THE DANISH AIR QUALITY MONITORING PROGRAMME

Annual Summary for 2014

Scientific Report from DCE – Danish Centre for Environment and Energy No. 162 2015

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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 2014 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_2 was found in concentrations above EU limit values while NO_2 levels in Odense, Aarhus and Aalborg were below the limit value. Model calculations indicate exceedances of NO_2 limit values at several streets in Copenhagen. Annual averages of PM_{10} and $PM_{2.5}$ were below limit values at all stations. The concentrations for most pollutants have been decreasing during the last decades.

Keywords: Atmospheric pollution, urban pollution, nitrogen compounds, ozone, sulphur compounds, heavy metals, volatile organic pollutants, dispersion models and measurements.

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Contents

Summary and Conclusion

This report presents the result from the Danish Air Quality Monitoring Programme in 2014. 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 (SO_2), nitrogen oxides (NO_x/NO_2), particulate mass (PM_{10} and $PM_{2.5}$), particle number, benzene (C_6H_6), toluene (C_7H_8), carbon monoxide (CO), ozone (O_3), polycyclic aromatic hydrocarbons (PAHs) and a number of heavy metals including lead (Pb), arsenic (As), cadmium (Cd), mercury (Hg) and nickel (Ni). In 2009 the programme was expanded with measurements of 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.

In 2014 the permitted number of exceedances in a year of the diurnal limit value of 50 $\mu g/m^3$ for PM_{10} were not exceeded at any stations in the measuring network, even at stations where exceedances previously has occurred (the two traffic stations in Copenhagen (HACB/1103 and Jagtvej/1257)). There were likewise no exceedances of the annual limit values for PM_{10} (40 $\mu g/m^3$) and $PM_{2.5}$ (25 $\mu g/m^3$ from 2015).

The number of particles in ambient air was about 14000 particles per cm^3 as annual average at the street station H.C. Andersens Boulevard. This is a factor of roughly about 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 PM_{10} on street stations was about 1.5 $\mu g/m^3$ corresponding to an estimated annual salt content (NaCl) of about 3.7 $\mu g/m^3$. High diurnal values of salt were observed during periods with winter salting of roads.

The annual limit value for NO_2 (40 $\mu g/m^3$) was exceeded at one street station in Copenhagen (H.C. Andersens Boulevard), whereas no exceedances were observed in Odense, Aalborg and Aarhus. The NO_2 concentrations were in 2014 on the same level as in 2013 for most of the stations. At H.C. Andersens Boulevard (HCAB/1103) the concentrations were lower in 2014 compared to 2013; however, there were still elevated concentrations of NO_2 compared to the situation before 2010. The main reason for these elevated concentrations is a permanent change in the traffic lanes at the street segment in front of the measurement station.

Model calculations at selected streets in Copenhagen and Aalborg indicate that the limit value was exceeded at 11 out of 98 calculated streets in Copenhagen but not at any streets in Aalborg in 2014.

The ozone levels were in 2014 higher than in 2013 at all rural and urban background stations, however the changes are small and no clear trend is observed for the average ozone concentration. The information threshold at 180 $\mu g/m^3$ was not exceeded in 2014. The target value for the max. 8 hours ozone concentration on 120 $\mu g/m^3$ was not exceeded, but the long-term objective for this parameter was exceeded at all Danish stations.

Measurements of volatile organic compounds (VOCs) at the urban background in Copenhagen showed concentration levels between 0.02 $\mu g/m^3$ and 0.89 $\mu g/m^3$ 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 an 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_2 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 particle bound PAH concentrations 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^3 and 0.39 ng/m^3 at H.C. Andersens Boulevard and Hvidovre, respectively. The higher concentrations at Hvidovre are due local use of wood burning for residential warming. The target value for benzo[a] pyrene (1 ng/m^3) was not exceeded in 2014.

Mearurements of the chemical content in $PM_{2.5}$ showed that the annual average concentrations of NH_4^+, Na^+, K^+, Mg^{2+}, Cl^-, NO_3^-, SO_4^{2-} 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^{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.

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 2014 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 (SO_2), nitrogenoxider (NO_x/NO_2), partikelmasse (PM_{10} og $PM_{2.5}$), partikel antal, benzen (C_6H_6) og toluen (C_7H_8), carbonmonoxid (CO), ozon (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. Samtidig tjener resultaterne 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 der 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 det nye luftkvalitetsdirektiv (EC 2008). En af de væsentligste ændringer i det nye direktiv i forhold til de tre første datterdirektiver (1999, 2000 og 2002) er, at der stilles krav om målinger af de fine partikler ($PM_{2.5}$), og at der er indført en grænseværdi for $PM_{2.5}$, som skal overholdes i 2015.

De væsentligste konklusioner fra overvågningsprogrammet i 2014 er følgende:

- I 2014 blev grænseværdien for NO_2 overskredet på en (H.C. Andersens Boulevard) af de to gademålestationer i København. I Odense, Aarhus og Aalborg var der ingen overskridelser. Koncentrationerne af NO_2 i 2014 var for hovedparten af målestationerne stort set på niveau med koncentrationerne målt i 2013. På gademålestationen ved H.C. Andersens Boulevard var koncentrationerne lavere i 2014 end 2013, men der er fortsat en forhøjet koncentration af NO_2 set i forhold til perioden før 2010. Den væsentligste årsag til den forøgede koncentration på H.C. Andersens Boulevard er en permanent ændring af vejbanerne ud for målestationen på H.C. Andersens Boulevard.
- Modelberegninger indikerer, at grænseværdien i 2014 var overskredet på 11 ud af 98 beregnede gadestrækninger i København, men ikke på udvalgte gadestrækninger i Aalborg.

- I 2014 var der ingen målestationer i måleprogrammet, hvor det tilladte antal af overskridelser af den daglige middelværdi for PM_{10} (50 µg/m^3 må ikke overskrides mere end 35 gange årligt) blev overskredet.
- Indholdet af partikler mindre end 2,5 µm ($PM_{2.5}$) overskred ikke den kommende grænseværdi på 25 µg/m^3 som årsmiddelværdi, der skal overholdes fra 2015.
- Antallet af partikler mellem 6 og 700 nm var omkring 14.000 partikler per cm^3 på gademålestationen H.C. Andersens Boulevard, mens det var betydeligt mindre i by- og landbaggrund. Antallet af partikler på H. C. Andersens Boulevard faldt fra 2013 til 2014 og ligger nu på niveau med 2012. Det overordnede billede er fortsat, at antallet af partikler er faldet med omkring en faktor 2 siden 2002 på H.C. Andersens Boulevard.
- Indholdet af natrium i PM_{10} på gademålestationerne var omkring 1,5 µg/m^3 svarende til et estimeret saltindhold (NaCl) på omkring 3,7 µg/m^3.
- Ozonkoncentrationerne var lidt højere i 2014 end i 2013, hvilket skyldes langtransport af ozon til Danmark. Der er ikke fastsat egentlige grænseværdier for ozon (O_3), men kun "målværdier" og ”langsigtede mål” (hensigtsværdier). Der var i 2014 ingen overskridelser af målværdierne for beskyttelse af sundhed, mens de langsigtede mål blev overskredet på alle bybaggrunds- og landstationerne. Tærsklen for information af befolkningen om høje ozonniveauer (timemiddel 180 µg/m^3) blev heller ikke overskredet i 2014.
- De øvrige målte stoffer findes i koncentrationer under grænseværdierne, og for flere stoffer (fx svovldioxid og bly) er koncentrationerne faldet betydeligt 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^3, og 0,39 ng/m^3 på henholdsvis H.C. Andersens Boulevard og ved målestationen i Hvidovre. De højere koncentrationer i Hvidovre skyldes lokal anvendelse af brændeovne til boligopvarmning. Målværdien på 1 ng/m^3 var således ikke overskredet i 2014.
- Målinger af 17 udvalgte flygtige organiske kulbrinter (VOC’er) i bybaggrund i København viser koncentrationsniveauer, som spænder fra 0,02 µg/m^3 til 0,89 µg/m^3 i 2013. 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 $PM_{2.5}$ ved gademålestationen ved H. C. Andersens Boulevard og ved landbaggrundsmålestationen på Risø viser ligesom i 2011-2013, at de årlige gennemsnitskoncentrationer for NH_4^+, Na^+, K^+, Mg^{2+}, Cl^-, NO_3^- og SO_4^{2-} er stort set ens på de to stationer. Dette skyldes, at de 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^{2+}, hvor koncentrationerne er højere på gadestationen som følge af udledninger 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 quality criteria's are identical with 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). In 2012-2014 there has only been minor changes were the most important are:

- During the course of 2012-2014 low volume samplers (LVS) for gravimetric determination of particle mass based on the reference method were introduced into the regular measuring programme and installed at eight stations in the network (HCAB PM_{10} and $PM_{2.5}$; HCØ PM_{10} and $PM_{2.5}$; Jagtvej PM_{10} and $PM_{2.5}$; Risø PM_{10} and $PM_{2.5}$) starting during August and September 2012 to replace some of the older SM200 instruments that needed to be renewed. See table 2.1 for explanation of station names.
- 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.
- The urban background measurement station in Aarhus was 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. 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 2014 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 1980's 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 loosen the criteria on the distance between the street station and urban background station, since it has not been possible to fulfil this criterion when new sites had to be found for example in Aarhus.

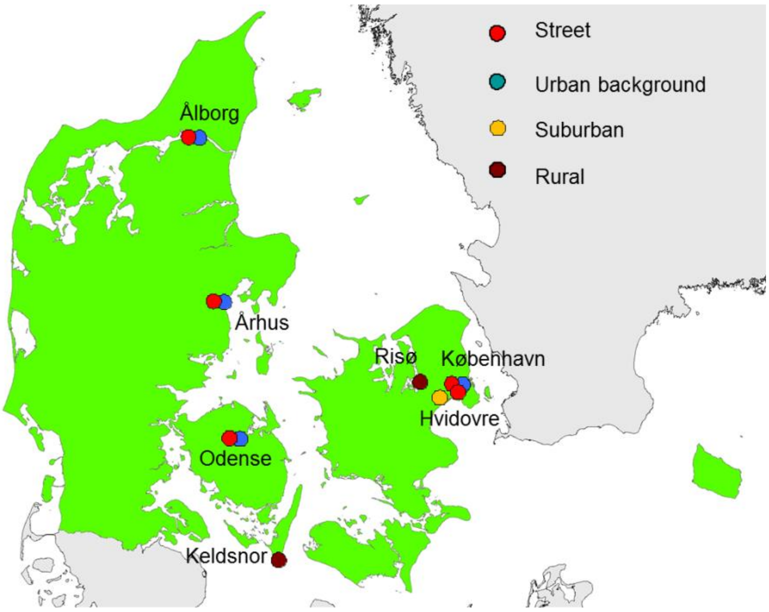


Figure 2.1. Main stations used for monitoring of air quality in relation to health.

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

Location	Station type	Station number
Copenhagen		
H.C. Andersens Boulevard (HCAB)	Street	1103
Jagtvej	Street	1257
H.C. Ørsted Institute (HCØ)	Urban background	1259
Hvidovre, Fjelstedvej 2650	Suburban	2650
Odense		
Albanigade	Street	9155
Town hall in Odense	Urban background	9159
Aarhus		
Banegårdsgade	Street	6153
Botanical Garden	Urban Background	6160
Aalborg		
Vesterbro, Limfjordsbroen	Street	8151
Aalborg/8158	Urban background	8150
Rural		
Lille Valby/Risø*	Rural background	2090l
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 there were three larger changes for the stations:

- The measurement station on Vesterbro at Limfjordsbroen in Aalborg was closed down temporarily on September 8th 2014 due to a large rebuild of the nearby laying house. The results for 2014 represent therefore only data for the 250 days (70%) of 2014.
- In Odense a traffic plan has been adopted by the municipality for the entire city centre and the realisation 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 January 2014 the urban background station in Aarhus was moved to a new site since the municipality had to sell the house that the measurements station was placed upon. The new site is laying 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 2014:

- Nitrogen oxides (NO, NO_2 and NO_x (= NO + NO_2)) were measured at all stations except Hvidovre.
- Particle mass (PM_{10} and/or $PM_{2.5}$) were measured as 24 hour averages at all stations except Odense/urban background and Hvidovre. At ten stations (HCAB PM_{10} and $PM_{2.5}$; HCØ PM_{10} and $PM_{2.5}$; Jagtvej PM_{10} and $PM_{2.5}$; Risø PM_{10} and $PM_{2.5}$; Aarhus/street $PM_{2.5}$; Aarhus/urban background $PM_{2.5}$) PM was measured throughout the year by using low volume samplers (LVS) for gravimetric determination of particle mass based on the reference method (EN 12341: 2014). At two stations (Aalborg/street $PM_{2.5}$ and Aalborg/urban background $PM_{2.5}$) the measurements for 2014 were a combination of LVS gravimetric and SM200 beta gauge determination of particle mass. At the remaining three stations (Aarhus/street PM_{10}; Keldsnor PM_{10}; Odense/street PM_{10}) PM was determined solely by using SM200 β-gauges measurements throughout the year.
- Elements (heavy metals) in PM_{10} were measured at Copenhagen/HCAB, Copenhagen/urban background, Aarhus/street, Odense/street and Risø.
- Additionally PM_{10} and $PM_{2.5}$ was measured at both Copenhagen/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/HCAB, Copenhagen/urban background and Risø in cooperation with particle research project funded separately by the Danish EPA.
- Ozone (O_3) was measured at all urban background and rural stations, and at the street stations Copenhagen/HCAB.
- Carbon monoxide (CO) was measured at all street stations as well as at the urban background station, Copenhagen/urban background and the rural site Risø.
- Benzene and Toluene were measured at Copenhagen/HCAB and Copenhagen/urban background using passive sampling on a weekly basis.
- PAHs were measured at Copenhagen/HCAB.
- SO_2 was measured at Aalborg/street and at Copenhagen/HCAB. The main purpose was to monitor episodic high concentrations.
- Elemental carbon (EC) and organic carbon (OC) were measured at Copenhagen/HCAB and 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_x, NO_2, O_3, CO, SO_2) and particle number were recorded as ½-hour averages. Particle mass (PM_{10} and $PM_{2.5}$) 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 at 24 hour averages.

2.2 Model calculations

In the monitoring programme the measurements at the permanent measurement stations are supplemented with model calculations using the THOR modelling system. In the present report model results are presented for NO_2 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 2014 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 nest is covering Europe with a resolution of 50 km x 50 km. The second nest 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 nest 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 2010/2011 compiled by DCE (http://envs.au.dk/en/knowledge/air/emissions/) and international emission inventories for the year 2012 collected and distributed by EMEP (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_x and ozone. The basic principles of the model are described in Berkowicz (2000b). In the recent year 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 newly developed SPREAD model that spatially distributes national emissions from 2010 from all sectors on a 1 km x 1 km grid for Denmark (Plejdrup & Gyldenkærne 2011). Updated spatial variations of emissions for more recent years are under development in a study.

