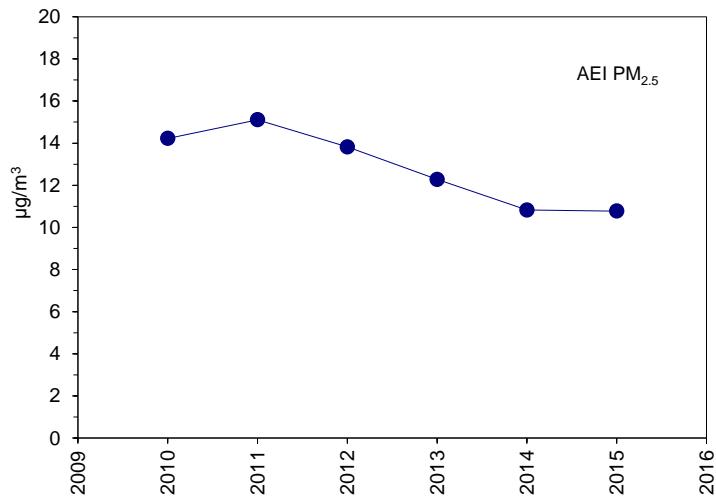
**Figure 7.1.** Annual averages for TSP measured at street stations (s) and at rural background station (r).



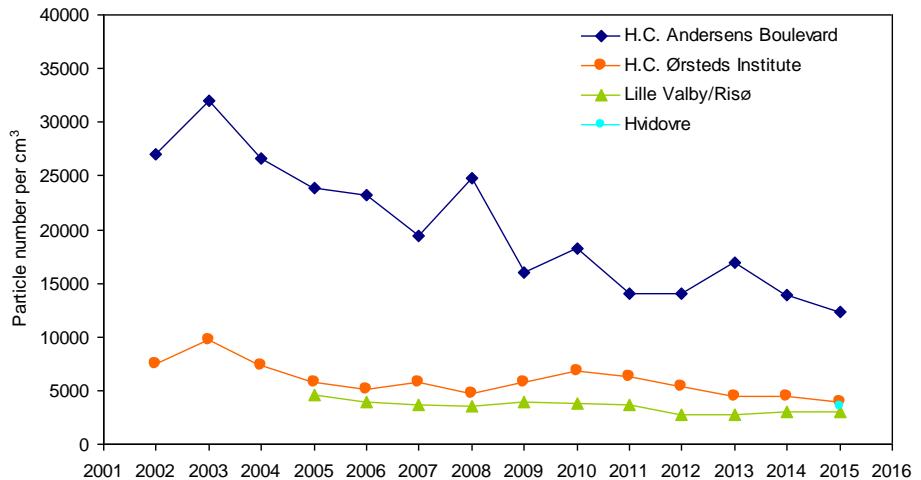
**Figure 7.2.** Annual averages for PM<sub>10</sub> measured at street stations (s), urban background stations (u) and at rural background stations (r). The change from gravimetric determination using the SM200 as a filter sampler to the use of the same instrument as a  $\beta$ -gauge from 2006 gives rise to a 5-10% increase due to the shift of method. The value for PM<sub>10</sub> at Copenhagen/1103 in 2008 and 2009 is based on the measurements with SM200 in combination with an estimated value. Data are given at standard temperature- and pressure conditions (0°C and 1 atm.). PM given at ambient temperature and pressure conditions is on an annual average approximately 3-4% lower than PM-results given at standard conditions. The site in Odense/9155 (Albanigade) was from 28 June 2014 affected by a major permanent rearrangement of the roads in Odense, and the site changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic. In 2015 the street was closed for traffic. The large change in PM<sub>10</sub> since 2013 is due to this change in traffic.



**Figure 7.3.** Annual averages for PM<sub>2.5</sub> measured at street stations (s), suburban (sub), urban background stations (u) and at rural background station (r). Only annual averages covering more than 2/3 of the years are shown except for the newly established suburban station at Hvidovre (began in 17 June 2015) and Aalborg(s) for 2014 (data covering the period 1/1 - 7/9). Data are given at standard temperature- and pressure conditions (0°C and 1 atm.). PM given at ambient temperature and pressure conditions is on an annual average approximately 3-4% lower than PM-results given at standard conditions.



Figur 7.4. The trend for AEI (the average exposure index for PM<sub>2.5</sub>). AEI is determined as the average PM<sub>2.5</sub> measured at urban background in Copenhagen, Aarhus and Aalborg averaged over a three year period. The value shown for 2010 corresponds to the average of the concentrations for 2008 to 2010 and likewise for the other years.



**Figure 7.5.** Annual averages for particle number at street level (H. C. Andersens Boulevard), suburban (Hvidovre), urban background (H.C. Ørsted Institute) and rural background (Lille Valby/Risø). It should be noted that the Hvidovre measurements started 1 October 2015 and thus only covers the period from 1 October – 31 December.

### 7.3 Impact of salt from winter salting and sea

The EU air quality directive (EC, 2008) gives the member states the possibility to compensate for the impact of salt from sea salt and winter salting on PM<sub>10</sub> (Article 20 and 21). Salt from sea salt can be subtracted from PM<sub>10</sub> prior to comparison with the limit values. If the limit values are exceeded due to winter salting then the member states do not have to prepare an air quality plan in order to reduce the levels of PM<sub>10</sub>. These rules account for both the annual limit value and the daily limit value that states that the daily PM<sub>10</sub> concentration must not exceed 50 µg/m<sup>3</sup> more than 35 days in a calendar year.

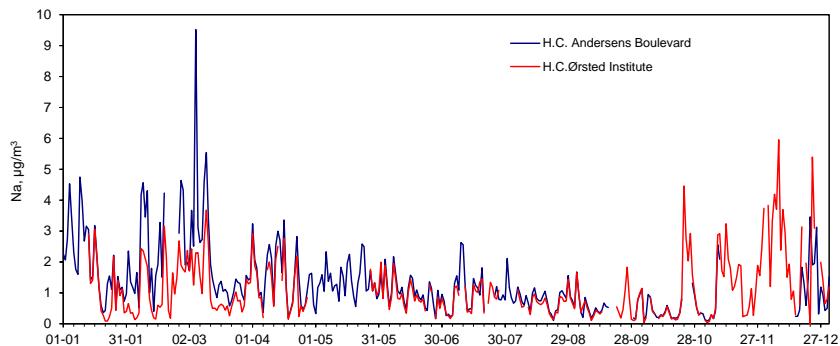
On this background the monitoring program was expanded in 2010 with daily sampling and analysis of sodium at the street stations H.C. Andersens Boulevard, Copenhagen (1103), Odense (9155) and Aarhus (6153) and at the urban background station in Copenhagen (H.C. Ørsted Institute/1259). Table 7.6 gives the annual average concentrations for sodium and estimate for total salt (NaCl) in 2013 (calculated from the measured sodium concentration).

