THE DANISH AIR QUALITY MONITORING PROGRAMME

Annual Summary for 2018

Scientific Report from DCE – Danish Centre for Environment and Energy No. 360 2020

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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 2018 the air quality was measured in four Danish cities and at two background sites. In addition, model calculations of air quality and the impact of air pollution on human health and related external costs were carried out. For 2018, no exceedances of the NO_2 EU limit values were observed. Model calculations were carried out for 98 streets in Copenhagen and 31 in Aalborg. Only one exceedance of the limit value for the annual average of NO_2 was modelled for a busy street in Copenhagen. Annual averages of PM_{10} and $PM_{2.5}$ were below limit values at all stations and the average exposure indicator ($PM_{2.5}$ in urban background) has decreased with about 30 % since 2010. 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, health effects, external cost.
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Summary and Conclusion

This report presents the result from the Danish Air Quality Monitoring Programme in 2018. The monitoring programme is carried out by the Danish Centre for Environment and Energy (DCE) at Aarhus University. The core part of this program consists of continuous measurements at thirteen monitoring stations. Eight of these stations are located in the four largest cities, four stations are located in background areas and one station is located 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), mass of particles with diameters less than 10 and 2.5 micrometers respectively (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), nickel (Ni), and a number of volatile organic compounds (VOCs) that are precursors for formation of O_3. The measurements and model calculations are applied for evaluating the Danish air quality in relation to limit values as well as to follow trends. Furthermore, the obtained data are used for determination of sources of the air pollutants, as basis for evaluation of the impact of regulations of emissions and as basis for various research projects related to air quality.

The permitted number of exceedances in a year of the diurnal limit value of 50 $\mu g/m^3$ for PM_{10} was not exceeded at any station in the measuring network. Likewise, there were no exceedances of the annual limit values for PM_{10} (40 $\mu g/m^3$) and $PM_{2.5}$ (25 $\mu g/m^3$). The average exposure indicator (AEI) determined as a running three-years average of the average urban background concentration of $PM_{2.5}$ has decreased with about 30 % since 2010 and hence the target (15 % reduction) has been reached.

Due to technical difficulties with two new instruments, it has not been possible to measure the number of particles between 11 and 41 nm in 2017-2018. Therefore, the particle number represents the particle range from 41 to 478/559 nm (dependent on instrument version but the difference is negletable). The particle number in ambient air was about 4,000 particles per cm^3 as an annual average at the street station H.C. Andersens Boulevard. This is roughly a factor of two higher than in suburban areas and in urban and rural background. Since 2002, significant reduction of more than 40 % in particle numbers has been observed. This reduction has mainly been attained by reduction of traffic emissions (cleaner fuel, particle filters etc.).

The limit values for NO_2 was not exceeded at any of the monitoring stations in Denmark. Model calculations at selected streets in Copenhagen and Aalborg in 2018 showed that the annual average concentration at one single street segment in Copenhagen were slightly above the limit value (40 $\mu g/m^3$).

The annual average O_3 concentrations in 2018 were at the same level as in the previous years but the maximum 8-hours running mean concentration was

higher in 2018 compared to 2017. This change was due to differences in the meteorological conditions. No clear trend is observed for the average O_3 concentration. The information threshold of 180 µg/m^3 was not exceeded at any of the measurement stations in 2018. The target value for the maximum daily 8-hours mean O_3 concentration of 120 µg/m^3 was not exceeded, but the long-term objective for this parameter was exceeded at all Danish stations. The target value entered into force in 2010 while the long-term objective has not entered into force and the date for this has not yet been decided.

Measurements of VOCs at the urban background in Copenhagen showed concentration levels between 0.01 µg/m^3 and 0.91 µg/m^3 for the selected 17 different compounds. VOCs can act as O_3 precursors, and the aim of these measurements is to improve the general understanding of the O_3 formation at a European level. The formation of O_3 in Denmark is in general small due to moderate solar radiation. O_3 pollution in Denmark is mainly the result of long-range 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 been decreasing for the last decades.