Finally, the street canyon model OSPM® (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 model 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;
http://envs.au.dk/videnudveksling/luft/model/airgis/).

The model calculations for 2014 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_2 concentration in 98 streets in Copenhagen and 31 streets in Aalborg.

2.2.1 Improved input data and re-calibration of OSPM

In the previous assessment for 2013 the model calculations with OSPM were improved through major revisions. These include changes related to the general building height, revision of NO_x 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 2014 is similar to that of 2013. As for the 2013 assessment OSPM has been calibrated against 2014 measurements at all street stations based on adjustment of one of the dispersion parameters in the model.

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. 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 42 out of the 98 calculation points had updated traffic data for 2014. For Aalborg 13 out of 31 streets had updated traffic data.

Manual traffic counts are now carried out annually for the street segments in front of the measurement stations of H.C. Andersens Boulevard and Jagtvej starting from April 2013. Manual counts for the 2014 assessment originate from September 2014. Aalborg does not have a systematic traffic counting program and traffic data is based on available traffic data from manual and automatic counts together with data from a traffic model. However, automatic traffic counts have been obtained for Vesterbro in Aalborg at the monitoring station. As something new detailed traffic data has also been obtained for Aarhus (Banegårdsgade) and Odense (Albanigade) based on traffic counts at the locations of the monitor stations for the 2014 assessment. Previously traffic data originated from the Low Emission Zone Project (Jensen et al., 2011). SpeedMap travel speeds have been applied for Aarhus and Odense in the street calculations for 2013 as well as for 2014.

Calculations with the full model chain of DEHM-UBM-OSPM have been compared to measured NO_2 concentrations in 2014 for the fixed street monitoring stations in Copenhagen, Aarhus, Odense and Aalborg. The model system slightly underestimates annual NO_2 concentrations with 5% for Jagtvej (Copenhagen), overestimates by 14% for H.C. Andersens Boulevard (Copenhagen), slightly underestimates by 3% for Banegårdgade (Aarhus), slightly overestimates by 9% for Albanigade (Odense) and slightly underestimates by 6% for Vesterbro (Aalborg). Calculations with the coupled DEHM-UBM models have also been compared to the fixed urban background monitoring stations in Copenhagen, Aarhus, Odense and Aalborg. Here the model system slightly overestimates annual NO_2 concentrations with 0-12%.

The comparison of the modelled NO_2 concentrations presented in this report for 2014 with measurements at the 5 street locations shows a good overall agreement within -5 to 14% (table 2.2).

For Odense, traffic has changed drastically during 2014 because Thomas B. Thrige Street was closed on a stretch of the road north of the monitor station, which has led to significantly less traffic on Albanigade where the monitoring station is located. The street closure took place on June 28, 2014. Therefore, model calculations were based on traffic data for the period January 1 to June 27, 2014 and another set of traffic data for the period June 28, 2014 to December 31, 2014.

Table 2.2. Comparison of modelled and measured annual means of NO_2 concentrations in 2014

Unit: µg/m^3	Measure-ments	Model results	Difference %	Models used
Traffic:				
Copenhagen/Jagtvej/1257	37	35	-5%	DEHM/UBM/OSPM
Copenhagen/HCAB/1103	51/43*	50	14%	DEHM/UBM/OSPM
Aarhus/6153	34	33	-3%	DEHM/UBM/OSPM
Odense/9155	23	25	9%	DEHM/UBM/OSPM
Aalborg/8151	32	30	-6%	DEHM/UBM/OSPM
Urban Background:				
Copenhagen/1259	17	17	0.3%	DEHM/UBM
Aarhus/6159	15	16	12%	DEHM/UBM
Odense/9159	14	15	6%	DEHM/UBM
Aalborg/8159	13	13	3%	DEHM/UBM
Rural:				
Risø/2090	8.8	14	63%	DEHM/UBM
Keldsnor/9055	8.3	9.3	13%	DEHM/UBM

* 51 µg/m^3 is measured at the monitoring 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 8-9 µg/m^3. Without the change in street layout, about 43 µg/m^3 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 monitor 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 setting. An ongoing PhD study together with other supporting studies is expected in a few years time to result in a new version of the model, which has improved capabilities to describe the complex distribution of emissions in streets, definition of location of calculation point etc. These improvements are expected to improve model calculations for complex street layouts, e.g. for HCAB.

3 Nitrogen oxides

The nitrogen oxides (NO, NO_2, NO_x) 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 2014 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_2 on 40 µg/m^3 (EC, 2008) at H.C. Andersens Boulevard (Copenhagen/1103). There were no exceedances of the hourly limit value for NO_2 on 200 µg/m^3. This value must not be exceeded more than 18 times in a calendar year (see 19th highest hourly concentration in table 3.1). In 2014 there was no information to the public due to exceedance of the information threshold for NO_2 (three hours average must not exceed 400 µg/m^3).

Table 3.1. Nitrogen dioxide (NO_2) in 2014. All parameters are based on hourly averages.

Unit: µg/m^3	Number	Average	Median	98. percentile	19. highest
Traffic:					
Copenhagen/1257	8310	37	34	90	114
Copenhagen/1103	8297	51*	48	110	142
Aarhus/6153	7360	34	32	77	96
Odense/9155 §§	8320	23	18	73	98
Aalborg/8151 §	5658	32	27	88	109
Urban Background:					
Copenhagen/1259	8213	16	14	49	72
Aarhus/6160	7691	15	12	47	64
Odense/9159	7682	14	12	42	61
Aalborg/8159	8238	13	10	43	64
Rural:					
Risø	8279	9	6	32	48
Keldsnor/9055	7618	8	6	29	42
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 2014 excluding hours used for calibration.

§) Aalborg/8151 (traffic) is only represented by data covering the period 1/1 - 8/9 as the station was closed temporarily due to construction work at the site.

§§) The site in Odense/9155 (Albanigade) was affected by a greater permanent rearrangement of the roads in Odense. The site changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on June 28th. 2014.

Table 3.2. Nitrogen oxides (NO_x=NO+NO_2) in 2014. All parameters are based on hourly averages.

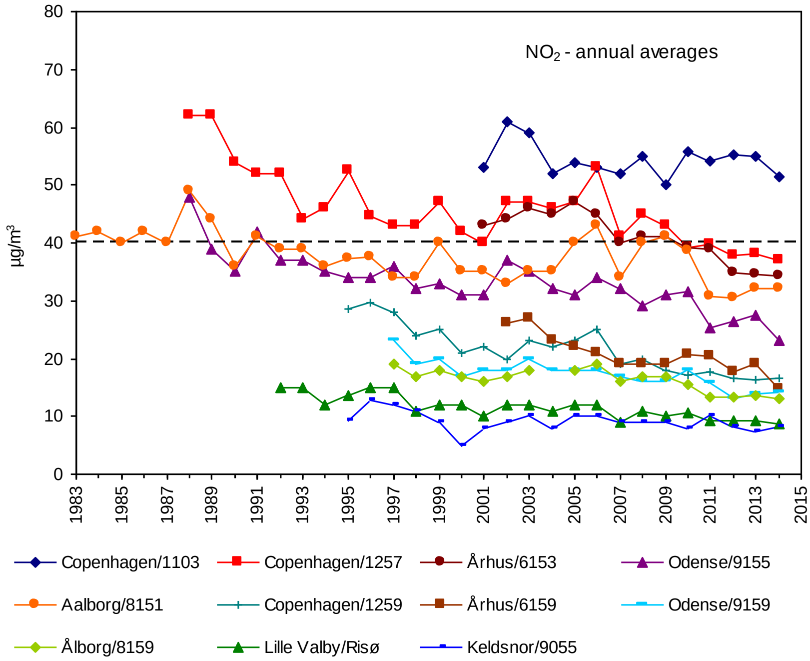
Unit: µg/m^3 (as NO_2)	Number	Average	Median	98. percentile	19. highest
Traffic:					
Copenhagen/1257	8310	82	62	277	453
Copenhagen/1103	8297	128	103	379	539
Aarhus/6153	7360	77	61	248	382
Odense/9155	8320	46	28	205	360
Aalborg/8151	5658	81	59	286	419
Urban Background:					
Copenhagen/1259	8213	21	16	74	150
Aarhus/6160	7691	19	14	76	210
Odense/9159	7682	18	14	64	149
Aalborg/8159	8238	18	12	68	226
Rural:					
Risø	8279	10	7	40	75
Keldsnor/9055	7618	10	7	38	61

3.2 Trends

The long term trends for NO_2 and NO_x are shown in figure 3.1. For 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 and obligatory use of catalytic converters.

The long term trend for nitrogen dioxide decreases much slower than observed for NO_x. This is mainly due to an increase in the share of diesel cars where up to about half of the emissions of NO_x consist of NO_2 (called direct NO_2). In comparison gasoline cars emit nearly all NO_x as NO. This increase of the direct emissions of NO_2 slows down the decrease of the concentrations of NO_2 compared to NO_x.

At Odense street station and Aarhus urban background station there were larger decreases from 2013 to 2014 than at the other measurement stations. In Odense there was a greater permanent rearrangement of the roads in Odense Centre that changed the traffic at the street station in Albanigade from a street with relatively high traffic intensity to a street with much reduced traffic intensity. This change took place on June 28th. 2014. This is the reason behind the larger decrease of the NO_2 and NO_x values for Odense/9155 in 2014 than for the other traffic stations in 2014 compared to 2013. The large change at Aarhus/background is due to the relocation of the measurement site (Chapter 2.1).



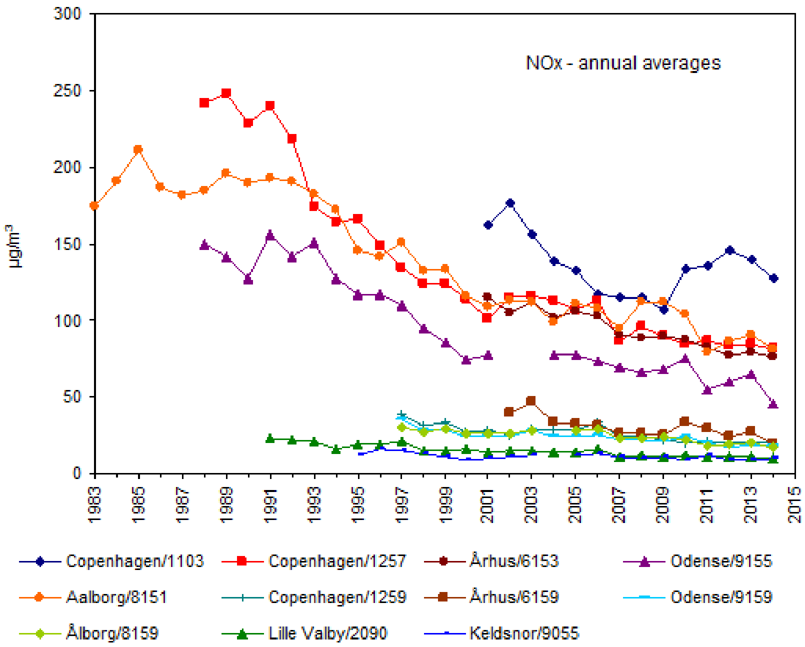


Figure 3.1. The graphs show the time series for the annual average values of NO_2 and NO_x. The dashed line on the upper graph shows the limit value that entered into force in 2010. Results from the previous (6159) and the new background station (6160) in Aarhus are shown on the same curve.

Both NO_2 and NO_x were higher in 2010-2014 compared to 2009 at the street station H.C. Andersens Boulevard (Copenhagen/1103). At all other street stations the levels in 2010-2014 were equal to or lower than observed in 2009. A detailed study at H.C. Andersens Boulevard has shown that the higher concentrations at the measurement station is mainly due to a permanent change in the traffic lanes, and that this is the main reason for the 8-9 µg/m^3 increase in NO_2 that has been observed since the middle of 2010 (Ellermann et al., 2014).

3.3 Results from model calculations

Model calculations of NO_2 have been performed for selected streets in Copenhagen (capital) and Aalborg (fourth largest city) as well as in a resolution of 5.6 km x 5.6 km for the entire country.

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. In Aalborg, on average traffic volumes increased 1%, heavy-duty share decreased about 4% and travel speed decreased 7% from 2013 to 2014. The reason for the decrease in travel speed is due to new assumptions as streets with no SpeedMap information were assumed to have similar average speed as streets with SpeedMap information. In Copenhagen, traffic volume on average decreased 1% and heavy-duty share increased 6% compared to 2013. Travel speeds are assumed to be unchanged.

Model calculations have been carried out in order to determine the annual concentrations of NO_2 for comparison with limit values. The air quality limit value for the annual mean is 40 µg/m^3. The number of exceedances is also given. An exceedance is registered if the calculated concentration is higher than 40.5 µg/m^3 since the limit value is given as an integer.

3.3.1 Model calculations for Copenhagen

The annual mean concentrations of NO_2 for streets in Copenhagen in 2014 are shown in figure 3.2 (bar chart) and figure 3.3 (map).

The average of NO_2 concentrations at all 98 streets has decreased 4% from 2013 to 2014. The general decrease is a result of a combination of changes in traffic, emission factors, urban background and meteorology. Modelled urban background concentrations decreased 3% or about 0.4 µg/m^3 which accounts for about one third of the reduction in street concentrations. Vehicle emission factors show a decrease due to the general replacement of the car fleet and leads to decrease in modelled concentrations. There is a slight decrease in ADT but an increase in heavy-duty share that should lead to slightly higher concentrations. In 2014 the limit value for the annual mean concentration was exceeded in 11 out of the 98 selected streets in Copenhagen according to the model results (figure 3.2). This is slightly less than in 2013 where the number of streets exceeding the limit value was 12 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.

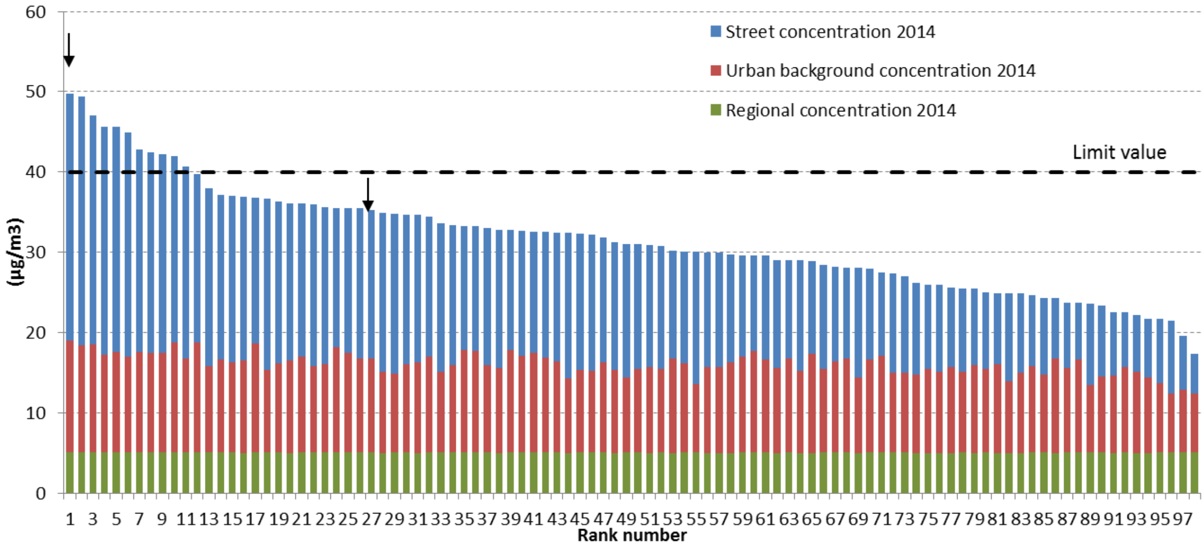


Figure 3.2. Annual mean concentrations of NO_2 in 2014 for 98 streets in Copenhagen. The contribution from traffic in the street canyons is based on the street canyon model OSPM®. The urban background (dark red colour) is obtained from calculations with the urban background model UBM with input from 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 measurement station it is the side of the location of the station. The names of the streets can be seen in table 3.3. Arrows indicate street segments with a measurement station.