**Table 7.6.** Annual statistics for sodium and estimate of total salt (NaCl) in 2015.

	Na $\mu\text{g}/\text{m}^3$	NaCl $\mu\text{g}/\text{m}^3$
<b>Street:</b>		
Copenhagen/1103	1.3	3.4
Odense/9155*	1.8	4.5
Aarhus/6153	1.6	4.0
<b>Urban Background:</b>		
Copenhagen/1259	1.1	2.8

\*) The site in Odense/9155 (Albanigade) was from 28 June 2014 affected by a major permanent rearrangement of the roads in Odense, and the site changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic. On 15 June 2015 the measurements had to be stopped.

Figure 7.6 shows the results from measurements of sodium at the street station H.C. Andersen's Boulevard, Copenhagen (1103) and at urban background in Copenhagen (H.C. Ørsted Institute/1259). The high concentrations at the street station during the winter months are due to winter salting of the roads. The high correlation between the sodium concentrations for the main part of the remaining year is due to long range transport of sea salt that have equal impact on the two stations.



**Figure 7.6.** Diurnal concentrations in 2015 of sodium at H.C. Andersens Boulevard, Copenhagen (1103) and at the urban background in Copenhagen (H.C. Ørsted Institute/1259).

In 2015 the permitted number of exceedances in a year of the diurnal limit value of  $50 \mu\text{g}/\text{m}^3$  for  $\text{PM}_{10}$  was not passed at any stations in the measuring network and therefore it has not been necessary to correct  $\text{PM}_{10}$  for the content of NaCl due to sea salt and winter salting of the roads.

## 8 Heavy Metals

Heavy metals in PM<sub>10</sub> is measured by collection of PM<sub>10</sub> on filters that are analysed by ICP-MS (Inductively Coupled Plasma Mass Spectrometry) for their content of elements. Results for 10 heavy metals are presented in table 8.1. Comparison between results from ICP-MS and the previously used PIXE-method (Proton Induced X-ray Emission) showed only minor changes in the annual averages, when the low concentration levels are taken in to account.

The table presents also results for analysis of heavy metals in total suspended particulate (TSP) at the measurement station Risø. The content of these heavy metals in PM<sub>10</sub> and TSP are approximately equal since these metals are mainly found in the fine particle fraction.

The ICP-MS analysis provides the measurements of Arsenic (As), Chromium (Cr) and Nickel (Ni) included in the EU Directive 2004/107/EC (EC, 2005) and Lead (Pb) included in EU Directive 2008/50/EC (EC, 2008). According to the directive (EC, 2005) also Mercury (Hg) has to be measured, however, these measurements can be carried out in cooperation with neighboring countries. As part of a bilateral agreement "Development of the mutual partnership on air pollution" between Denmark and Sweden, it has been agreed that the Swedish measurements at Röå (table 8.2) can fulfil the Danish obligations on measurements of Hg. This agreement is based on the fact that the spatial variation of background Hg concentrations is small.

### 8.1 Annual statistics

The annual statistics for the selected heavy metals are shown in table 8.1 and 8.2. The concentrations are low for all of the heavy metals and there were no exceedances of the target/limit values for the four metals (As, Cd, Ni, and Pb) that are regulated by use of target/limit values (EC, 2005, 2008).

**Table 8.1.** Annual statistics for Vanadium (V), Chromium (Cr), Manganese (Mn), Nickel (Ni), Copper (Cu), Zink (Zn), Arsenic (As), Selenium (Se), Cadmium (Cd) and Lead (Pb) measured in PM<sub>10</sub> during 2015. For comparison the table includes also results for these heavy metals measured in total suspended particulate (TSP) at the rural background station Risø.

Unit ng/m <sup>3</sup>	V	Cr	Mn	Ni	Cu	Zn	As	Se	Cd	Pb
<b>PM<sub>10</sub>, Street</b>										
Copenhagen/1103	1.3	8.0	22	2.1	72	37	0.9	0.6	0.08	3.5
Odense/9155***	0.6	1.0	6.8	0.8	12	17	0.5	0.5	0.07	2.1
Aarhus/6153	1.0	2.8	7.1	2.6	30	22	0.5	0.5	0.07	2.3
<b>PM<sub>10</sub>, Urban background:</b>										
Copenhagen/1259	1.0	0.9	5.0	1.1	11	12	0.5	0.5	0.07	2.9
<b>TSP, Rural Background</b>										
Risø	0.6	0.7	3.0	0.5	2.5	7.4	0.6	0.5	0.06	1.7
EU Target (Limit) Values *					20		6		5	500
Guideline value (WHO)**	1000		150						5	
Life time risk level at 1:10 <sup>5</sup>				25			6.6			

\*) Target values for Ni, As and Cd are implemented through EU Council Directive 2004/107/EC (EC, 2005). The limit value for Pb is found in EU Directive 2008/50/EC (EC, 2008).

\*\*) The guidelines and life time risk for the carcinogenic metals are established by WHO (WHO, 2000). The lifetime risk level is defined as the concentration that through a lifelong exposure is estimated to give an excess risk of 1:105 for developing cancer.