Measurements of concentrations of particle bound PAH were performed at H.C. Andersens Boulevard, Copenhagen and at the suburban measurement station at Hvidovre. The average concentration of benzo[a]pyrene was 0.25 ng/m^3 and 0.28 ng/m^3 at H. C. Andersens Boulevard and Hvidovre, respectively. The target value for benzo[a]pyrene (1 ng/m^3) was not exceeded in 2018.

Due to minor revisions of the program measurements of the chemical content in $PM_{2.5}$ were only carried out at the rural background station at Risø. The concentrations were slightly higher in 2018 compared to 2017 as a consequences of the low precipitation in 2018. Low precipitation gives higher particulate concentrations.

Model calculations show that air pollution causes about 4,200 premature deaths in Denmark as average for 2016-2018 and a large number of other negative health effects. This is about 1,000 premature deaths more compared to the reporting for 2017. These higher numbers are due to a major update of the model systems and not due to an increase in the air pollution. About 1,220 (29 %) of the premature deaths are due to Danish emission sources while the remaining premature deaths are caused mostly by sources outside Denmark. The total health related external costs for Denmark have been calculated to 79 billion DKK as an average over the three years 2016-2018. This is more than a doubling of the external costs compared to the previous reporting. This higher number is mainly due to an increase in the economic value of a statistic life. The negative health effects and external costs have declined with about 38 % since 1988-1990. It should be noted that the calculation of health impacts and external costs are constrained with considerably uncertainties.

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 2018 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 otte målestationer placeret i de fire største danske byer og ved fire baggrundsmålestationer uden for byerne samt en station i et forstadsområde. Disse måleresultater suppleres med resultater fra modelberegninger udført med DCE's luftkvalitetsmodeller.

Formålet med programmet er at overvåge den luftforurening, som har betydning for befolkningens sundhed. Målingerne udføres i overensstemmelse med EU's luftkvalitetsdirektiver. I henhold til disse og under hensyntagen til øvrige danske behov måles koncentrationer af svovldioxid (SO_2), nitrogenoxider (NO_x/NO_2), massen af partikler med diametre mindre end 10 og 2,5 mikrometer (hhv. PM_{10} og $PM_{2,5}$), partikelantal, benzen (C_6H_6), toluen (C_7H_8), kulmonoxid (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 O_3. Målingerne og modelberegningerne anvendes til at vurdere, om EU's grænseværdier for luftkvalitet er overholdt. Rapporten beskriver endvidere udviklingen i koncentrationerne. Samtidigt tjener resultaterne fra måleprogrammet som grundlag for vurdering af effekt af reduktionstiltag. Og som grundlag for en række videnskabelige undersøgelser, blandt andet vurdering af små partiklers effekt på sundhed.

Der er fastsat grænse- og målværdier for flere af de målte stoffer. Grænseværdierne skal være overholdt fra 2005, 2010 eller 2015 alt efter hvilke stoffer, det drejer sig om. En detaljeret beskrivelse af gældende mål- og grænseværdier og deres gennemførelse i dansk lov findes i en bekendtgørelse fra Miljø- og Fødevareministeriet (2016). Bekendtgørelsen er baseret på det 4. datterdirektiv om tungmetaller og PAH'er (EC 2005) samt EU's luftkvalitetsdirektiv fra 2008 (EC 2008). En af de væsentligste ændringer i direktivet fra 2008 i forhold til de tre første datterdirektiver (1999, 2000 og 2002) er, at der i direktivet fra 2008 stilles krav om målinger af de fine partikler ($PM_{2,5}$), og at der med dette direktiv er indført en grænseværdi for $PM_{2,5}$, som skulle overholdes fra 2015.

I 2018 blev grænseværdierne for NO_2 ikke overskredet. Koncentrationerne af NO_2 målt på gadestationerne i 2018 var stort set på niveau med det der blev målt for 2017. Modelberegninger viser en lille stigning i koncentrationerne i NO_2 som følge af primært højere baggrundskoncentrationer. Endvidere indikerer modelberegningerne for udvalgte gader i København og Aalborg, at der på et enkelt gadesegment i København, hvor årsmiddelkoncentrationen var over grænseværdien for årsmiddelkoncentrationen (40 µg/m^3).

PM_{10} overholdt grænseværdien på 40 µg/m^3 som årsmiddelværdi på alle målestationer. Ligeledes var der ingen målestationer i måleprogrammet, hvor det tilladte antal overskridelser af den daglige middelværdi for PM_{10} (50 µg/m^3 må ikke overskrides mere end 35 gange årligt) blev overskredet.