The streets where the limit value were exceeded all have average daily traffic in the range of 12,200 to 67,600 vehicles per day. However, it is not only the traffic intensity alone which determines the concentration of NO_2. 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 façade, orientation of the street and background concentrations and meteorology have impacts on the concentration of NO_2 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_2 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 Nørre Søgade to Øster Søgade.

There have been minor changes in the ranking of streets according to NO_2 concentrations from 2013 to 2014. The highest modelled NO_2 concentration in 2014 is at H.C. Andersens Boulevard (1) (50 µg/m^3) which is the location of the monitoring station. Observed concentrations are 51/43 µg/m^3 (explanation in table 2.2). This location was also ranked number one in 2013.

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_2 levels in 2014 (1 = highest, 98 = lowest). The numbers in parenthesis refer to different segments of the same street that has more than one model calculation. * indicate the street segments with measurement stations. 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.

Number	Street name	Number	Street name	Number	Street name
1	H C Andersens Boulevard(1)*	34	Gammel Kongevej(1)	67	Tagensvej(4)
2	H C Andersens Boulevard(2)	35	Vester Farimagsgade	68	Jagtvej(2)
3	H C Andersens Boulevard(3)	36	Nørre Farimagsgade	69	Frederikssundsvej(5)
4	Øster Søgade	37	Toldbodgade	70	Bülowsvej(2)
5	Gyldenløvesgade	38	Nordre Fasanvej(1)	71	Øster Voldgade(2)
6	Ågade	39	Nørre Voldgade(2)	72	Amager Boulevard
7	Nørre Søgade	40	Nordre Fasanvej(3)	73	Røde Mellemvej(1)
8	Åboulevard(3)	41	Gothersgade(1)	74	Ålholmvej(2)
9	Åboulevard(1)	42	Tagensvej(3)	75	Tuborgvej(1)
10	Stormgade	43	Jagtvej(3)	76	Frederikssundsvej(2)
11	Vesterbrogade(1)	44	Strandvejen(1)	77	Slotsherrensvej(2)
12	Bernstorffsgade(1)	45	Torvegade	78	Folke Bernadottes Allé
13	Toftegårds Allé(1)	46	Amagerfælledvej	79	Peter Bangs Vej(1)
14	Fredensgade	47	Godthåbsvej(3)	80	Blegdamsvej
15	Bredgade	48	Tuborgvej(2)	81	Bellahøjvej
16	Frederikssundsvej(3)	49	Søndre Fasanvej(2)	82	Frederiksborgvej(1)
17	Bernstorffsgade(2)	50	Folehaven(1)	83	Slotsherrensvej(1)
18	Østerbrogade(4)	51	Jyllingevej(1)	84	Dag Hammarskjølds Allé
19	Tagensvej(2)	52	Gammel Køge Landevej(1)	85	Peter Bangs Vej(2)
20	Tomsgårdsvej(2)	53	Hulgårdsvej(2)	86	Godthåbsvej(2)
21	H.C. Ørsteds Vej(2)	54	Kalvebod Brygge	87	Strandvænget(2)
22	P Knudsens Gade(2)	55	Amagerbrogade(1)	88	Halmetgade
23	Enghavevej	56	Ålholmvej(1)	89	Englandsvej(1)
24	Hammerichsgade	57	Roskildevej(1)	90	Vesterfælledvej
25	Scandiagade	58	Rebildvej	91	Strandvejen(2)
26	Falkoner Alle(2)	59	Hillerødgade(1)	92	Artillerivej
27	Jagtvej(1)*	60	Ingerslevsgade	93	Frederiksborgvej(2)
28	Frederikssundsvej(8)	61	Nørrebrogade	94	Vigerslevvej(2)
29	Amagerbrogade(2)	62	Tagensvej(1)	95	Gammel Køge Landevej(2)
30	Øster Voldgade(1)	63	Istedgade	96	Amagerbrogade(3)
31	Lyngbyvej(2)	64	Grøndals Parkvej	97	Røde Mellemvej(2)
32	Frederikssundsvej(1)	65	Hillerødgade(3)	98	Englandsvej(2)
33	Vesterbrogade(3)	66	Østerbrogade(1)		

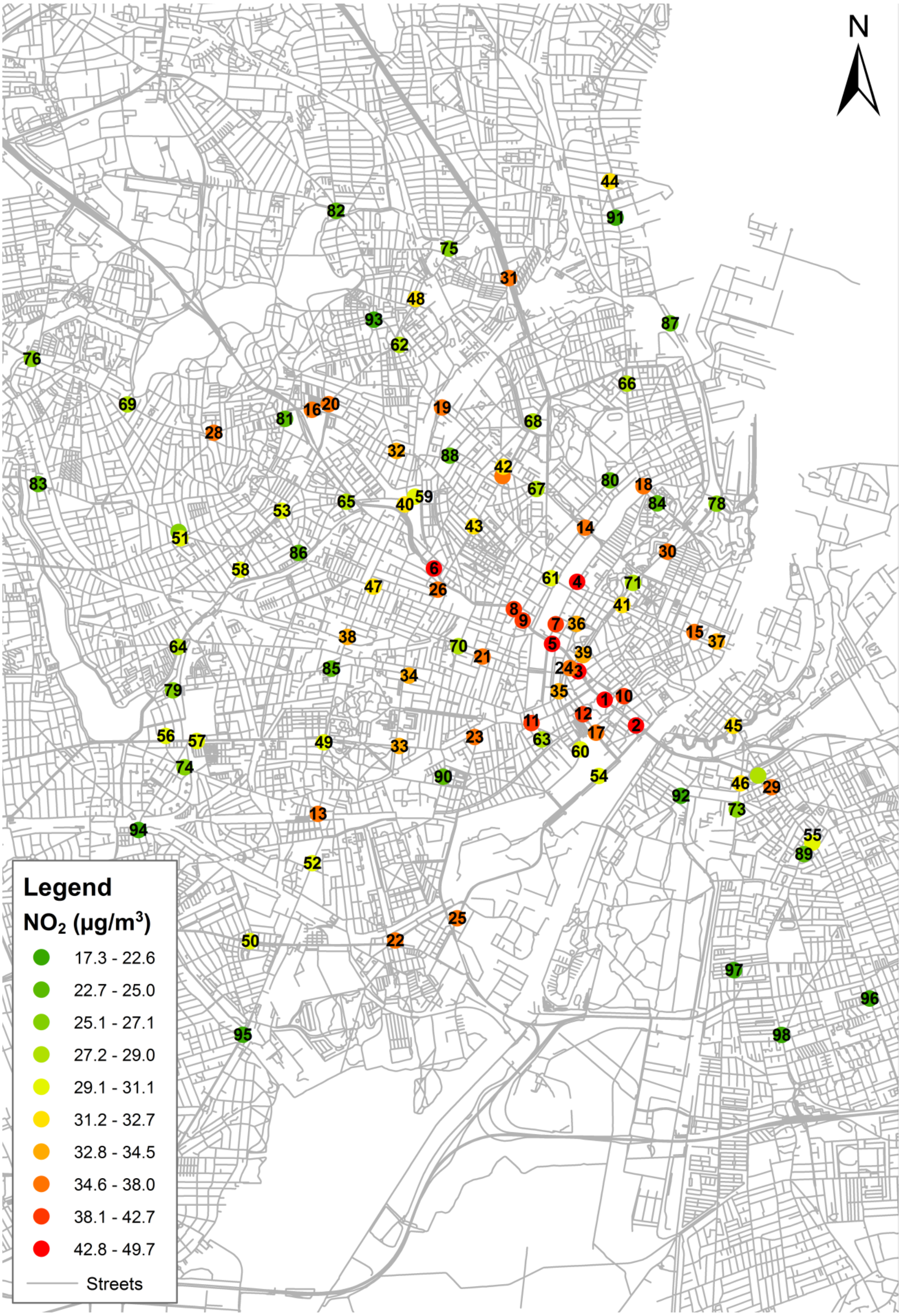


Figure 3.3. Map showing the locations of the selected streets in Copenhagen and the annual mean concentrations of NO_2 for 2014. The contribution from traffic in the street canyons is based on the street canyon model OSPM®. The urban background is obtained from calculations with the urban background 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 of the location of the station. The names and numbers for the streets are shown in table 3.3.

3.3.2 Model calculations for Aalborg

For Aalborg the modelled street concentrations show an average increase of 4% for NO_2 compared to 2013 when considering all 31 street segments. The general increase is a result of a combination of several factors. A modelled increase in urban background concentrations of 0.7 µg/m³ accounts for more than half of the average increase in street contribution. The assumption of reduced travel speeds for some roads also contributes to higher modelled concentrations. On average ADT increased about 1% whereas the heavy-duty share of vehicles decreased about 4% that should lead to slightly lower concentrations. Reduced emissions due to replacement of the car fleet should also lead to lower concentrations.

According to the model calculations the limit value for the annual mean concentration in 2014 was not exceeded at any of the 31 selected streets which was also the case in 2013 (figure 3.4 and figure 3.5). The order of some of the streets has changed slightly due to changes in traffic data.

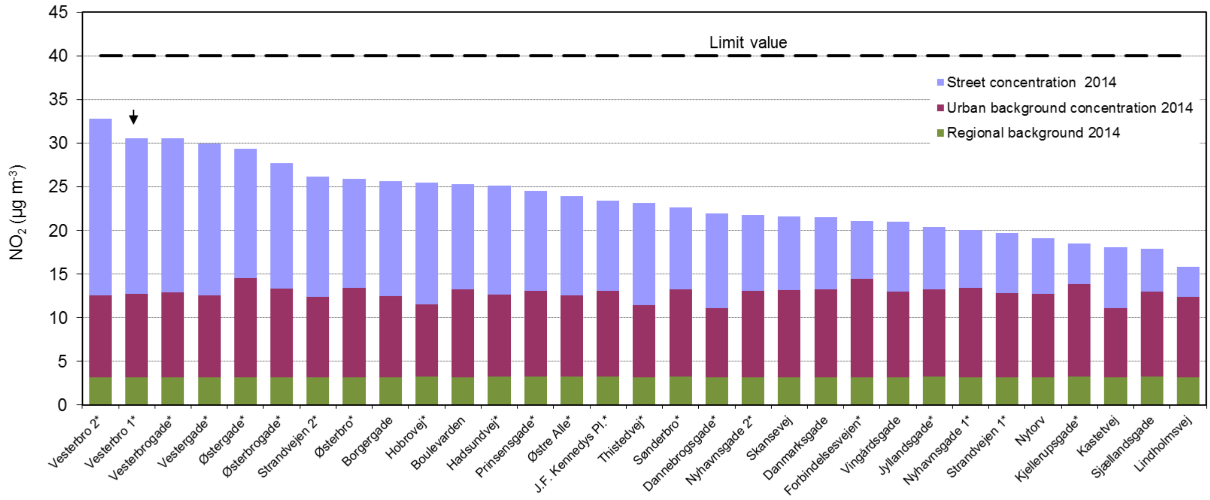


Figure 3.4. Modelled annual mean concentrations of NO_2 in 2014 for 31 streets in Aalborg. The contribution from traffic in the street canyons is based on the street canyon model OSPM®. The urban background (dark red colour) is obtained from calculations with the urban background model UBM with input 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 of the location of the station. The arrow indicates the street segment (Vesterbro 1) with the measurement station. '*' at the end of the street name indicates that calculations were based on SpeedMap data.

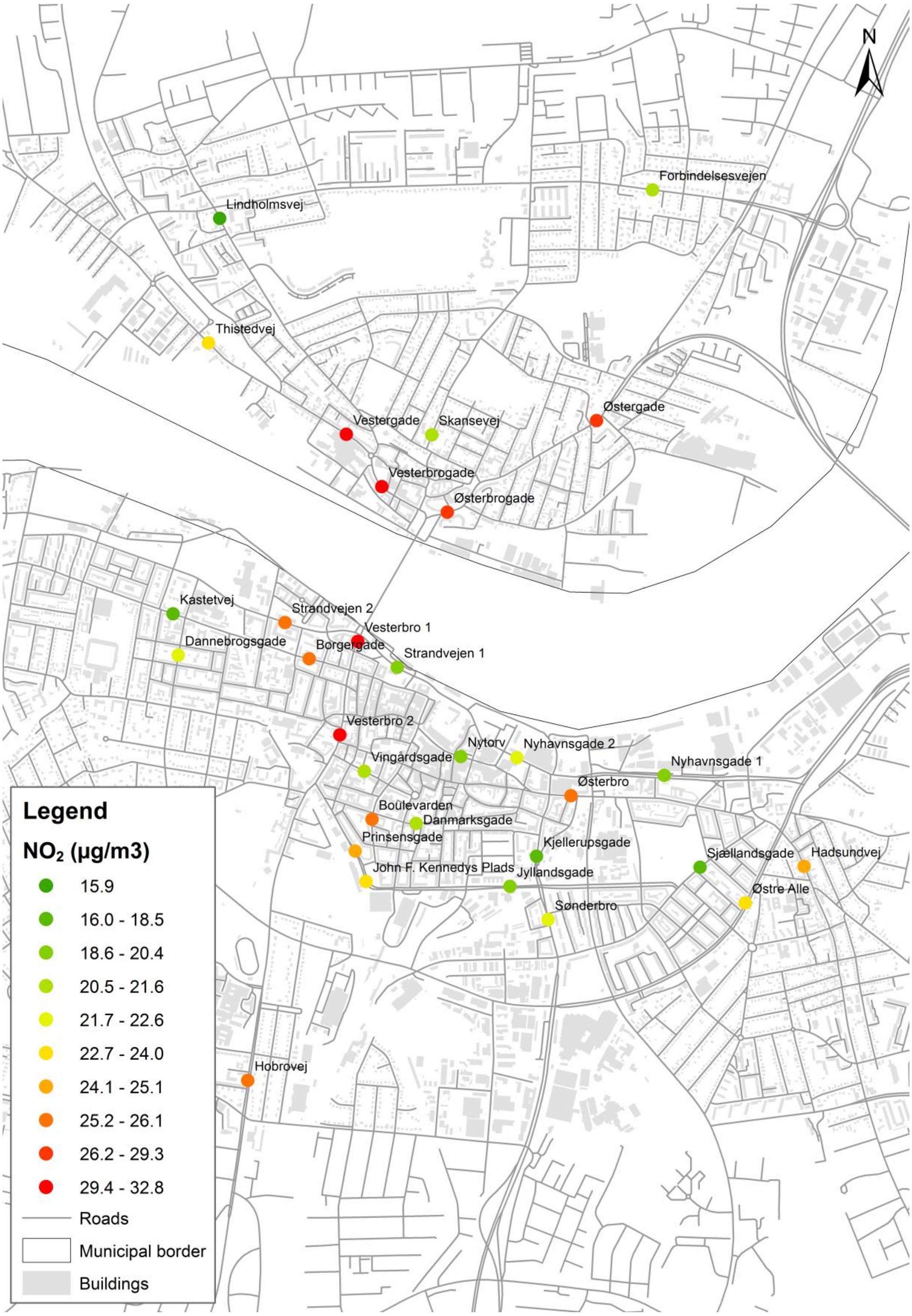


Figure 3.5. Map showing the location of the selected streets in Aalborg and the annual mean concentrations of NO_2 for 2014. The contribution from traffic in the street canyons is based on the street canyon model OSPM®. The urban background is obtained from calculations with the urban background model 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 of the location of the station. Vesterbro 1 is the street segment with the measurement station.