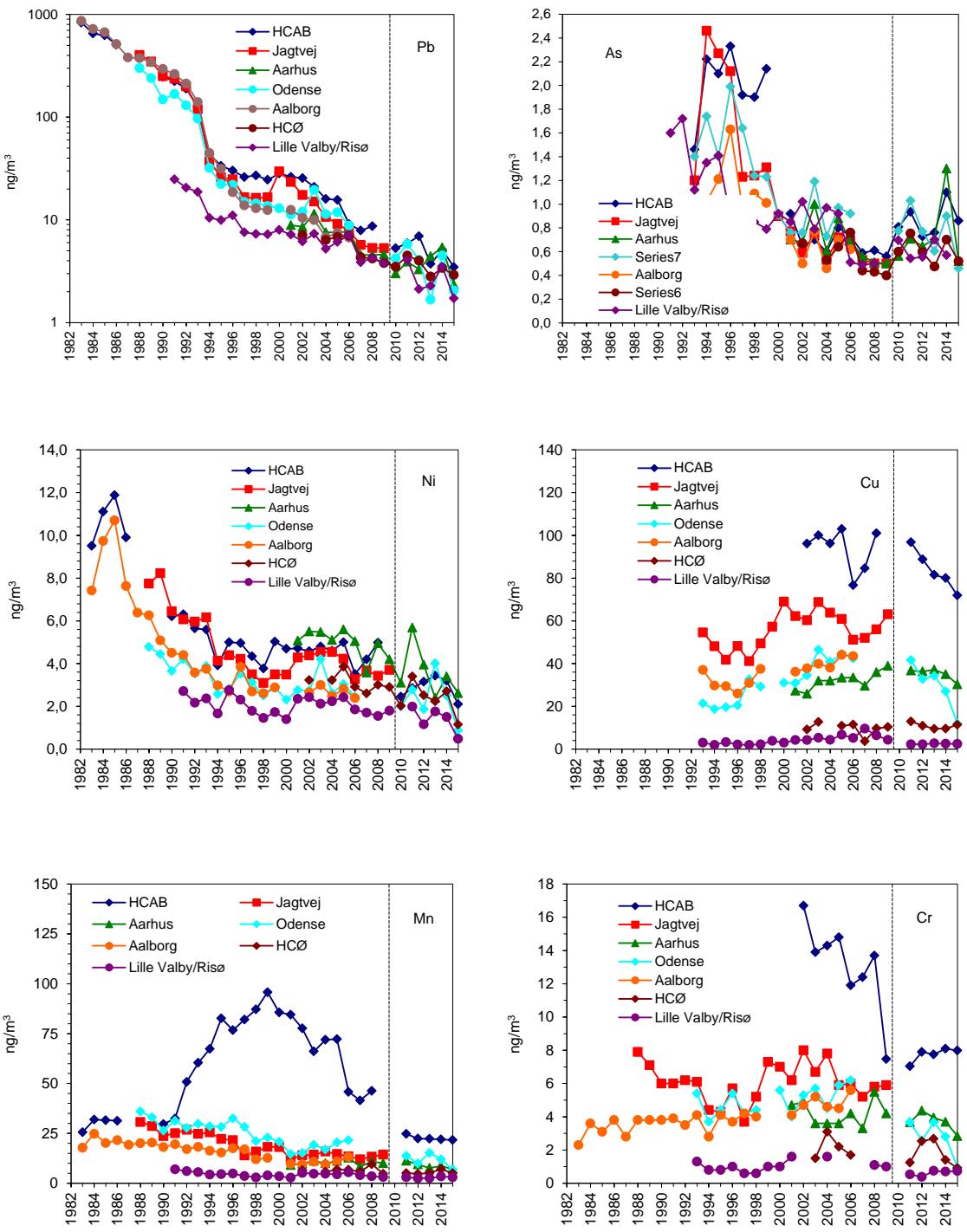
\*\*\*) The site in Odense/9155 (Albanigade) was from 28 June 2014 affected by a major permanent rearrangement of the roads in Odense, and the site changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic. On 15 June 2015 the measurements had to be stopped.

**Table 8.2.** Annual statistics for Mercury 2015. Measured at Råö in southern Sweden by the Swedish Environmental Research Institute.

Unit: ng/m <sup>3</sup>	Total Gas Hg (ng/m <sup>3</sup> )	Total Particles Hg (ng/m <sup>3</sup> )
Råö (SE00014)	1.4	0.003

## 8.2 Trends

The long term trends for six of the heavy metals are shown in figure 8.1. For Pb, As, Ni and Manganese (Mn) there are clear reductions in the concentrations due to national and international regulations of the emissions. Most pronounced for Pb where removal of Pb from gasoline has resulted in large reductions of the concentrations. For Mn the long term trend at HCAB deviates from the other stations. This is believed to be due to high Mn concentrations in the asphalt used at HCAB during the period from 1991 to 2008. The concentration of Cu increases mainly due to increased use of Copper (Cu) in brakes.



**Figure 8.1.** Annual averages from selected stations for some heavy metals in particulate matter. Until 2000 in TSP and later in PM<sub>10</sub> – except for Copenhagen/1103 where PM<sub>10</sub> replaced TSP from the beginning of 2006. The heavy metals are usually found in fine particles, which make the TSP and the PM<sub>10</sub> values comparable. Note that the scale for Pb is logarithmic. The dashed line indicate that the analysis method has been changed from 2009 to 2010.

## 9 Sulphur dioxide

The concentration of sulphur dioxide has reached very low levels in Denmark and it is therefore only necessary with a limited monitoring of the concentrations of sulphur dioxide; both with respect to the number of stations and the quality of the measurements. Hence it is only measured at two traffic stations (Copenhagen and Aalborg) with focus on episodes with high concentrations of sulphur dioxide. It is measured using gas monitors based on ultraviolet fluorescence. The concentrations of sulphur dioxide are often below the detection limit of the instruments and hence the uncertainties of the measurements are large. The concentrations are measured continuously throughout the year with a time resolution on minute scale that is aggregated to hourly averages for this reporting.

### 9.1 Annual statistics

The annual statistics for 2014 for sulphur dioxide are shown in table 9.1. None of the limit values (EU, 2008) were exceeded in 2015. In 2015 there was no information to the public due to exceedance of the alert threshold for SO<sub>2</sub> (one hour average 500 µg/m<sup>3</sup>).

**Table 9.1.** Annual statistics for SO<sub>2</sub> in 2015. All parameters are calculated based on hourly average. The detection limit for the monitors is a few µg/m<sup>3</sup>, which makes the average and median values encumbered with high relative uncertainties.