$PM_{2,5}$ overholdt grænseværdien på 25 µg/m^3 som årsmiddelværdi på alle målestationer. AEI-værdien (average exposure indikator, som er defineret som middel af tre års gennemsnit af årsgennemsnittet af $PM_{2,5}$ i bybaggrund) er

faldet med omkring 30 % siden 2010. Dermed er målværdien (15 % reduktion siden 2010) fastlagt i EU-direktivet (EC 2008) allerede nået.

Grundet tekniske vanskeligheder med de nye måleinstrumenter har det ikke været muligt at udføre målinger af de små partikler i området fra 11– 41 nm og derfor er data for 2018 foreløbige. Derfor angives antallet af partikler for 2018 i intervallet fra 41 – 478/550 nm (øvre grænse afhænger af instrumenttype, men forskellen er ubetydelig). Antallet af partikler var omkring 4.000 partikler per cm^3 på gademålestationen H. C. Andersens Boulevard, hvilket er en faktor 2 højere end ved forstadsstationen Hvidovre og ved by- og landbaggrundsstationen hhv. H. C. Ørsted Instituttet og Risø. Siden 2002 har der været et fald på ca. 40 % i antal partikler med diameter mellem 41 – 478/550 nm. Faldet er blandt andet sket som følge af krav om partikelfilter på alle nye dieselkøretøjer.

Ozonkoncentrationerne i 2018 var på niveau med tidligere år. Der er ikke fastsat egentlige grænseværdier for O_3, men kun "målværdier" og "langsigtede mål" (hensigtsværdier). Der var i 2018 ingen overskridelser af ozonmålværdien for beskyttelse af sundhed. Målværdien for ozon trådte i kraft i 2010. Det planlagte langsigtede mål (120 $\mu g/m^3$) er endnu ikke trådt i kraft, og der er ikke taget beslutning om hvornår, dette skal ske. Såfremt dette mål havde været gældene, så ville det have været overskredet på tre bybaggrundsstationer, København (ved H. C. Ørsted Instituttet), Aarhus (lokaliseret ved den botaniske have) og Odense (på taget af Rådhuset). Tærsklen for information af befolkningen om høje ozonniveauer (timemiddel 180 $\mu g/m^3$) blev ikke overskredet i 2018.

De øvrige målte stoffer findes i koncentrationer under grænseværdierne, og for flere stoffer (fx benzen, svovldioxid og bly) er koncentrationerne faldet markant siden 1990.

Målinger af partikelbundet PAH blev fortaget på H.C. Andersens Boulevard i København. Middelværdien for benz[a]pyren var 0,25 ng/m^3 og 0,28 ng/m^3 på henholdsvis H. C. Andersens Boulevard og ved målestationen i Hvidovre. Målværdien på 1 ng/m^3 blev således ikke overskredet i 2018.

Målinger af 17 udvalgte VOC'er i bybaggrund i København viser koncentrationsniveauer, som spænder fra 0,01 $\mu g/m^3$ til 0,91 $\mu g/m^3$ i 2018. Disse VOC'er bidrager til den kemiske dannelse af O_3 på europæisk plan, og målingerne skal først og fremmest understøtte den generelle forståelse af ozondannelsen i Europa. I Danmark er størstedelen af de målte O_3–niveauer hovedsageligt resultat af langtransport af luftforurening fra centrale og sydlige dele af Europa.

Grundet revision i måleprogrammet blev målinger af det kemiske indhold i $PM_{2,5}$ i 2018 kun gennemført ved landbaggrundsmålestationen på Risø. Målingerne i 2018 ligger lidt højere end i 2017 grundet den lavere nedbørsmængde i 2018, hvilket fører til højere luftkoncentrationer.