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 this reporting.

4.1 Annual statistics

The annual statistics for 2014 for ozone are shown in table 4.1. The maximum 8 hours daily mean value must not exceed 120 µg/m^3 more than 25 days per calendar year averaged over three years (EC, 2008). This target value was not exceeded for 2012-2014 at any of the stations. The long term objective (maximum 8 hours daily mean value must not exceed 120 µg/m^3; table 4.1 column 5) was exceeded at six of the stations. However, the long term objective has not entered into force.

In 2014 there was no exceedance of the information threshold (hourly average 180 µg/m^3) and no exceedance of the alert threshold (hourly average 240 µg/m^3) for ozone.

Table 4.1. Ozone (O_3) in 2014. All parameters are based on one-hour average values. The eight 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 eight hour average exceeds 120 µg/m^3 averaged over 2012-2014.

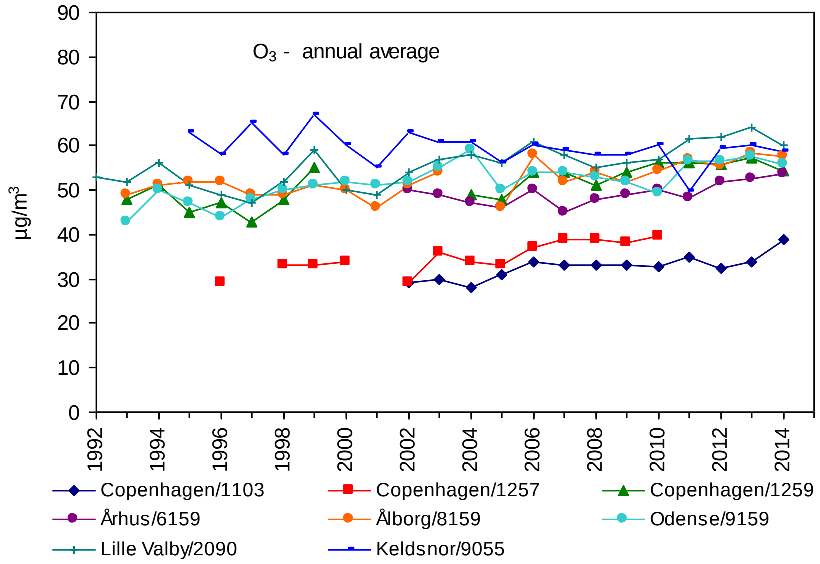
Unit: µg/m^3	Number of results	Average	Median	Max 8 hours	Days above target value 8 hours	Max 1 hour
Urban Background:						
Copenhagen/1259	7775	54	55	156	3	171
Århus/6160	7912	54	56	120	2	133
Odense/9159	7502	56	57	131	5	155
Aalborg/8158	7776	58	59	121	1	135
Rural						
Risø/2090	8010	60	61	165	8	173
Keldsnor/9055	7236	59	61	139	3	148
Traffic						
Copenhagen/1103	8002	39	38	127	0	147
Target value[1]	-	-	-	-	25	-
Long term objective	-	-	-	120	-	-
Information threshold	-	-	-	-	-	180
Data capture*	>7154	-	-	-	-	-

[1] As average over 3 years.

*) 90% data capture of number of hourly measurements in relation to total number of hourly measurements in 2014 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 (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.



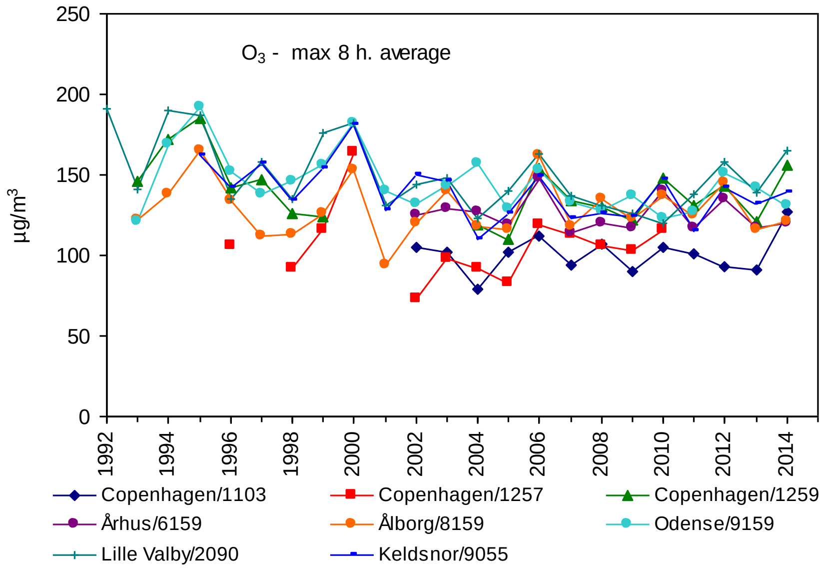


Figure 4.1. Annual average values and the max. 8 hour average value. The latter is calculated as 8 hourly running averages according to the provisions in the EU Directive (EC, 2008). Results from the previous (6159) and the new background station (6160) in Aarhus are shown on the same curve.

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.

The target value for protection of human health is that the running 8 hour mean concentration of ozone must not exceed 120 µg/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 µg/m^3. The target value and long term objective are given in the EU Directive (EC, 2008). Results from the model calculations for 2014 show that the maximum daily 8 hour mean value of 120 µg/m^3 was exceeded up to 16 days during 2014, which is higher than usual (figure 4.3). The target value that is determined as an average over three years (2012-2014), 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 Bornholm, because it is a small island sourround by sea and the long range transported ozone is only deposited slowly over sea.

Both model results and measurements show higher ozone concentrations in 2014 compared to the previous years. This is due to the hot summer in 2014 with larger long range transport of ozone to Denmark than usual.

According to the directive (EC, 2008) the public has to be informed if the one hour average concentration exceeds the information threshold at 180 µg/m^3. Both measurements and model calculations shows that this threshold was not exceeded in 2013 (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.

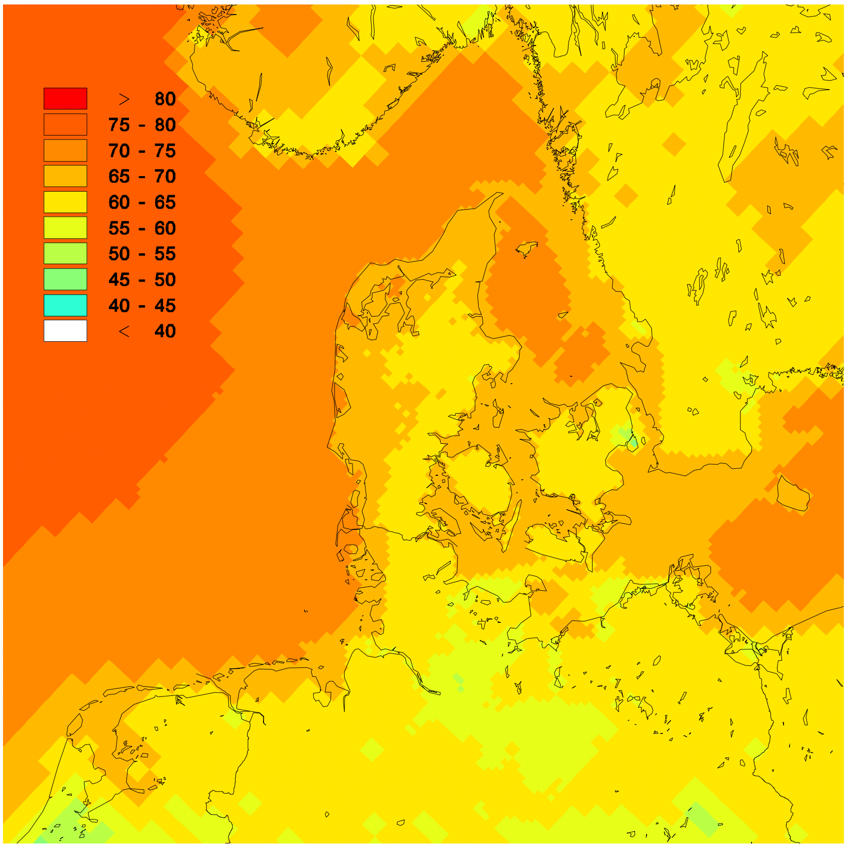


Figure 4.2. Annual mean concentrations of O_3 (µg/m^3) for 2014 calculated using DEHM. The figure shows the average concentrations for the 6 km x 6 km grid cells used in the model.

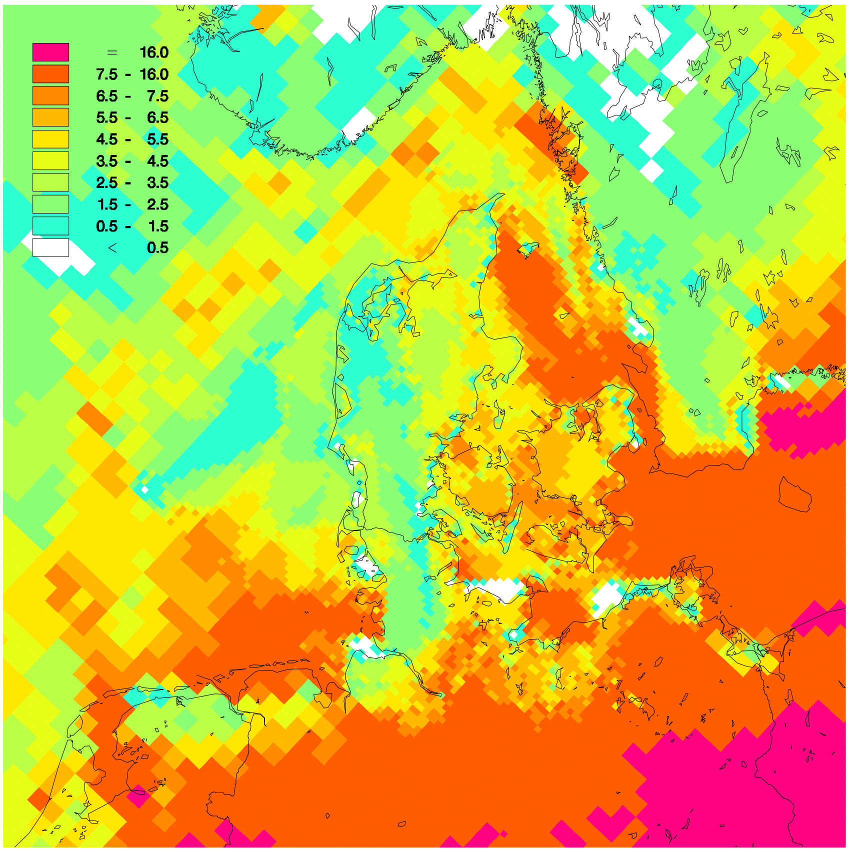


Figure 4.3. Number of exceedances of 120 µg/m^3 for 8-hour running mean concentrations of ozone in 2014. The calculations were carried out using DEHM.

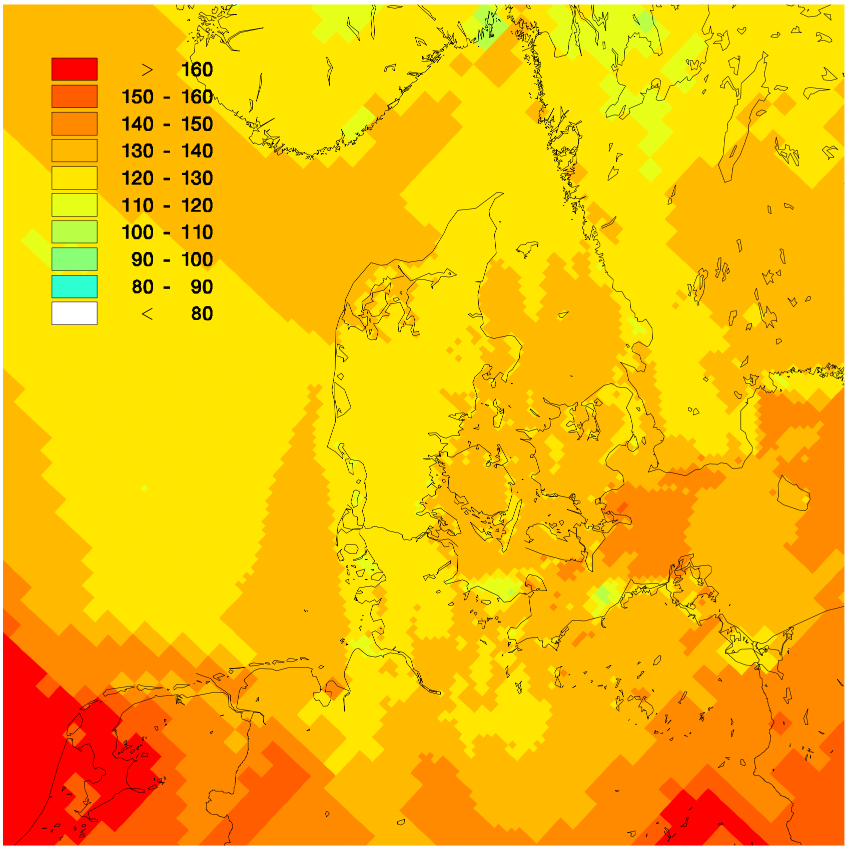


Figure 4.4. Maximum 8 hour running mean concentration (µg/m^3) of ozone in 2014 calculated using DEHM.