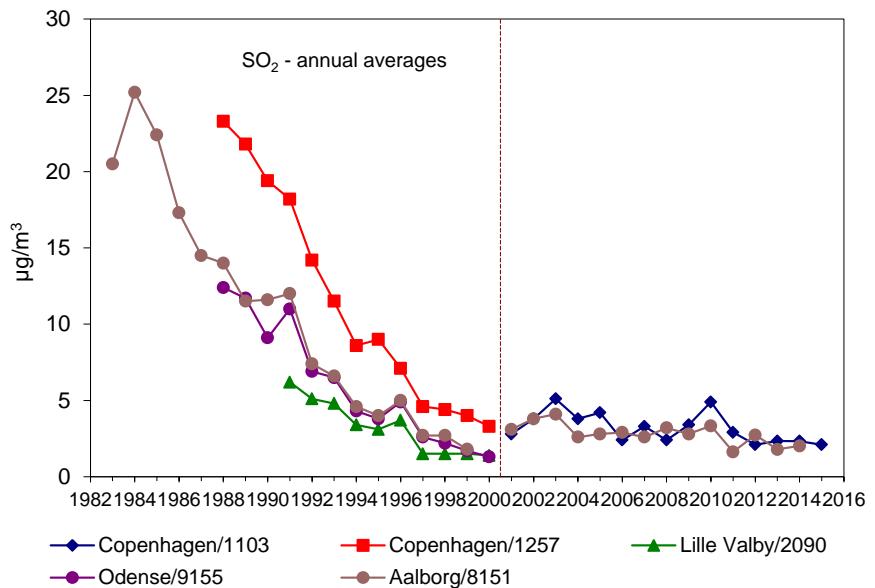
Unit: µg/m <sup>3</sup>	Number of results	Average year	Average winter	Median	98-percentile	Max. Hour	4th highest diurnal mean
<b>Traffic:</b>							
Copenhagen/1103	8032	2.1	2.4	1.6	7.4	17.0	6.9
Aalborg/8151 §	0	-	-	-	-	-	-
Limit values	>7467*	20	20			350	125

\*) 90% data capture of number of hourly measurements in relation to total number of hourly measurements in 2015 excluding hours used for calibration.

§) Aalborg/8151 (traffic) there is no data since the station has been shut down due to construction work at the site. It has not yet been possible to reinitiate the measurements at the street station in Aalborg.

## 9.2 Trends

The long term trends for sulphur dioxide are shown in figure 9.1. Since the beginning of the 1980s the annual concentrations have decreased by more than a factor of five due to effective national and international regulations of the emissions. The emission reductions are due to use of effective cleaning technologies in combination with decrease of the sulphur content in fuel.



**Figure 9.1.** Annual averages for SO<sub>2</sub>. Until 2001 the results were obtained using KOH impregnated filters for collection of SO<sub>2</sub>. These measurements ceased in 2000. After 2000 the SO<sub>2</sub> measurements have been carried out using SO<sub>2</sub> monitors in order to monitor episodic results. The detection limit for the monitors is a few µg/m<sup>3</sup>, which makes the average and median values encumbered with high relative uncertainties. The shift in level from 2000 to 2001 is due to shift of the methods. The station in Aalborg (traffic) has temporarily been shut down due to construction work at the site. There is therefore no data from Aalborg in 2015.

## 10 Polyaromatic Hydrocarbons (PAHs)

Following the EU Directive 2004/107/EC (EC, 2005), measurements of atmospheric concentrations of benzo[a]pyrene and other particle bound PAHs have been introduced in the air quality monitoring programme starting from June 2007. The target value for benzo[a]pyrene in ambient air is set to 1 ng/m<sup>3</sup> averaged over a calendar year (EC, 2005). Benzo[a]pyrene is used as a marker for the carcinogenicity of PAHs.

Particulate matter (PM<sub>10</sub> fraction) is collected at the urban station of H.C. Andersens Boulevard (Copenhagen/1103) in Copenhagen and at a temporary station in a suburban area in Hvidovre. PM is collected by high volume sampling (HVS) at a flow rate of 0.5 m<sup>3</sup> min<sup>-1</sup> over a period of 24 hours for an average total volume of 700 m<sup>3</sup>. The filters are kept frozen until analysis. Weekly based PAH concentrations are obtained by analysis of pooled fractions of daily collected samples. For each day 4 x 1.5 cm<sup>2</sup> are taken from the filter and the fractions from the whole week are pooled and extracted. The pooled filters are extracted with dichloromethane and cleaned up on silica. Before extraction, the filters are spiked with deuterium-labeled PAH. Analysis of the extracts is carried out by gas chromatography-mass spectrometry (GC-MS). Concentrations of individual PAHs in samples are corrected for recovery of a deuterium-labelled PAH standard with the closest molecular weight. A total of 18 PAHs are analysed with the method.

### 10.1 Annual Statistics

The average concentration of benzo[a]pyrene in 2015 was 0.29 ng/m<sup>3</sup> and 0.25 ng/m<sup>3</sup> at the street station on HCAB and the suburban station in Hvidovre, respectively. It can therefore be concluded that the target value for benzo[a]pyrene on 1 ng/m<sup>3</sup> was not exceeded in 2015.

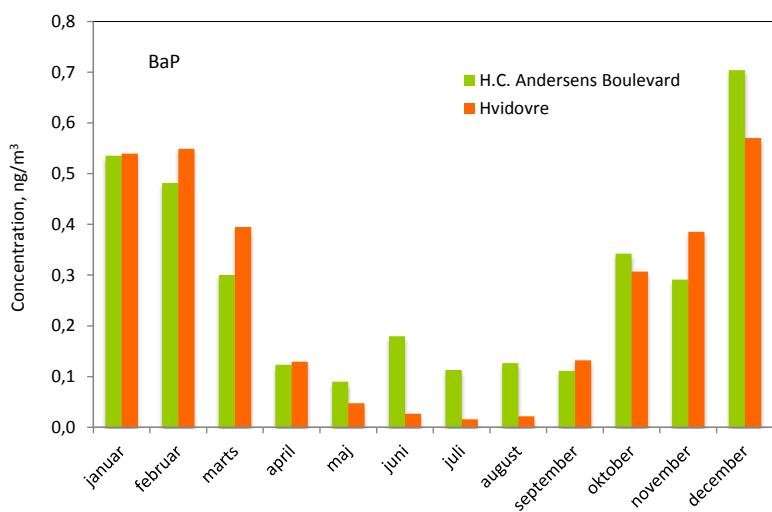
In 2015 the concentrations were highest at HCAB, whereas Hvidovre had the highest concentrations in 2014. The difference from year to year shows that the variations in the concentrations can be large. This is thought to be due to variations in the number and magnitude of local episodes with large concentrations of benzo[a]pyrene created by the meteorological conditions i.e. temperature inversions during cold winter mornings.

The average annual concentrations of the other five PAHs listed as relevant in the EU Directive are shown in table 10.1.