Modelberegningerne af helbredseffekterne viser, at luftforureningen som gennemsnit for 2016-2018 er skyld i omkring 4.200 for tidlige dødsfald og en lang række andre negative helbredseffekter. Antallet af for tidlige dødsfald er omkring 1.000 højere end rapporteret for 2017. Dette højere antal skyldes en gennemgribende opdatering af modelsystemerne, som anvendes til bereg-

ning af helbredseffekter og eksterne omkostninger relateret til luftforureningen. Det højere antal er derfor ikke begrundet i højere luftforurening. Omkring 1.220 (29 %) af de for tidlige dødsfald skyldes danske kilder, mens resten hovedsageligt stammer fra det øvrige Europa. De eksterne omkostninger fra luftforurening beløber sig til omkring 79 milliarder kr. Dette er mere end en fordobling af de eksterne omkostningerne set i forhold til rapporteringen for 2017. Årsagen til dette er opdateringen af modelsystemet, hvor forøgelsen af værdisætningen af et statistisk liv spiller den væsentligste rolle for de øgede eksterne omkostninger. De negative helbredseffekter og de eksterne omkostninger er faldet med omkring 38 % siden 1988-1990. Det skal bemærkes at beregningerne af helbredseffekterne er behæftet med betydelige usikkerheder.

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 and food (Miljø- og Fødevareministeriet, 2016) that implements the EU directives on air quality in Denmark (EC, 2005; EC, 2008).

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

The program was revised in 2016. The majority of the revisions were implemented from January 2017 except for the modelling part of the program that has been extended, so that they now also include model calculations of the health impacts and the external costs of air pollution.

Since 2012 there have been some important changes for the measurements stations and methods. These are:

- Starting in August 2012 low volume samplers (LVS) for gravimetric determination of particle mass based on the reference method were introduced into the regular measuring program and gradually installed at the PM-stations in the network to replace some of the older SM200 instruments that needed to be renewed. See introduction to Chapter 7 for an overview.
- A new measurement station at a suburban area in Hvidovre was initiated in the beginning of 2013 with measurements of polycyclic aromatic hydrocarbons (PAHs) in relation to use of wood burning as residential heating. In June 2015, the measurement program in Hvidovre was supplemented with measurements of $PM_{2.5}$ by LVS, elementary (EC) and organic carbon (OC), particle number and nitrogen oxides (NO and NO_2).
- The urban background measurement station in Aarhus was in January 2015 moved to another position (Chapter 2.1).

- The street station in Aalborg had to be temporarily closed down from September 2014 and onwards due to nearby construction work (Chapter 2.1).
- At the street station in Albanigade in Odense there was a large decrease in daily traffic intensity from late June 2014 and the street was closed down for traffic in spring 2015. This change was due to major changes in the traffic patterns in Odense (section 2.1). A new street station was opened in 2016 in Odense at Grønnelykkevej (section 2.1).
- In October 2016 the measurement station at H. C. Andersens Boulevard was moved 2.7 m (corresponds approximately to the width of a traffic lane) further away from the inner traffic lane. The aim of this relocation is to compensate for the changes in traffic lanes in 2010 that moved the traffic closer to the measurement station. The data presented for 2016 (in plots) covers data from both the old and the new position. This report shows the full impact of the relocation of the measurement station.
- The model system used for the calculation of air quality, health impact and external costs related to air pollution has undergone major revision for this year's reporting.
- Data from the rural back ground station at Anholt and Ulborg has been included in this year's reporting.

In the following chapters, the results from measurements and model calculations for 2018 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, information and alert thresholds.

2. Measurements and model calculations

2.1 Measurements

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

The Danish measuring strategy is 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 as close as possible to the street station and is placed so that it is representative for the urban background pollution; meaning that its location 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 relatively short distance between street station and urban background station makes it possible to directly determine 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/).