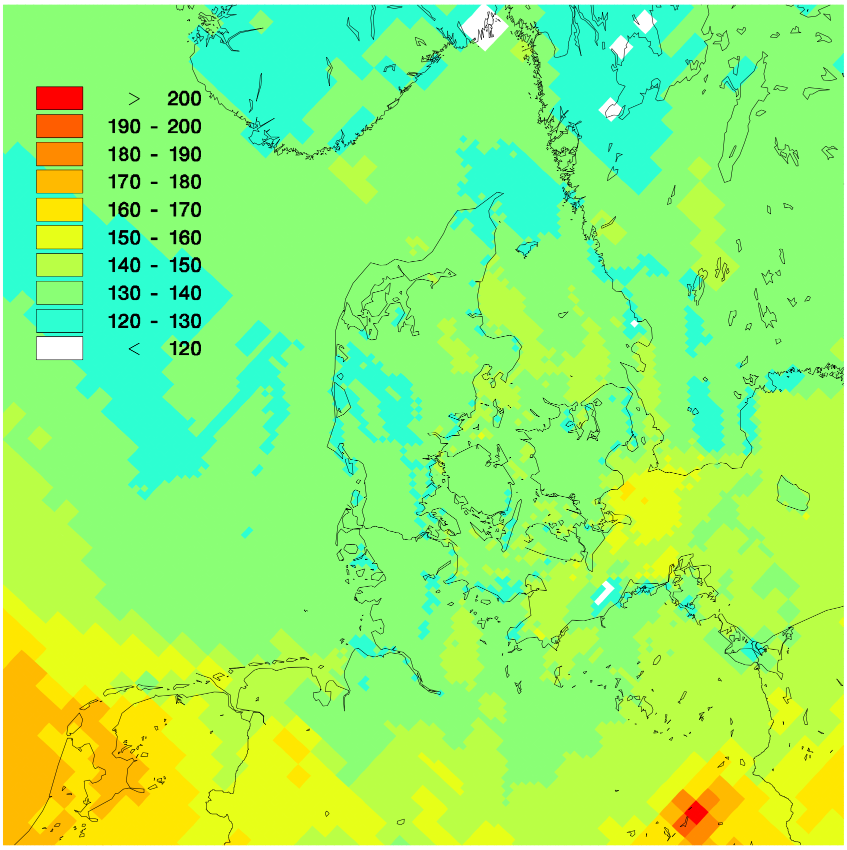


Figure 4.5. Maximum one hour mean concentration of ozone (µg/m^3) in 2014 calculated using DEHM.

5 Carbon monoxide

Carbon monoxide is measured at the four traffic oriented monitoring sites, 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 2014 for carbon monoxide are shown in table 5.1. The limit value for carbon monoxide is based on the maximum daily eight hour average concentration that must not exceed 10.000 µg/m³ (EC, 2008). This limit value was not exceeded at any of the stations.

Table 5.1. Annual statistics for carbon monoxide (CO) in 2014. All parameters are based on hourly average. The 8-hour values are calculated as a moving average based on hourly results.

Unit: µg/m³	Number	Average	Median	98-percentile	99.9-percentile	Max. 8-hours	Max. hour
Traffic:							
Copenhagen/1103	8303	376	349	815	1263	1142	1362
Århus/6153	7682	283	258	625	1659	1734	1799
Odense/9155 §	8339	320	270	785	1176	1077	1312
Aalborg/8151 §§	5671	373	337	809	1181	1049	1550
Urban Background:							
Copenhagen/1259	8159	227	215	442	599	578	658
Rural:							
Risø	8316	172	158	346	459	465	483
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 2014 excluding hours used for calibration.

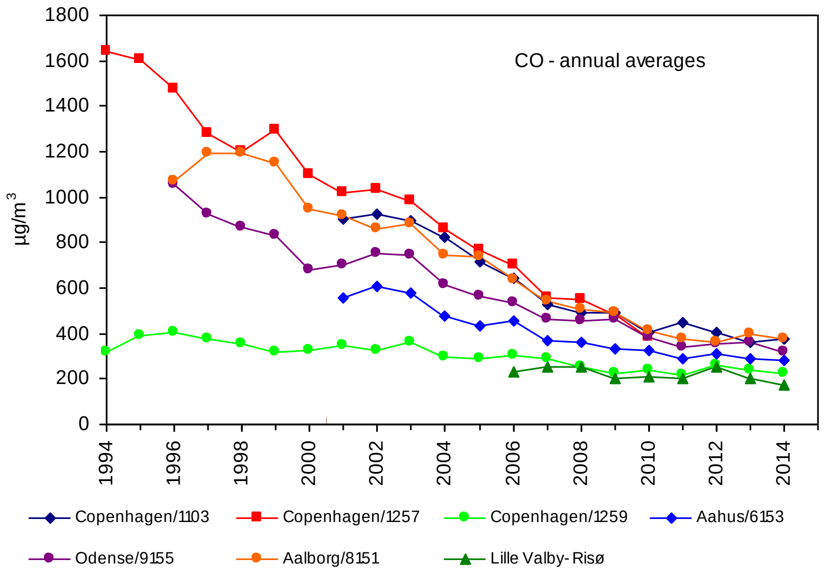
§) The site in Odense/9155 (Albanigade) was affected by a greater permanent rearrangement of the roads in Odense changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on June 28th. 2014.

§§) Aalborg/8151 (traffic) is only represented by data covering the period 1/1 - 8/9 as the station was closed due to construction work at the site.

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 2014 than at the other stations. This is due to a greater permanent rearrangement of the roads in Odense that resulted in a large reduction in the traffic intensity in Albanigade.



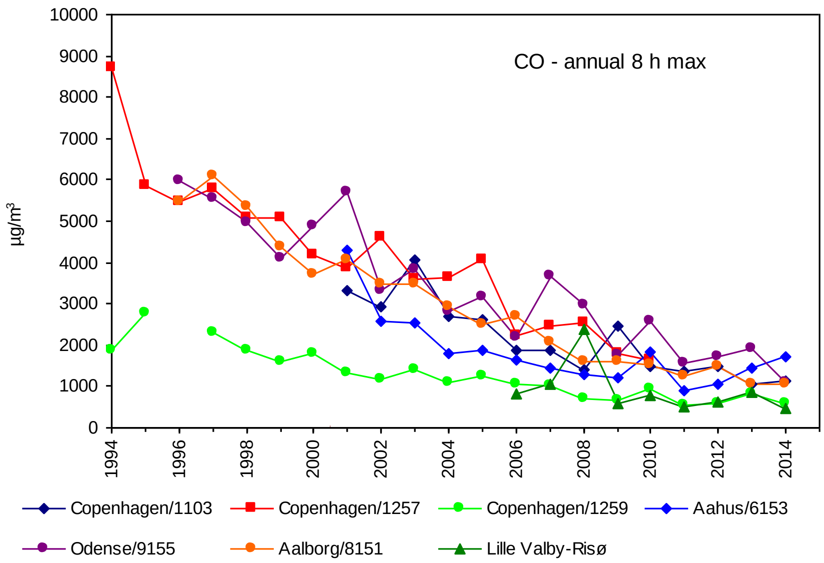


Figure 5.1. Annual average values and highest 8-hour values calculated based on an hourly moving average. The site in Odense/9155 (Albanigade) was due to a greater permanent rearrangement of the roads in Odense changed from a traffic site with relatively high traffic intensity to a site with much reduced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

6 Benzene and Toluene

Benzene and toluene are measured at two kerbside stations in Copenhagen, Jagtvej/1257 and H.C. Andersen's Boulevard/1103, using a passive sampling method of time resolution: one week. Benzene, toluene and 15 other ozone precursors are also measured in urban background (H.C. Ørsted Institute/1259) using active sampling of time resolution: one day (Chapter 12).

6.1 Annual statistics

The annual averages of benzene and toluene in 2014 are listed in table 6.1 and table 6.2. The annual average of benzene was about 20% of the EU-limit value (EC, 2008), whereas the annual average for toluene was far below the WHO guide line value (WHO, 2000).

At the two street stations in Copenhagen the concentrations of benzene and toluene were roughly the same (table 6.1 and 6.2). Benzene and toluene in urban background (HCØ/1259) were respectively, 0.53 μg/m^3 and 0.89 μg/m^3 (Chapter 12), which for both compounds is less than 50% of the concentrations measured at the street stations.

The ratio between toluene and benzene is influenced by the distance to sources and atmospheric degradation. A factor of about 3 is typical for traffic sources, whereas wood combustion is characterised by a factor < 1 (Hedberg et al., 2002). At the street stations the ratio was measured to about 2.7 in agreement with the fact that traffic is the main source. At Hvidovre the ratio was lower (about 1.6) showing that wood combustion is a more important source in the suburbs compared to the streets. Moreover, for this reason the toluene/benzene ratio is not constant throughout the year, but lowest in the heating season, where benzene from wood combustion adds to the ambient benzene concentration.

Table 6.1. Annual statistics for benzene in 2014 based on weekly average concentrations at 1 atm. and 293 K. The limit value is based on the EU Directive 2008/50/EC (EC, 2008).

Concentration μg/m^3	Number of results	Average	Max. weekly average
Copenhagen/1103	52	1.1	2.1
Copenhagen/1257	51	1.0	2.2
Limit value		5	

Table 6.2. Annual statistics for toluene in 2014 based on weekly average concentrations at 1 atm. and 293 K. The Maximum weekly average is the maximum value for the weekly measurements (WHO, 2000). The guideline value is established by WHO (WHO, 2000).

Concentration μg/m^3	Number of results	Average	Max. weekly average
Copenhagen/1103	52	2.8	4.7
Copenhagen/1257	51	2.8	4.5
Guideline value	-	-	260

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

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:

Benzene = 0.0044·CO - 0.37

where benzene and CO are in units of µg/m^3.

Based on this and the concentrations of CO (table 5.1) the annual average concentrations of benzene is estimated to about 1 µg/m^3 for all the three street stations in Aarhus, Odense and Aalborg in 2014.

6.2 Trends

Benzene has decreased from approximately 6 µg/m^3 on Jagtvej (Copenhagen/1257) in 1998, to a value below the lower assessment threshold (EC, 2008) of 2 µg/m^3 since 2007. In 2014 the annual averages were 1.0 - 1.1 µg/m^3 at the street stations in Copenhagen. Toluene shows a similar trend, which indicates that a significant fraction of benzene and toluene is emitted from traffic on these locations. Annual averages for toluene were 2.8 µg/m^3 at both kerbside stations. The main reasons for the significant decrease of benzene and toluene up to 2008 are reductions of the emissions from gasoline-fuelled traffic due to increased use of catalysts and higher ratio of diesel cars.

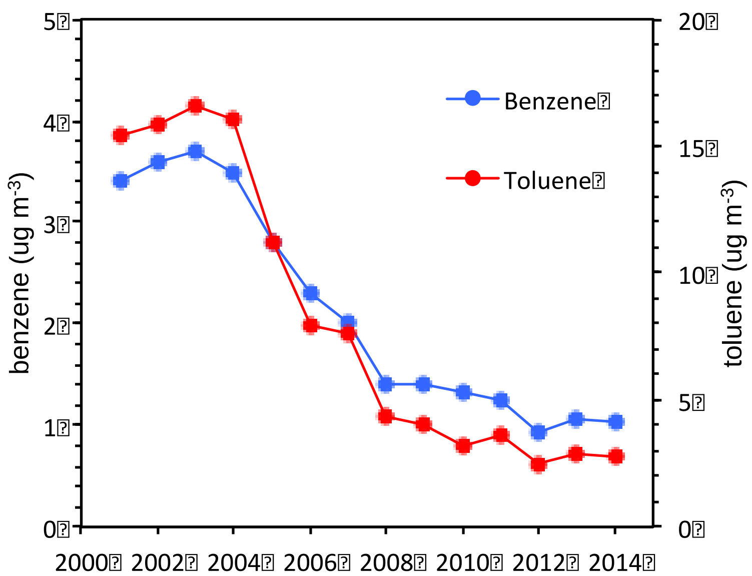


Figure 6.1. Annual average concentrations of benzene and toluene on the kerbside station Jagtvej, Copenhagen/1257.

7 Particles (TSP, PM_{10}, $PM_{2.5}$ and particle number)

The SM200 sampler manufactured by OPSIS, Sweden, has been used in Denmark to measure PM_{10} in accordance with the EU Directive (EC, 2008). Measurements with this instrument have from 2007 been extended to also include $PM_{2.5}$. 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_{10} given in the EU Directive, than the results from weighing of the filters using the SM200 as a filter sampler for PM_{10} (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.

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_{10} and $PM_{2.5}$ (EN 12341: 2014, into which the previous standards for PM_{10}, EN 12341: 1998, and for $PM_{2.5}$, 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^3/hr with following gravimetric determination of the sampled mass in the laboratory. Results from comparison of LVS gravimetric determination and the SM200 β-method for PM measurements has not documented any systematic deviation between the two measuring methods except for an improved reproducibility using the LVS instruments.

During the course of 2012-14 LVS 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 mesurement method (i.e. either SM200 or LVS) does not cover a full calender year consists of a combination of SM200- and LVS measurements.

Initiation of LVS measurements:

2012

HCAB PM10 (Copenhagen traffic): September 5th 2012

HCAB PM2.5(Copenhagen traffic): September 12th 2012

Jagtvej PM10 (Copenhagen traffic): August 28th 2012

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

Risø PM2.5 (Rural): August 29th 2012

2013

HCØ PM10 (Copenhagen urban background): June 26th 2013

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

2014

Risø PM10 (Rural): January 4th 2014

Aarhus PM10 (traffic): September 10th 2014

Aahus PM2.5 (traffic): September 10th 2014

Aahus PM2.5 (urban background): January 1th 2014

Aalborg PM2.5 (traffic): April 9th 2014

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

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 particle with mobility diameter between 6 and 700 nm.

7.1 Annual statistics

At all the PM_{10}- and/or $PM_{2.5}$ 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 2014 the permitted number of exceedences in a year of the diurnal limit value of 50 $\mu g/m^3$ for PM_{10} was not exceeded at any stations in the measuring network, even at stations where exceedances previously has occurred (the two traffic stations in Copenhagen (HACB/1103 and Jagtvej/1257)).

There were likewise no exceedances of the annual limit value for PM_{10} (of 40 µg/m³) and $PM_{2.5}$ (of 25 µg/m³ [from 2015]) 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 $PM_{2.5}$. For the years 2012-2014 the AEI is determined to 11 µg/m³. In Denmark the average exposure indicator is measured in urban background at Copenhagen/1259, Århus/6159 and Aalborg/8158.

In 2014 the number of particles in ambient air was about 14.000 particles per cm³ at the street station H.C. Andersens Boulevard (table 7.5). This is a factor of roughly about 3 and 5 higher than in urban and rural background, respectively.

Table 7.1. Annual statistics for PM_{10} in 2014. All parameters are based on diurnal averages at ambient temperature and pressure.