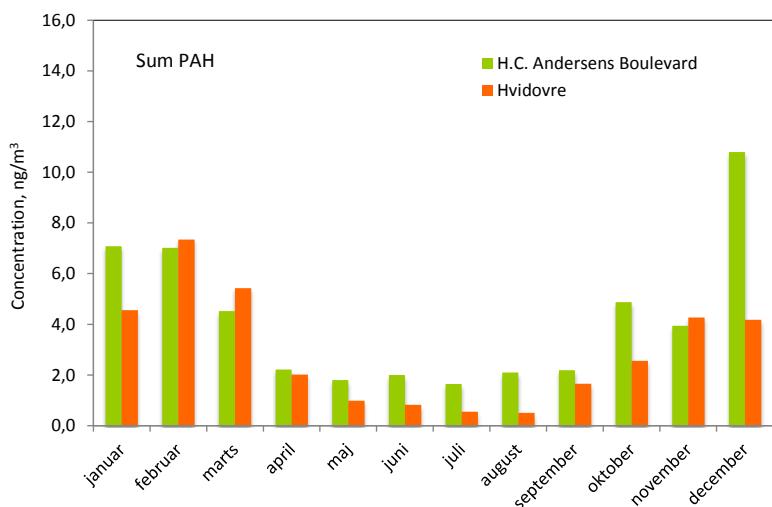
**Table 10.1.** Annual average concentrations for the six PAHs listed in the EU Directive.

	HCAB ng/m <sup>3</sup>	Hvidovre ng/m <sup>3</sup>
Benzo[a]pyrene	0.29	0.25
Benzo[a]atracene	0.19	0.17
Benzo[b]fluoranthene	0.42	0.35
Benzo[j+k]fluoranthenes	0.40	0.36
Indeno[1,2,3-cd]pyrene	0.23	0.20
Dibenzo[a,h]anthracene	0.03	0.10

The seasonal trends in PAH concentrations are summarized in figure 10.1 and 10.2. As expected, the atmospheric concentrations are low during summer months, while concentrations increase in winter months due to higher emissions and less photochemical degradation of the compounds. The seasonal variation seems also to vary between the two measurements stations (table 10.2). The winter concentrations at Hvidovre are higher than at HCAB in 2013-2015 while the summer concentrations are at the same level in 2013 and 2014 and lower at Hvidovre than at HCAB in 2015. This is because the sources of benzon[a]pyrene in Hvidovre is largely wood burning for house hold heating while the sources at HCAB are both wood burning and traffic. The seasonal variation in the emissions for traffic is small compared to that of wood burning.



**Figure 10.1.** Monthly average concentrations of benzo[a]pyrene at H.C. Andersens Boulevard and Hvidovre in 2015.



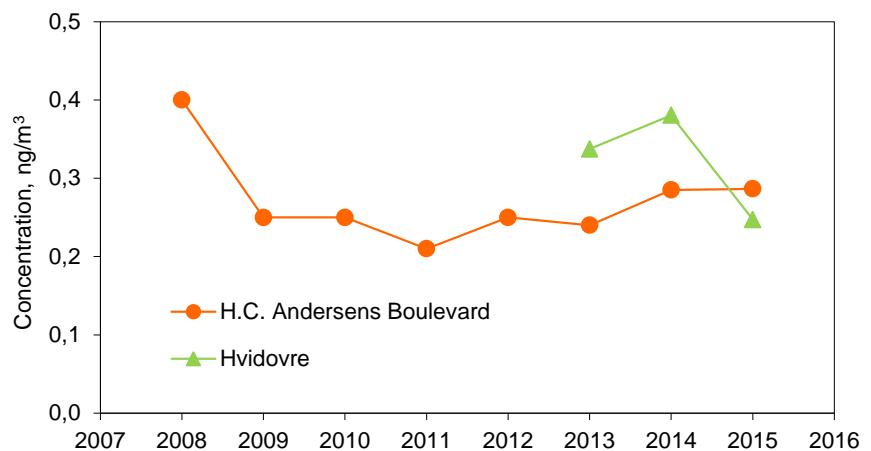
**Figure 10.2.** Monthly average concentrations of the sum of all analyzed PAH at H.C. Andersens Boulevard and Hvidovre in 2015.

**Tabel 10.2.** Winter, summer and annual average concentrations of benzo[a]pyrene for 2013-2015.

	Hvidovre			HCAB		
	2013	2014	2015	2013	2014	2015
Winter	0.53	0.73	0.46	0.38	0.50	0.44
Summer	0.12	0.10	0.06	0.11	0.10	0.12
Annual	0.34	0.38	0.25	0.24	0.29	0.29

## 10.2 Trends

The annual averages of benzo[a]pyrene since 2008 at the street station on HCAB are shown in figure 10.3. A slight decrease in the annual averages of benzo[a]pyrene is observed since 2008, however, longer time series are needed in order to show whether or not this tendency is persistent.



**Figure 10.3.** Annual average concentrations of benzo[a]pyrene at H.C. Andersens Boulevard and Hvidovre.

# 11 Organic carbon and elemental carbon

Ambient concentrations of particulate organic carbon (OC) and elemental carbon (EC) are measured on the kerbside station H.C. Andersens Boulevard/1103 in Copenhagen and the rural background station Risø north of Roskilde. EC is additionally measured on H.C. Ørsted Instituttet /1259 (Urban background, Copenhagen) and at Fjeldstedvej (suburban site, Hvidovre), which is considered to be a hot spot for residential wood burning. PM<sub>2.5</sub> particulate matter is sampled on two filters in tandem, i.e. quartz-behind-quartz, to correct for positive artifacts from adsorption of volatile and semivolatile organic compounds, which are not particulate material. The filters are analyzed for OC and EC by a thermal/optical method according to the European EUSAAR2 temperature protocol (Cavalli et al., 2010).

## 11.1 Annual statistics and trends

Organic Carbon (OC) and Elemental Carbon (EC) have been measured in PM<sub>2.5</sub> since 2010. During this relatively short period, the annual averages of OC in rural background has oscillated between 1.2 and 1.8 µg/m<sup>3</sup>. Since biogenic sources are expected to account for the majority of the OC in PM<sub>2.5</sub> a constant trend impacted by natural variation is expected. OC at the kerbside station HCAB co-varies with rural background with an almost constant increment corresponding to the traffic source (Figure 11.1). On the contrary, EC in rural background (0.22 µg/m<sup>3</sup>) has decreased to 50% of its 2010 concentration in 2015. The kerbside station (1.6 µg/m<sup>3</sup>), which is largely impacted by local traffic, has experienced a 34% decrease in EC. In 2015, urban background (0.38 µg/m<sup>3</sup>) and the suburban site (0.60 µg/m<sup>3</sup>) experienced EC concentrations at intermediate values. The ratio of EC to total carbon (TC) differs markedly between rural background (0.15) and the kerbside station in Copenhagen (0.41), however both ratios have decreased from 2010 to 2015 as expected (Figure 11.1).