Unit µg/m³	Number of results	Average	Median	Days above 50 µg/m³	90 percentile	Max. day
Traffic						
Copenhagen/1103[1]	359	30	27	26	46	77
Copenhagen/1257[1]	354	26	23	23	43	95
Århus/6153[2]	337	25	23	14	40	103
Odense/9155[2] §	364	22	20	7	37	75
Urban background						
Copenhagen/1259[1]	352	19	16	3	32	65
Rural						
Risø[1]	350	17	14	3	31	66
Keldsnor/9055[2]	313	22	19	9	36	84
Limit value (2005)	>328**	40		35***		

1 Measurements based on LVS with gravimetric determination of particle mass
2 Measurements based on SM200 beta gauge determination of particle mass

* Measurements is a combination of LVS gravimetric and SM200 beta gauge determination of particle mass

** 90% data capture of number of diurnal measurements in relation to number of days in 2014

*** Permitted number of exceedances in a year of the diurnal limit value of 50 µg/m³

§ The site in Odense/9155 (Albanigade) was affected by a greater permanent rearrangement of the roads in Odense changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on June 28th. 2014

Table 7.2. Annual statistics for $PM_{2.5}$ in 2014. All parameters are based on diurnal averages at ambient temperature and pressure.

Unit µg/m³	Number of results	Average	Median	90 percentile	Max. day
Traffic:					
Copenhagen/1103[1]	358	18	15	30	69
Copenhagen/1257[1]	354	17	14	31	67
Århus/6153*	303	15	13	27	54
Aalborg/8151* §	244	14	12	24	57
Urban background:					
Copenhagen/1259[1]	346	13	10	23	57
Århus/6159[1]	339	12	10	23	50
Aalborg/8158*	345	12	9	21	52
Rural:					
Risø[1]	**342**	**12**	**9**	**24**	**61**
Limit value (2015) (parenthesis gives proposed value for 2020)	>328**	25(20)			

[1] Measurements based on LVS with gravimetric determination of particle mass.

[2] Measurements based on SM200 beta gauge determination of particle mass.

* Measurements a combination of LVS gravimetric and SM200 beta gauge determination of particle mass.

**90% data capture of number of diurnal measurements in relation to number of days in 2014.

§ Aalborg/8151 (street) is only represented by data covering the period 1/1 - 7/9 as the station was closed due to construction work at the site.

Table 7.3. Annual statistics for PM_{10} measured in 2014 using TEOM. The values are based on ½-hourly averages. Total annual number of ½-hours is 17.520.

Unit: µg/m³	Number of results	Average	Average x 1.3
***Traffic**:*			
Copenhagen/1103	12088	29	37
Rural:			
Risø	9811	15	20
Limit value			40

Table 7.4. Annual statistics for $PM_{2.5}$ measured in 2014 using TEOM. The values are based on ½-hourly averages. Total annual number of ½-hours is 17.520.

Unit: µg/m³	Number of results	Average	Average x 1.3
Traffic:			
Copenhagen/1103	10162	14	19
Rural:			
Risø	11388	10	13
Limit value (2015) (parenthesis gives proposed value for 2020)			25(20)

Table 7.5. Annual statistics for particle number measured in 2014 using DMPS. All values are based on ½-hourly averages. Total annual number of ½-hours is 17520.

Unit: particles per cm^3	Number of results	Average
Traffic:		
Copenhagen/1103	14717	13957
Urban Background:		
Copenhagen/1259	12789	4506
Rural:		
Risø	14717	2956

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 was changed from TSP to PM_{10} according to the EU directive adopted in 1999 (EC, 1999) and PM_{10} 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_{10} 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_{10} 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_{10} at this station. However, this is mainly due to a major reduction (7 µg/m^3) in PM_{10} 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 greater permanent rearrangement of the roads in Odense changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on June 28th. 2014. This has affected the measured PM_{10} levels in the second half of 2014 and this is the reason why there is unchanged PM_{10} value for Odense/9155 in 2014 while all the other traffic stations displays an increase in 2014 compared to 2013.

The measurements of $PM_{2.5}$ started in 2007 at Copenhagen/1103 and at the other stations in 2008. figure 7.3 presents all the results from measurements of $PM_{2.5}$ until now. There seems to be a tendency to a small reduction in $PM_{2.5}$, although this tendency is uncertain due to the relatively short period with measurements.

Values of the AEI (the average exposure index for $PM_{2.5}$) have been determined over the period to:

2008-2010: 14 µg/m^3;

2009-2011: 15 µg/m^3;

2010-2012: 14 µg/m^3;

2011-2013: 13 µg/m^3;

2012-2014: 11 µg/m^3.

Like for $PM_{2.5}$ in general there seems to be a tendency to a small reduction in the AEI, although this tendency is uncertain due to the relatively short period with measurements.

The measurements show a significant reduction of particle number in ambient air (figure 7.4) over the entire measuring period. On HCAB the number of particles has decreased by a factor of about 2 during the period 2002-2014. At the urban background station (HCØ) and rural background station (LVBY/Risø) a reduction in particle numbers was also observed though the decrease is smaller than at HCAB. The decreases are only about 30% at HCØ and LVBY/Risø, respectively.

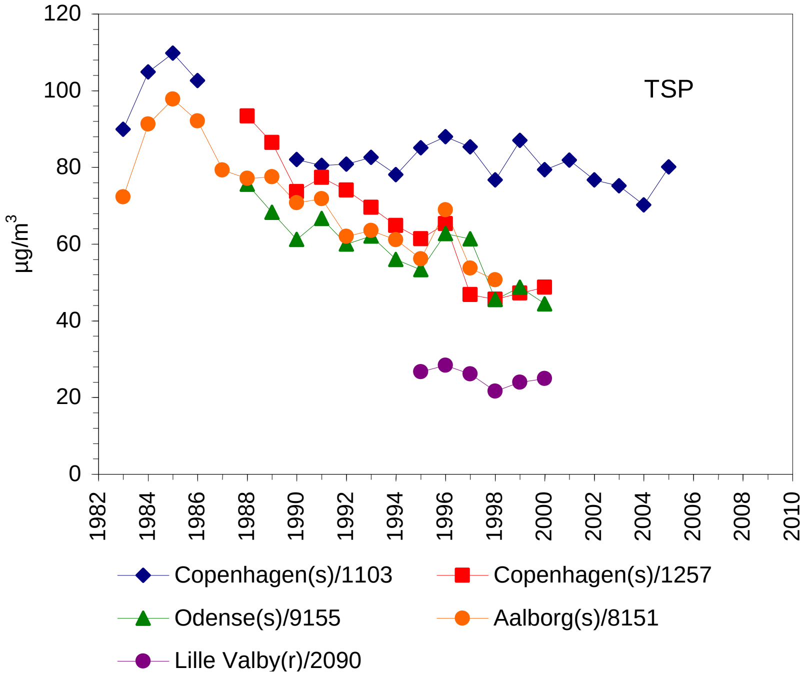


Figure 7.1. Annual averages for TSP measured at street stations (s) and at rural background station (r).

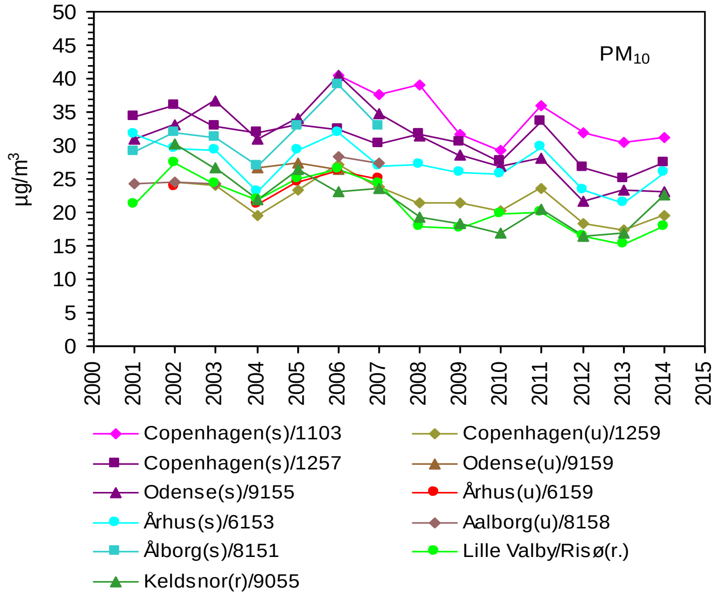


Figure 7.2. Annual averages for PM_{10} measured at street stations (s), urban background stations (u) and at rural background stations (r). The change from gravimetric determination using the SM200 as a filter sampler to the use of the same instrument as a β-gauge from 2006 gives rise to a 5-10% increase due to the shift of method. The value for PM_{10} 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 (0ºC 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 due to a greater permanent rearrangement of the roads in Odense changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on June 28th. 2014. This is the reason why there is a unchanged PM_{10} value for Odense/9155 in 2014 while all the other street stations displays an increase for 2014 compared to 2013.

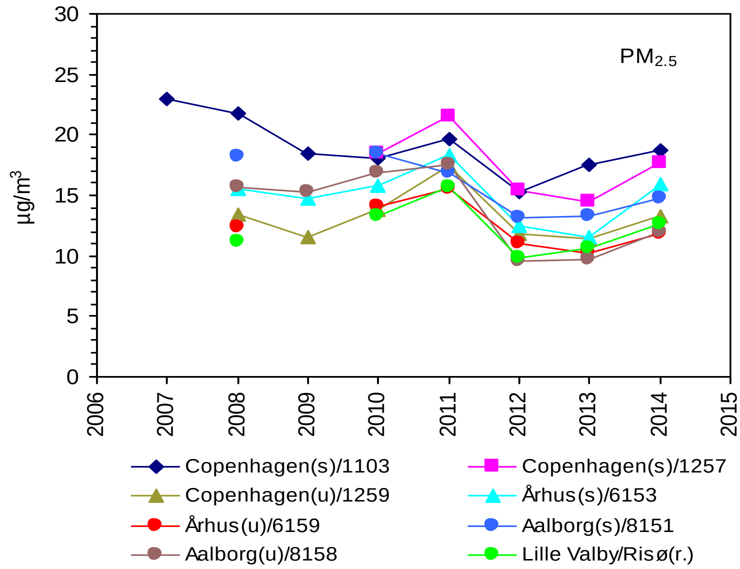


Figure 7.3. Annual averages for $PM_{2.5}$ measured at street stations (s), urban background stations (u) and at rural background station (r). Only annual averages covering more than 2/3 of the years are shown. Data are given at standard temperature- and pressure conditions (0ºC and 1 atm.). PM given at ambient temperature and pressure conditions is on an annual average approximately 3-4% lower than PM-results given at standard conditions. Aalborg(s) is only represented by data covering the period 1/1 - 7/9.

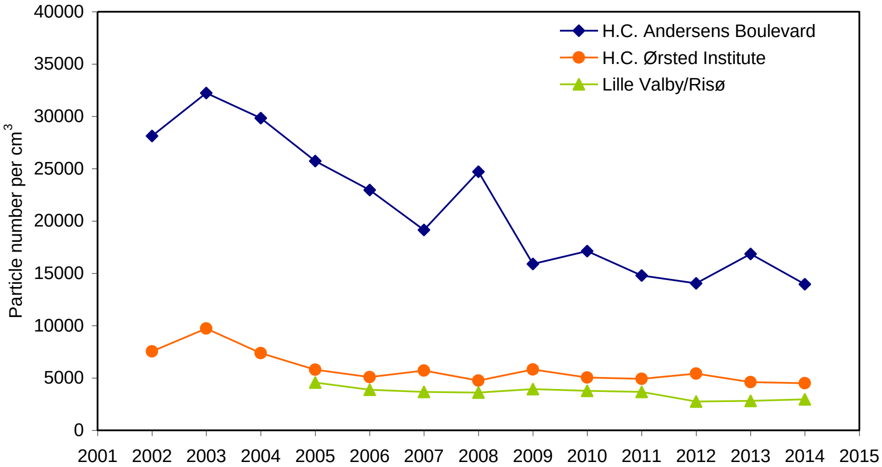


Figure 7.4. Annual averages for particle number at street level (H. C. Andersens Boulevard), urban background (H.C. Ørsted Institute) and rural background (Lille Valby/Risø)..

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_{10} (Article 20 and 21). Salt from sea salt can be subtracted from PM_{10} 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_{10}. These rules account for both the annual limit value and the daily limit value that states that the daily PM_{10} concentration must not exceed 50 µg/m³ 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 2014.

	Na µg/m³	NaCl µg/m³
Traffic:		
Copenhagen/1103	1.0	2.7
Odense/9155	1.1	2.8
Aarhus/6153	1.3	3.3
Urban Background:		
Copenhagen/1259	0.9	2.2

Figure 7.5 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.

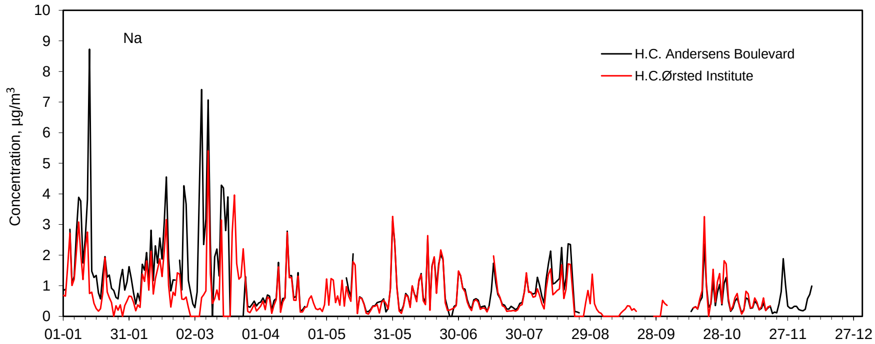


Figure 7.5. Diurnal concentrations in 2014 of sodium at H.C. Andersen's Boulevard, Copenhagen (1103) and at urban background in Copenhagen (H.C. Ørsted Institute/1259).

In 2014 the permitted number of exceedances in a year of the diurnal limit value of 50 µg/m³ for PM_{10} was not passed at any stations in the measuring network and therefore it has not been necessary to correct PM_{10} for the content of NaCl due to sea salt and winter salting of the roads.

8 Heavy Metals

Heavy metals in PM_{10} is measured by collection of PM_{10} 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_{10} and TSP are approximately equal since these metals are mainly found in the fine particle fraction.

The ICP-MS analysis provides the measurements of As, Cr and Ni included in the EU Directive 2004/107/EC (EC, 2005) and Pb included in EU Directive 2008/50/EC (EC, 2008). According to the directive (EC, 2005) also Hg has to be measured, however, these measurements can be carried out in cooperation with neighbouring 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), Cupper (Cu), Zink (Zn), Arsenic (As),Selenium (Se), Cadmium (Cd) and Lead (Pb) measured in PM_{10} during 2014. 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^3	**V**	**Cr**	**Mn**	**Ni**	**Cu**	**Zn**	**As**	**Se**	**Cd**	**Pb**
PM_{10}, Traffic:										
Copenhagen/1103	3.1	8.1	22	3.2	80	45	1.1	0.8	0.1	5.1
Odense/9155	2.0	2.8	12	2.5	27	30	0.9	0.9	0.1	4.5
Aarhus/6153	2.4	3.7	8.7	3.4	35	25	1.3	0.7	0.1	5.4
PM_{10},Urban background:										
Copenhagen/1259	2.5	1.4	7.0	2.7	9.6	17	0.7	0.7	0.1	3.4
TSP, Rural Background										
Risø	1.9	0.7	3.4	1.5	2.5	11	0.7	0.6	0.1	3.5
EU Target (Limit) Values *				20			6		5	500
Guideline value (WHO)**	1000		150						5	
Life time risk level at 1:10^5				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.