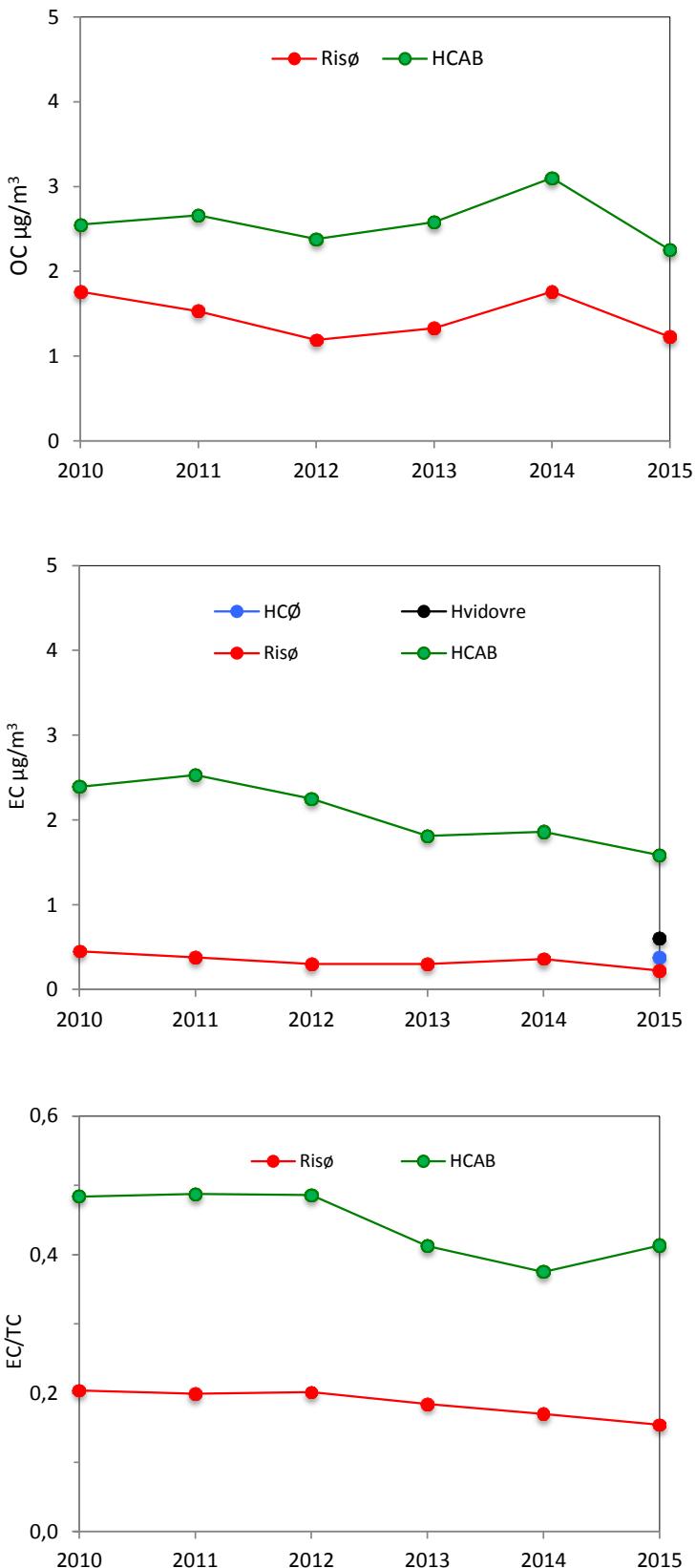
A clear seasonal pattern was observed for EC and OC at the rural and urban background with minimum summer concentrations and higher winter concentrations. EC and OC showed only minor seasonal variation at the kerbside station.

**Table 11.1.** Annual statistics for OC in 2015. The values are based on daily averages at H. C. Andersens Boulevard and in semi-rural background.

Concentration µg/m <sup>3</sup>	Data capture	OC, average.	90% percentile
Copenhagen/1103	98%	2.3	3.9
Risø	93%	1.2	2.5

**Table 11.2.** Annual statistics for EC in 2015. The values are based on daily averages at H.C. Andersens Boulevard and in semi-rural background.

Concentration µg/m <sup>3</sup>	Data capture	EC, average.	90% percentile
Copenhagen/1103	98%	1.6	2.6
Copenhagen/1259	93%	0.38	0.66
Risø	93%	0.22	0.45
Hvidovre	18%	0.60	1.2



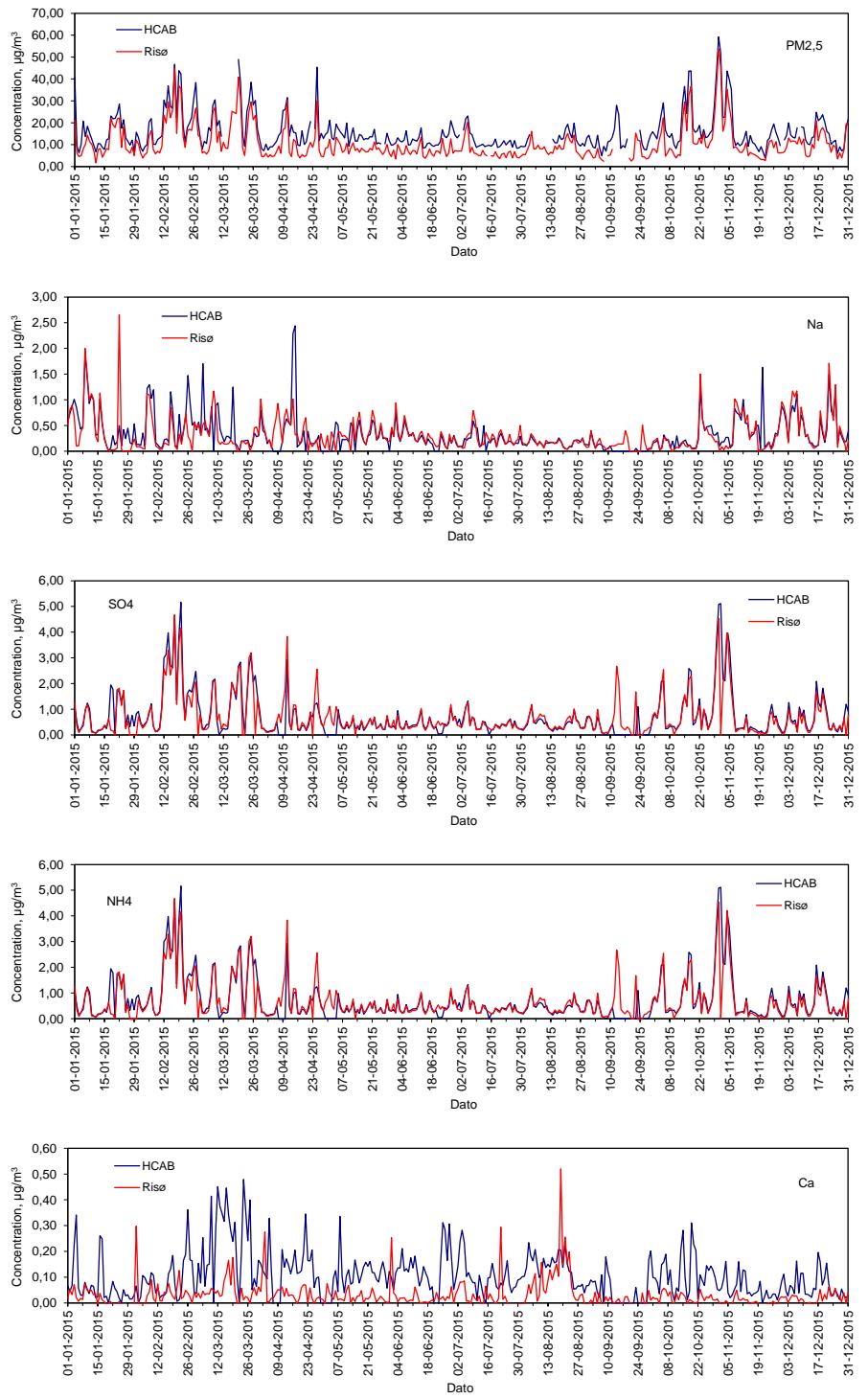
**Figure 11.1.** Organic Carbon (OC), Elemental Carbon (EC) and the ratio of EC to total carbon (EC/TC) at street (HCAB), semi-rural background (Risø), urban background (HCØ) and the suburban site for residential wood combustion (Hvidovre).