Table 8.2. Annual statistics for Mercury 2014. Measured at Råö in southern Sweden by the Swedish Environmental Research Institute.

Unit: ng/m^3	Total Gas Hg (ng/m^3)	Total Particles Hg (ng/m^3)
Råö (SE00014)	1.5	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 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 Cu in brakes.

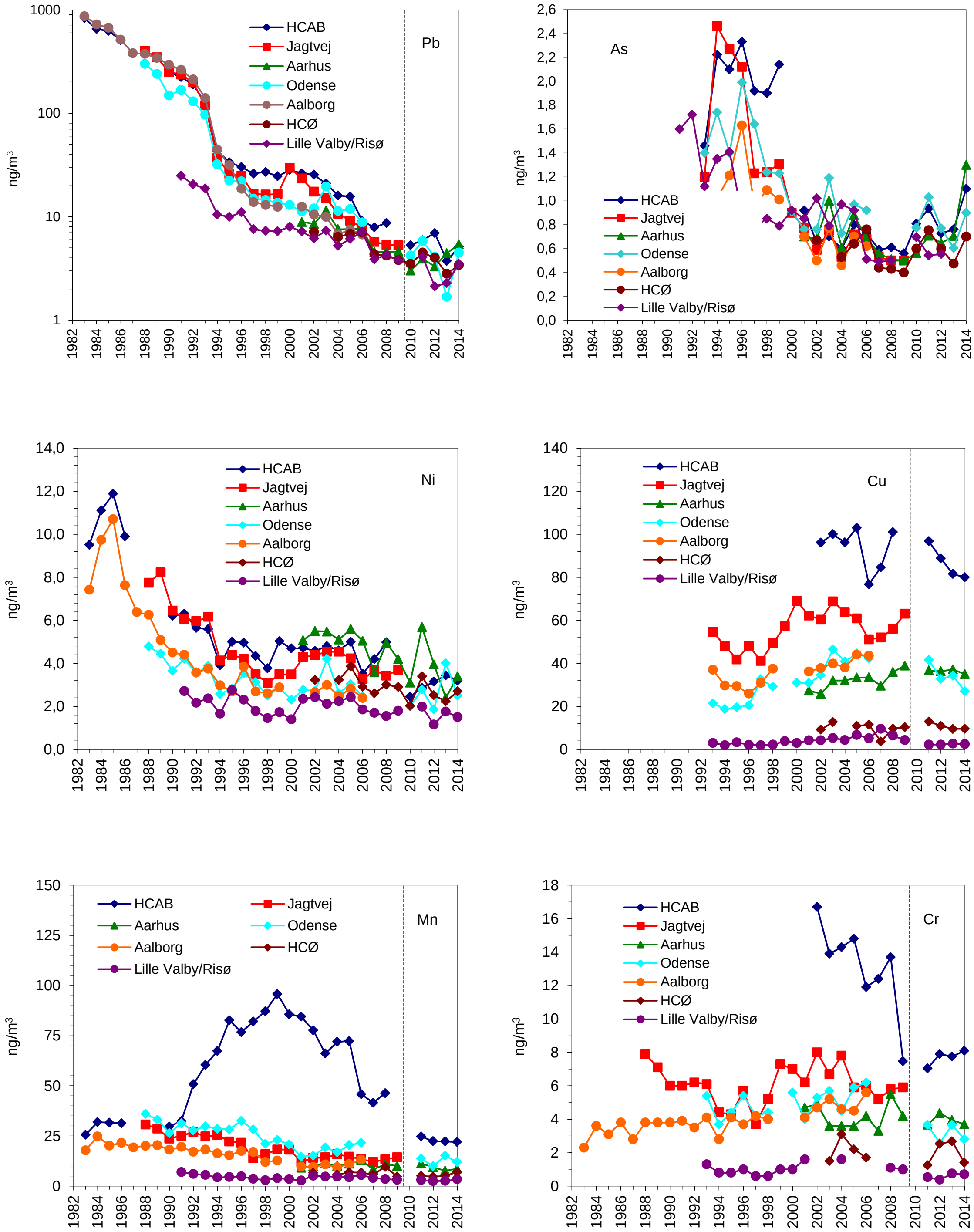


Figure 8.1. Annual averages from selected stations for some heavy metals in particulate matter. Until 2000 in TSP and later in PM_{10} – except for Copenhagen/1103 where PM_{10} replaced TSP from the beginning of 2006. The heavy metals are usually found in fine particles, which make the TSP and the PM_{10} 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 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 2014. In 2014 there was no information to the public due to exceedance of the alert threshold for SO_2 (one hour average 500 µg/m^3).

Table 9.1. Annual statistics for SO_2 in 2014. All parameters are calculated based on hourly average. The detection limit for the monitors is a few µg/m^3, which makes the average and median values encumbered with high relative uncertainties.

Unit: µg/m^3	Number of results	Average year	Average winter	Median	98-percentile	Max. Hour	4th highest diurnal mean
Traffic:							
Copenhagen/1103	8203	2.3	2.6	1.8	8.4	19.9	9.3
Aalborg/8151 §	5476	2.0	1.9	2.0	5.4	13.3	5.6
Limit values	>7467*	20	20			350	125

*) 90% data capture of number of hourly measurements in relation to total number of hourly measurements in 2014 excluding hours used for calibration.

§) Aalborg/8151 (traffic) is only represented by data covering the period 1/1 - 8/9 as the station was closed due to construction work on the site.

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 with 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.

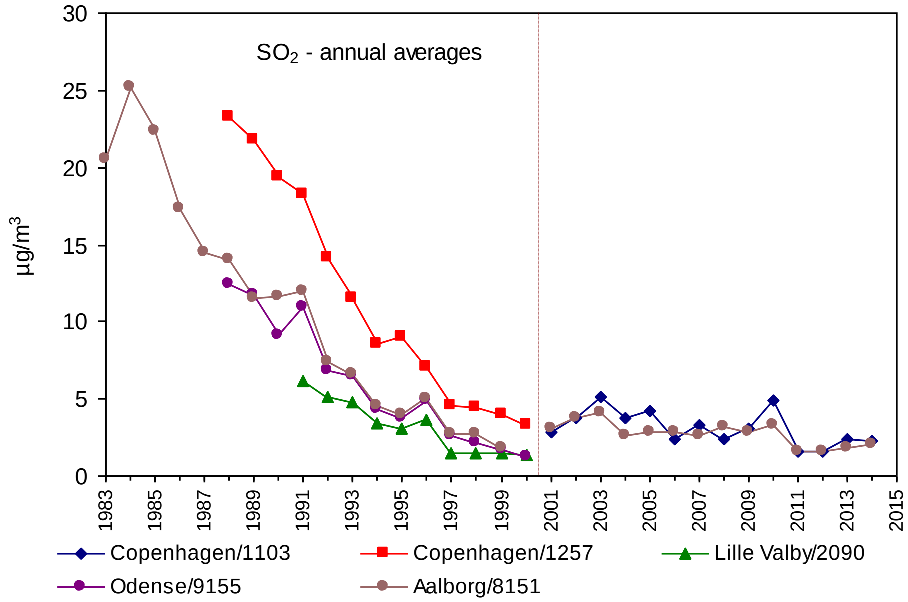


Figure 9.1. Annual averages for SO_2. Until 2001 the results were obtained using KOH impregnated filters for collection of SO_2. These measurements ceased in 2000. After 2000 the SO_2 measurements have been carried out using SO_2 monitors in order to monitor episodic results. The detection limit for the monitors is a few µg/m^3, 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.

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^3 averaged over a calendar year (EC, 2005). Benzo[a]pyrene is used as a marker for the carcinogenicity of PAHs.

Particulate matter (PM_{10} 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^3 min^{-1} over a period of 24 hours for an average total volume of 700 m^3. 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^2 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 2014 was 0.29 ng/m^3 and 0.38 ng/m^3 at the street station on HCAB and suburban station in Hvidovre, respectively. The higher concentrations in Hvidovre are due to local wood burning for house hold heating. The minimum, maximum and average monthly concentrations of benzo[a]pyrene are summarized in table 10.2 and 10.3. The average annual concentrations of the other five PAHs listed as relevant in the EU Directive are shown in table 10.1.

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

	HCAB Ng/m^3	Hvidovre Ng/m^3
Benzo[a]pyrene	0.29	0.38
Benzo[a]atracene	0.27	0.27
Benzo[b]fluoranthene	0.49	0.65
Benzo[j+k]fluoranthenes	0.44	0.90
Indeno[1,2,3-cd]pyrene	0.29	0.35
Dibenzo[a,h]anthracene	0.09	0.05

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. It can be concluded that the target value for benzo[a]pyrene on 1 ng/m^3 was not exceeded in 2014.

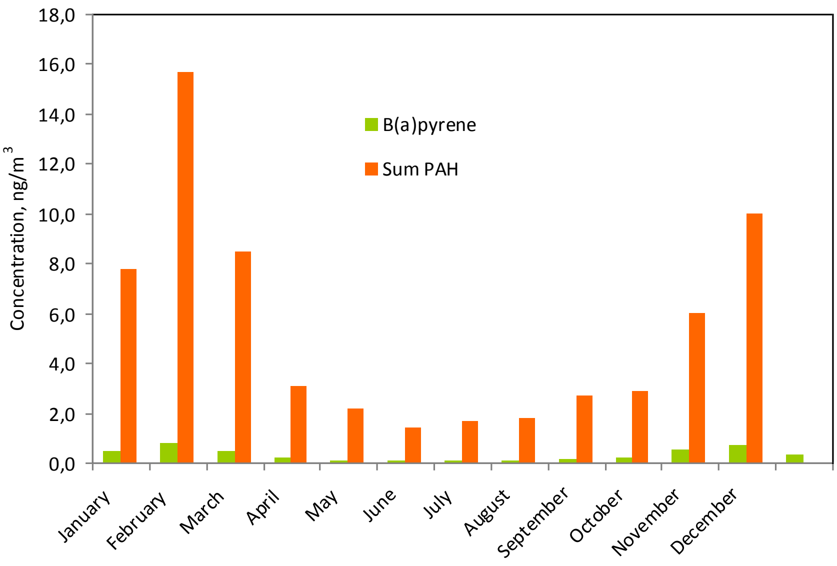


Figure 10.1. Monthly average concentrations in 2014 of benzo[a]pyrene and the sum of the analysed PAHs in Copenhagen (HCAB).

Table 10.2. Weekly minimum, maximum and average monthly concentrations (ng/m^3) of benzo[a]pyrene during 2014 in Copenhagen (HCAB).

Month	Minimum conc.	Maximum conc.	Average conc.
January	0.13	0.78	0.45
February	0.27	1.86	0.79
March	0.07	1.05	0.44
April	0.10	0.24	0.16
May	0.07	0.12	0.09
June	0.05	0.07	0.06
July	0.05	0.16	0.08
August	0.05	0.13	0.08
September	0.11	0.16	0.14
October	0.11	0.20	0.17
November	0.04	0.67	0.48
December	0.33	1.30	0.70
Annual	0.04	1.55	0.29

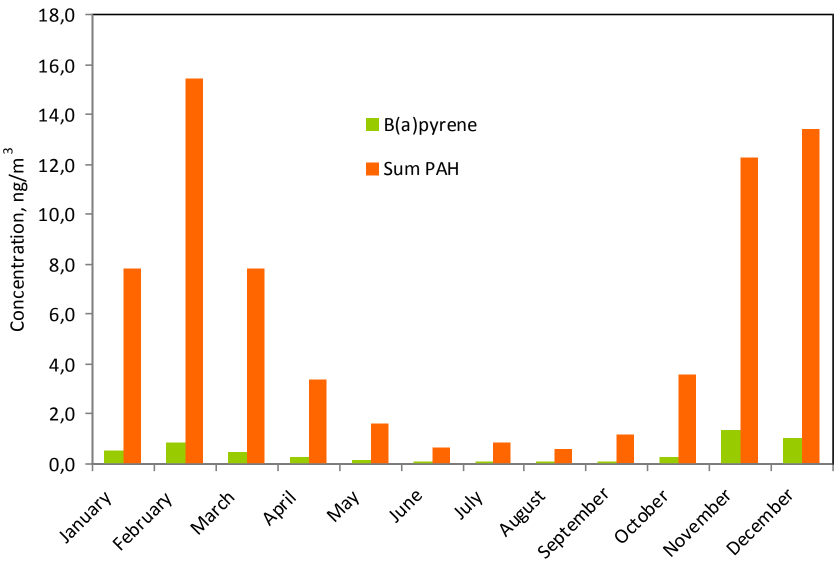


Figure 10.2. Monthly average concentrations in 2014 of benzo[a]pyrene and the sum of the analysed PAHs in Hvidovre.

Table 10.3. Daily minimum, maximum and average monthly concentrations (ng/m^3) of benzo[a]pyrene during 2014 in Hvidovre.

Month	Minimum conc.	Maximum conc.	Average conc.
January	0.18	0.65	0.49
February	0.20	1.96	0.84
March	0.07	1.10	0.47
April	0.08	0.51	0.24
May	0.08	0.22	0.12
June	0.06	0.07	0.06
July	0.05	0.09	0.07
August	0.04	0.06	0.05
September	0.07	0.10	0.09
October	0.21	0.35	0.26
November	0.33	2.11	1.32
December	0.22	2.02	1.00
Annual	0.04	4.27	0.38

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.

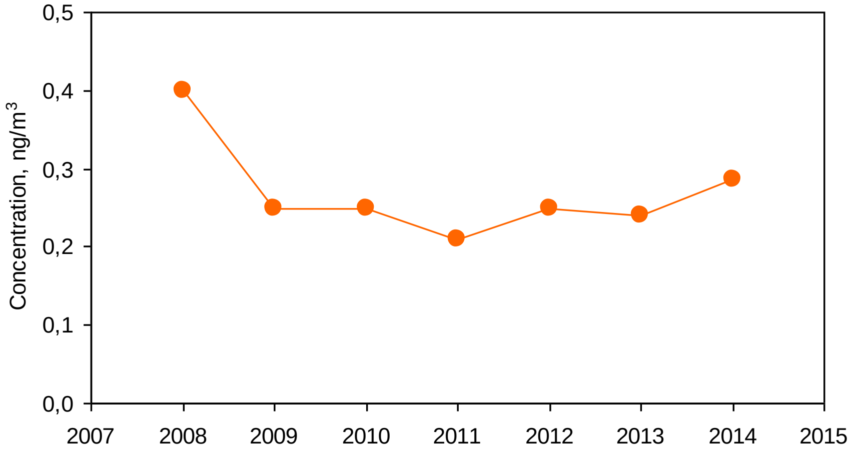


Figure 10.3. Annual average concentrations of benzo[a]pyrene at H.C. Andersens Boulevard (Copenhagen/1103).

11 Organic carbon and elemental carbon

Ambient concentrations of particulate organic carbon (OC) and elemental carbon (EC) are measured on the street station H.C. Andersens Boulevard/1103 in Copenhagen and the semi-rural background station Risø North of Roskilde based on the requirements in the EU air quality directive (EC, 2008). $PM_{2.5}$ 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.

11.1 Annual statistics and trends

The measurements of organic carbon (OC) and elemental carbon (EC) were initiated in 2009. The ratio of EC to total carbon (TC) differ markedly when comparing rural background and the street station in Copenhagen/1103: In the last 5 years EC amounted to 37-49% of the total particulate carbon at kerbside compared to about 16-20% in rural background calculated as annual averages (figure 11.1, table 11.1 and 11.2).

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

Table 11.1. Annual statistics for OC in 2014. The values are based on daily averages at H. C. Andersens Boulevard and in semi-rural background.

Concentration µg/m^3	Data capture	OC, average.	90% percentile
Copenhagen/1103	99%	3.1	4.1
Risø	98%	1.8	5.2

Table 11.2. Annual statistics for EC in 2014. The values are based on daily averages at H.C. Andersens Boulevard and in semi-rural background.

Concentration µg/m^3	Data capture	EC, average.	90% percentile
Copenhagen/1103	99%	1.9	2.8
Risø	98%	0.3	0.7

EC (black lines) in the rural background and at the kerbside station in 2014 show only little deviation from the 2013 concentrations, while decreasing concentrations of EC has generally been observed from 2011 - 2013 at the Kerbside station. On the other hand, OC has increased 20 -30% from last year in rural background and at the kerbside station in Copenhagen. Generally, the largest contributor to ambient OC is atmospheric oxidation of biogenic compounds.

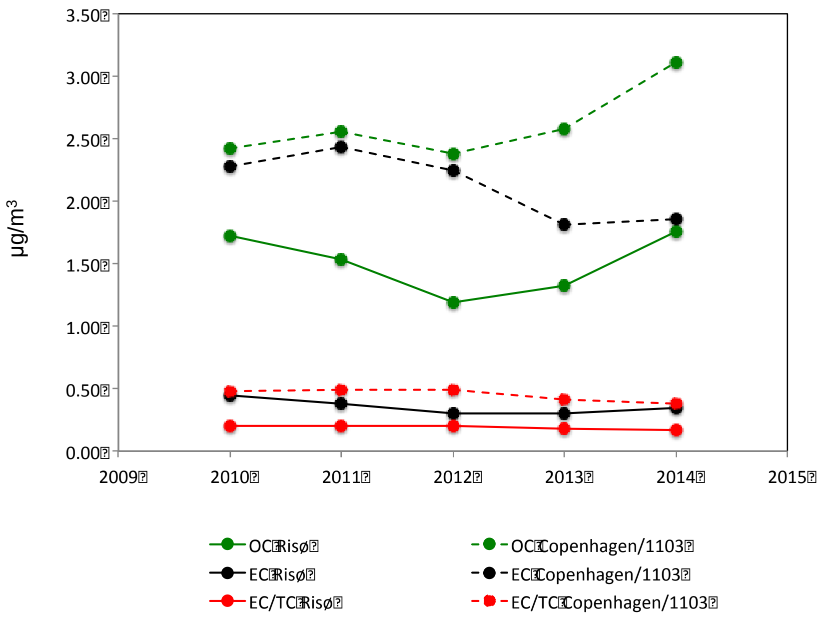


Figure 11.1. Elemental carbon (EC), organic carbon (OC) and the ratio between elemental carbon and total carbon (EC/TC) at H.C. Andersens Boulevard (Copenhagen/1103) and in semi-rural background at Risø 2010 - 2014.

12 Chemical composition of $PM_{2.5}$

In addition to the measurements of elemental and organic compound, there has also been carried out measurements of the main inorganic compounds in $PM_{2.5}$ (NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_3^-, SO_4^{2-}) 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 metehod is chemical analysis of the daily $PM_{2.5}$ particle filters sampled using the SM200 monitors.

Examples on the daily variations of the concentrations are shown in figure 12.1 together with the variation of $PM_{2.5}$. For Na^+ the concentrations are similar at HCAB and Risø due to long range transport of sea salt. For the winter months Na^+ is higher at HCAB than Risø due to winter salting of the roads in Copenhagen. The variations of Cl^- follow the variations of Na^+ because the main source is sea salt and winter salting. Mg^{2+} originates only from sea salt and there are therefore similar concentrations at the two stations throughout the year. SO_4^{2-} and 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 NO_3^- and K^+. 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 $PM_{2.5}$ of the different compounds are shown in figure 12.2. The annual average concentrations of NH_4^+, Na^+, K^+, Mg^{2+}, Cl^-, NO_3^-, 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 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 $PM_{2.5}$ and the sum of all the analysed constituents. The unknown mass is water attached to the particles, dust (e.g. SiO_2), heavy metals and other trace constituents.

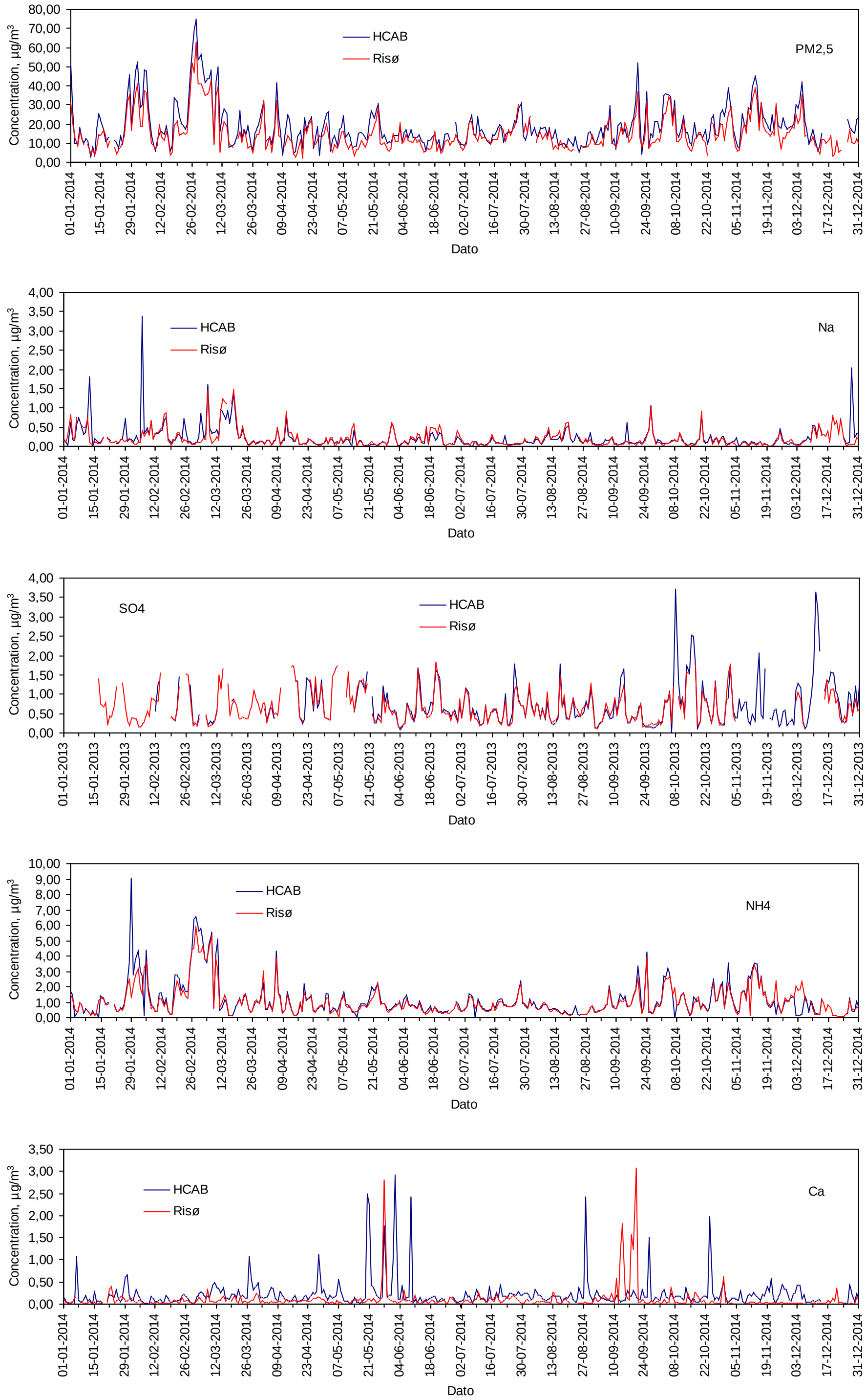


Figure 12.1. Daily variations of the concentrations of $PM_{2.5}$, Na^{+}, SO_4^{2-}, NH_4^{+} and Ca^{2+} at H.C. Andersens Boulevard (HCAB, Copenhagen/1103) and Risø in 2014. The large gap in data for calcium on Risø is due to problems with filter material.

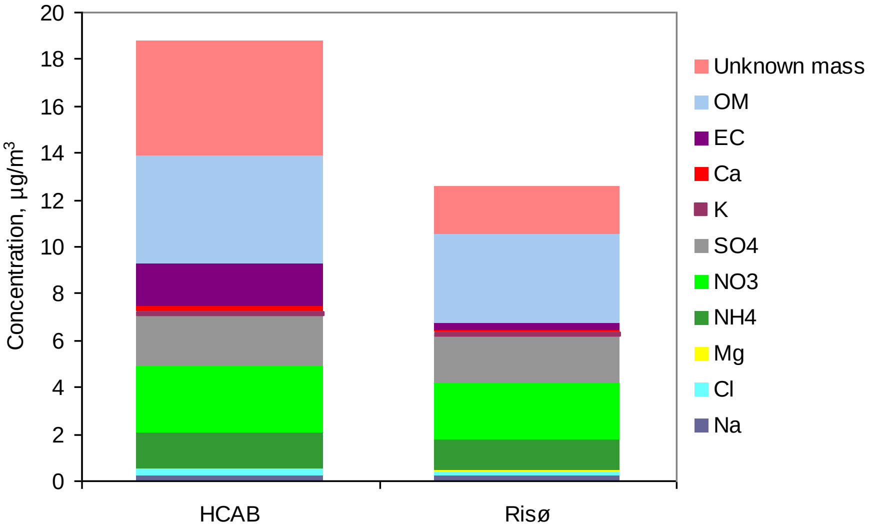


Figure 12.2. Annual average contributions to the chemical composition of $PM_{2.5}$ at H.C. Andersens Boulevard (HCAB, Copenhagen/1103) and Risø in 2014. Organic matter (OM) has been estimated from the measured concentrations of OC (organic carbon) by multiplication of OC with a factor of 1.5 for the 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.

13 Ozone precursors

Measurements of mainly anthropogenic volatile organic compounds in urban background, which may act as ozone precursors, were initiated in 2009 at the urban background station HCØ. Ambient air is sampled as 24-hour averages on adsorbent tubes packed with Carbopack X and analysed using Thermal Desorption Gas Chromatography Mass Spectrometry. 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_5-C_7 alkanes: pentane, 2-methylpentane hexane and heptane. The more reactive unsaturated compounds are less abundant.

13.1 Annual statistics

The urban background concentration of the major ozone precursors benzeneand toluene correspond to 51% and 32% of the corresponding concentrations at the kerbside station 1257, respectively. The urban background ratio between toluene and benzene is somewhat smaller than the ration at the traffic dominated kerbside stations, i.e. 1.7 versus 2.7 (1257) and 2.6 (1103). This reflects a faster atmospheric decomposition of toluene, and different sources to benzene and toluene, i.e. the kerbside stations are more dominated by traffic.

Table 12.1. Annual statistics for selected ozone precursors in urban background in Copenhagen (1259) based on daily average concentrations at 1 atm. and 293 K.

Concentration µg/m^3	Average concentration	90% Percentile	Number of results
1-Pentene	0.04	0.06	326
n-Pentane	0.86	1.76	329
Trans-2-pentene	0.02	0.02	328
Isoprene	0.04	0.09	329
2-Methylpentane	0.33	0.56	329
n-Hexane	0.18	0.31	329
Benzene	0.53	0.99	329
n-Heptane	0.22	0.38	329
2,2,2-Trimethylpentane	0.05	0.09	308
Toluene	0.89	1.45	329
n-Octane	0.05	0.09	329
Ethylbenzene	0.17	0.29	329
m,p-Xylene	0.47	0.85	329
o-Xylene	0.20	0.36	329
1,3,5-Trimethylbenzene	0.04	0.09	329
1,2,4-Trimethylbenzene	0.16	0.36	329
1,2,3-Trimethylbenzene	0.04	0.08	329
Σ	4.28	7.85	

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Appendix 1

Pollutants measured in the LMP Network

NO and partly NO_2 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_2 can cause respiratory problems.

Most of the NO_2 in the urban atmosphere is produced by oxidation of nitrogen monoxide (NO) by ozone (O_3). The reaction will take place immediately, if sufficient O_3 is present. O_3 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_2 is further oxidised to nitrate and/or nitric acid, which may cause acid precipitation and eutrophication. NO_2 is a toxic gas, which may cause respiratory problems. There are limit values for the allowed concentration of NO_2 in the atmosphere.

O_3 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_3 measured in Denmark originates from sources outside the country. Usually the highest concentrations are found at rural and urban background sites. O_3 is removed by NO at street level. O_3 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_3 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 Hydrocarbones).

The main sources for PM_{10} and $PM_{2.5}$ 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_{10} concentration in the atmosphere are implemented at present.

PM_{10} and $PM_{2.5}$ is measured using three different methods in the monitoriong 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 β-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 PM10 or PM2.5 mass concentration of suspended particulate matter in ambient air (EN 12341: 2014, into which the previous standards for PM_{10}, EN 12341: 1998, and for $PM_{2.5}$, 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_2) is formed by burning of fossil fuel and biomass. The SO_2 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_2 with one order of magnitude. SO_2 may cause respiratory problems. There are limit values for the allowed concentration of SO_2 in the atmosphere.

THE DANISH AIR QUALITY MONITORING PROGRAMME

Annual Summary for 2014

The air quality in Danish cities has been monitored continuously since 1982 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 2014 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_2 was found in concentrations above EU limit values while NO_2 levels in Odense, Aarhus and Aalborg were below the limit value. Model calculations indicate exceedances of NO_2 limit values at several streets in Copenhagen. Annual averages of PM_{10} and $PM_{2.5}$ were below limit values at all stations. The concentrations for most pollutants have been decreasing during the last decades.

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