## 12 Chemical composition of PM<sub>2.5</sub>

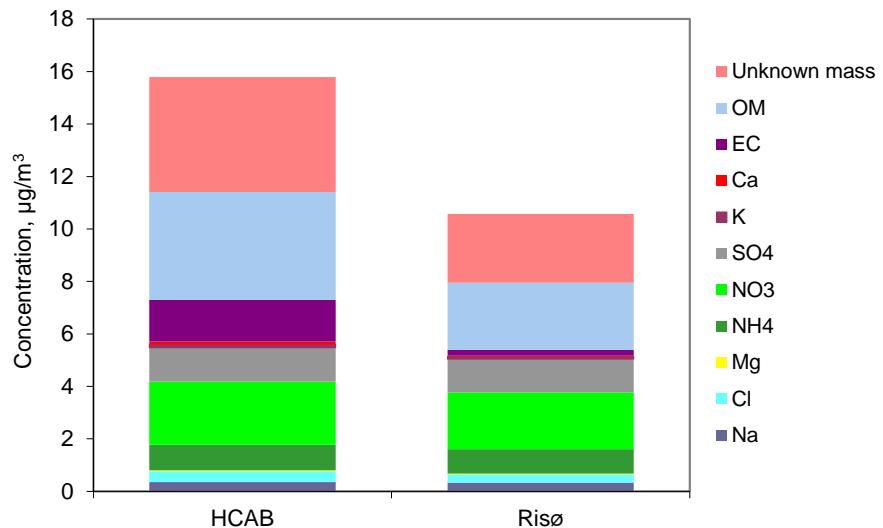
In addition to the measurements of elemental and organic compounds, also measurements of the main inorganic compounds in PM<sub>2.5</sub> ( $\text{NH}_4^+$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ) have been conducted at H.C. Andersen's Boulevard (HCAB, Copenhagen/1103) and Risø. These measurements are carried out on the basis of the air quality directive from 2008 (EC, 2008). The method is chemical analysis of the daily PM<sub>2.5</sub> particle filters sampled using the SM200 monitors.

Examples of the daily variations of the concentrations are shown in figure 12.1 together with the variation of PM<sub>2.5</sub>. For  $\text{Na}^+$  the annual average concentrations are similar at HCAB and Risø due to long range transport of sea salt. For the winter months  $\text{Na}^+$  is higher at HCAB than Risø due to winter salting of the roads in Copenhagen. The variations of  $\text{Cl}^-$  follow the variations of  $\text{Na}^+$  because the main source is sea salt and winter salting.  $\text{Mg}^{2+}$  originates only from sea salt and there are therefore similar concentrations at the two stations throughout the year.  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  originate mainly from long range transport and there are therefore only minor differences between the two stations (figure 12.1). This is also the case for  $\text{NO}_3^-$  and  $\text{K}^+$ .  $\text{Ca}^{2+}$  is in general higher at HCAB than at Risø. This is due to road dust at HCAB since asphalt contains large quantities of calcium.

The annual contributions to PM<sub>2.5</sub> of the different compounds are shown in figure 12.2. The annual average concentrations of  $\text{NH}_4^+$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  are very similar at the two stations, just as the daily variation. The main variations between the two stations are for EC, OM (organic matter) and  $\text{Ca}^{2+}$  where the concentrations are higher at the street station compared to the rural background station. This is mainly due to emissions of these compounds from the traffic in Copenhagen. As in the previous year, the unknown mass is higher at HCAB than at Risø. The mass of the unknown is very uncertain because it is calculated from the difference between PM<sub>2.5</sub> and the sum of all the analysed constituents. The unknown mass is water attached to the particles, dust (e.g.  $\text{SiO}_2$ ), heavy metals and other trace constituents.



**Figure 12.1.** Daily variations of the concentrations of PM<sub>2.5</sub>, Na<sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup> and Ca<sup>2+</sup> at H.C. Andersens Boulevard (HCAB, Copenhagen/1103) and Risø in 2015. The large gap in data for calcium on Risø is due to problems with filter material.



**Figure 12.2.** Annual average contributions to the chemical composition of PM<sub>2.5</sub> at H.C. Andersens Boulevard (HCAB, Copenhagen/1103) and Risø in 2015. Organic matter (OM) has been estimated from the measured concentrations of OC (organic carbon) by multiplication of OC with a factor of 1.5 for the fresh OM at HCAB and 2.1 for the aged OM at HCAB and Risø, respectively (Turpin and Lim, 2001). This is in order to account for the contribution of hydrogen, oxygen, nitrogen etc. to the mass of the organic compounds.

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## Appendix 1

### Pollutants measured in the network

NO and partly NO<sub>2</sub> are formed by combustion at high temperatures. The main sources are power plants and traffic. At the street stations the traffic is the main source. The application of catalytic converter in the exhaust reduces the emission considerably. NO is relatively harmless, but NO<sub>2</sub> can cause respiratory problems.

Most of the NO<sub>2</sub> in the urban atmosphere is produced by oxidation of nitrogen monoxide (NO) by ozone (O<sub>3</sub>). The reaction will take place immediately, if sufficient O<sub>3</sub> is present. O<sub>3</sub> is often the limiting component for a complete oxidation in the street canyons, but practically all NO is oxidised at the urban background and rural stations. Within a few hours the NO<sub>2</sub> is further oxidised to nitrate and/or nitric acid, which may cause acid precipitation and eutrophication. NO<sub>2</sub> is a toxic gas, which may cause respiratory problems. There are limit values for the allowed concentration of NO<sub>2</sub> in the atmosphere.

O<sub>3</sub> is formed by photochemical reactions (i.e. by the influence of sunlight) between nitrogen oxides and volatile organic compounds (VOC's). The VOC's can be of natural and anthropogenic origin. The major part of the O<sub>3</sub> measured in Denmark originates from sources outside the country. Usually the highest concentrations are found at rural and urban background sites. O<sub>3</sub> is removed by NO at street level. O<sub>3</sub> is a toxic gas, which may cause respiratory problems and damage on crops and forests. There are so-called target values for the concentration of O<sub>3</sub> in the atmosphere.

The main source of CO in urban air is petrol-fuelled cars. The CO is formed due to incomplete combustion. The application of catalytic converter in the exhaust reduces the emission considerably. CO is only slowly removed from the atmosphere. CO is a toxic gas that may prevent the uptake of oxygen in the blood. There are limit values for the allowed concentration of CO in the atmosphere.

Benzene is present in petrol. It may also be formed in engines due to incomplete combustion. Since 1994 the benzene content in petrol has been reduced by up to a factor of 5. The concentration in the atmosphere has been reduced correspondingly. Benzene is a carcinogenic gas. There is a limit value for the average content in the atmosphere.

Many different VOC's are present in the air. Several of these are emitted by incomplete combustion in e.g. engines and wood burning stoves. Several of the VOC's are carcinogenic. A "target value" is implemented through an EU Council Directive in 2004 for Benzo[a]-pyrene as indicator for PAH (Polycyclic Aromatic Hydrocarbons).

The main sources for PM<sub>10</sub> and PM<sub>2.5</sub> are combustion and resuspended dust. PM are also produced by chemical reactions in the atmosphere e.g. oxidation of nitrogen dioxide, sulphur dioxide and VOC. The submicron particles, which are formed by combustion and chemical reactions in the atmosphere, are suspected to be the most harmful for the health. There are still a lack of knowledge about the connection between health effects and particle size. Limit values for the PM<sub>10</sub> concentration in the atmosphere are implemented at present.

PM<sub>10</sub> and PM<sub>2.5</sub> is measured using three different methods in the monitoring program:

- The Beta method: The particles are collected on filters for 24 hours intervals. The mass on the filters is automatic determined by measurements in the instrument of  $\beta$ -absorption in the filter with sampled dust. This method is considered to be equivalent to the reference method (EN 12341:1999 and EN14907:2005).
- The LVS method: The particles are collected on filters for 24 hour intervals by a low volume sampler (LVS). The mass on the filters is subsequently determined in the laboratory by gravimetric measurements of the dust. This method is the current reference method for the determination of the PM<sub>10</sub> or PM<sub>2.5</sub> mass concentration of suspended particulate matter in ambient air (EN 12341: 2014, into which the previous standards for PM<sub>10</sub>, EN 12341: 1998, and for PM<sub>2.5</sub>, EN 14907:2005, have been merged).
- The TEOM method: The particles are continuously collected on a "tapered oscillating microbalance" (TEOM) and heated to 50°C. During heating volatile compounds may evaporate. The loss will be most pronounced for "secondary aerosols" containing ammonium nitrate. PM results are given with a time resolution as ½-hourly averages.

There are a number of different heavy metals (HM) in the atmosphere. They are emitted from e.g. coal and oil fired power plants, waste incinerators and industries. HM's may also be emitted from traffic due to wear on engines, tires and brake pads. Several HM's are toxic even in low concentrations and a few also carcinogenic. A limit value is implemented for lead. Target values are values are implemented for arsenic, cadmium, nickel and mercury. WHO has proposed guideline values for the toxic non-carcinogenic and estimated life time risks for the carcinogenic HM's.

Sulphur dioxide (SO<sub>2</sub>) is formed by burning of fossil fuel and biomass. The SO<sub>2</sub> is oxidised in the atmosphere to particulate sulphuric acid and sulphate. The conversion time depends strongly on the temperature and humidity in the air. It is typically of the order of one day. Sulphuric acid contributes to "acid rain" and the deposition of sulphate causes damage to sensitive ecosystems. Since the beginning of the 1980'thies the reduction of sulphur in fossil fuel and improved flue gas cleaning has reduced the concentration of SO<sub>2</sub> with one order of magnitude. SO<sub>2</sub> may cause respiratory problems. There are limit values for the allowed concentration of SO<sub>2</sub> in the atmosphere.

# THE DANISH AIR QUALITY MONITORING PROGRAMME

Annual Summary for 2015

The air quality in Danish cities has been monitored continuously since 1981 within the Danish Air Quality Monitoring network. The aim is to follow the concentration levels of toxic pollutants in the urban atmosphere and to provide the necessary knowledge to assess the trends, to perform source apportionment, and to understand the governing processes that determine the level of air pollution in Denmark. In 2015 the air quality was measured in four Danish cities and at two background sites. In addition model calculations were carried out to supplement the measurements. At one street station (H.C. Andersens Boulevard) in Copenhagen NO<sub>2</sub> was found in concentrations above the EU limit value while NO<sub>2</sub> levels in Odense, Aarhus and Aalborg were below the limit value. Model calculations indicate exceedances of the NO<sub>2</sub> limit value at several streets in Copenhagen. Annual averages of PM10 and PM2.5 were below limit values at all stations and the average exposure indicator (PM2.5 in urban background) has decreased with about 20% since 2010. The concentrations for most pollutants have been decreasing during the last decades.

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