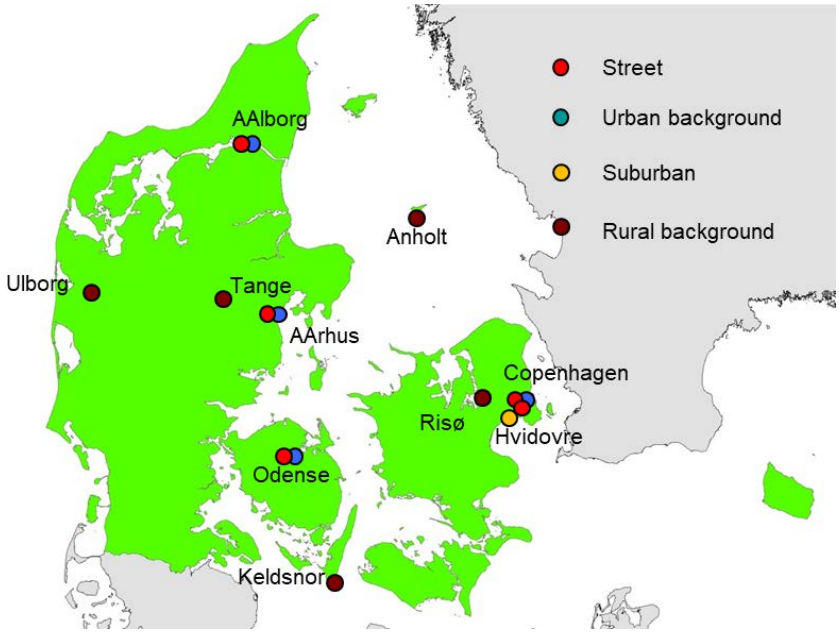


Figure 2.1. Main stations used for monitoring of air quality in relation to health.

Table 2.1. Main stations used in 2018 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, Fjeldstedvej	**Suburban**	**2650**
Odense		
Grønløkkevej	Street	9156
Town hall in Odense	Urban background	9159
Aarhus		
Banegårdsgade	Street	6153
Botanical Garden	Urban background	6160
Aalborg		
Vesterbro (not active)	Street	8151
Østerbro	Urban background	8150
Rural		
Lille Valby/Risø*	Rural background	2090
Keldsnor	Rural background	9055
Anholt	Rural background	6001
Ulborg	Rural background	7060

* 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 2014-2018 there were four major changes regarding the stations:

- The measurement station on Vesterbro at Limfjordsbroen in Aalborg was temporarily closed down on 8 September 2014 due to a major construction work at the nearby house. Therefore, the results for 2014 only represent data for 250 days (70 %). The station has finally been reestablished ultimo 2019 at a different site on Vesterbro. The reporting for 2018 includes therefore no data from this measurement station.
- In Odense a traffic plan has been adopted by the municipality for the entire city centre and the implementation of this plan began in late June 2014. This resulted in a major decrease in the traffic intensity at Albanigade, where the street station is situated. In spring 2015, Albanigade was closed for traffic. The station was shut down on 16 June 2015 and was moved to a new position at Grønnelykkevej in summer 2016 (figure 2.2).
- In January 2014, the urban background station in Aarhus moved to a new site since the municipality sold the house that the measurements station was placed upon and it was not any longer possible to carry on with the measurements. The new site is situated in the southeasterly part of the Botanical Garden that belongs to Aarhus University.
- On 3 October 2016 the station at H. C. Andersen Boulevard (HCAB) closed and a new station was placed nearby the old station (figure 2.2). The majority of the measurements were initiated on 19 October 2016. The new station is located 2.7 m further away from the inner traffic lane in order to compensate for the road change in 2010 (see Appendix 1 for a sketch of the location). Thus, it is possible to follow changes in the level of pollution in the street as measurements can be directly compared to previous years' measurements at HCAB. Moreover, the station was moved about 2 m parallel with the street further away from a tree close to the station. The EU

directive (EC, 2008) specifies that measurements have to be carried out several meters from trees in order to avoid influence of the trees on the measurements.

Figure 2.1. The old measurement station (left) at H.C. Andersen Boulevard closed down 3 October 2016. The new measurement station (right) began measurements 19 October 2016.

The following compounds were measured in 2018:

- Nitrogen oxides (NO, NO_2 and NO_x (= NO + NO_2)) were measured at all stations.
- Particle mass (PM_{10} and/or $PM_{2.5}$) as 24-hour averages, were measured throughout the year at all stations except at Aalborg/street ($PM_{2.5}$) were no data were measured in 2018 due to relocation of the station and at the urban background station Odense Town hall, where PM measurements has not been performed since primo 2007. At all the PM sites for 2018, PM was measured using low volume samplers (LVS) for gravimetric determination of particle mass according to the reference method EN 12341: 2014.
- Elements (heavy metals) in PM_{10} were measured at Copenhagen/street (HCAB), Copenhagen/urban background, Aarhus/street and the rural site Risø.
- Additionally, PM_{10} and $PM_{2.5}$ were measured by TEOM (Tapered-Element Oscillating Microbalance) on a half hourly basis at selected stations: HCAB (PM_{10} and $PM_{2.5}$), Risø (PM_{10}) and Århus street station (PM_{10}). The high time resolution is making it possible to resolve the diurnal variation. Part of these measurements was carried out in a research project funded separately by the Danish EPA.
- Particle number was measured at Copenhagen/street (HCAB), Copenhagen/urban background and Risø in cooperation with a particle research project funded separately by the Danish EPA. Additionally, measurements were started at a suburban site in Hvidovre in autumn 2015.
- Ozone (O_3) was measured at all urban background and rural stations (except Anholt), and at the street stations Copenhagen/street (HCAB).
- Carbon monoxide (CO) was measured at all street stations except Jagtvej as well as at the urban background station, Copenhagen/urban background and the rural site Risø.
- Benzene and toluene were measured at Copenhagen/street (HCAB) and Copenhagen/urban background using passive sampling on a weekly basis.

- PAHs were measured at Copenhagen/street (HCAB) and at the suburban site in Hvidovre.
- Sulphur dioxide (SO_2) was measured at Copenhagen/street (HCAB). The main purpose was to monitor episodic high concentrations.
- Elemental carbon (EC) and organic carbon (OC) in $PM_{2.5}$ were measured at Copenhagen/street (HCAB), at the rural station Risø and at the suburban station Hvidovre. EC in $PM_{2.5}$ was measured at the urban background station (HCØ). In addition, the main inorganic ions in $PM_{2.5}$ was determined at 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 2.

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 as 24-hour averages using LVS (gravimetric method) but also to a lesser extend as hourly averages using TEOM. Elements in the particles as well as PAH were measured as 24-hour averages. EC and OC were measured as 24-hour averages. Benzene and toluene were measured weekly by passive sampling. Furthermore, volatile organic compounds were sampled as 24-hour averages.

2.2 Air quality model calculations

In the monitoring programme, the measurements at the fixed site measuring stations are supplemented with model calculations of NO_2, $PM_{2.5}$, and PM_{10} at street level and O_3 at the regional level. These model calculations are carried out with an integrated multiscale model system (the THOR modelling 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 THOR system includes the Danish Eulerian Hemispheric Model, DEHM (Christensen, 1997; Brandt et al., 2012), the Urban Background Model, UBM (Brandt et al., 2001; Brandt et al., 2003) and the Operational Street Pollution Model, OSPM® (Berkowicz 2000a; Ketzel et al., 2012).

The modeling system has been updated on several aspects regarding the reporting for 2018. The main updates concern the following aspects:

- *Meteorological input data.* A new meteorological model has been used to provide the meteorological data that are used to drive the model calculations. The new model WRF (Weather Research and Forecasting Model) has been developed by National Center for Atmospheric Research, USA (NCAR). This model replaces the old model MM5 (also from NCAR) that no longer is subject to development and support.
- *Emission inventories*. The emission inventories for Europe are based on emissions provided by EMEP. EMEP has improved the geographical resolution of the inventories from 50 km x 50 km to 0.1° x 0.1° (about 11 km in N-S direction). In addition EMEP has adopted a new system for categorizing the emission sources.

- *Description of chemical reactions*. The description of the chemical scheme in the models has been further developed in order to improve the modelling of particles. The new scheme for particles includes black carbon (BC), primary organic matter (OM) and secondary organic aerosols (SOA).

These major changes and a number of minor changes are described further in Ellermann et al. (2019).

The following paragraphs provide a short description of the models that are used for carrying out the model calculations presented in this report.

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 at http://www.au.dk/thor). The same model setup is also used for an air pollution map that shows modelled urban background and street concentrations at all 2.4 million addresses in Denmark presented at a publicly available website (luftenpaadinvej.au.dk; Jensen et al., 2017).

Model calculations of air quality on national scale are carried out using DEHM (version Feb 2019), which is an Eulerian model where emissions, atmospheric transport, chemical reactions, and dry and wet depositions of air pollutants are calculated in a 3D grid covering the northern hemisphere with a resolution of 150 km x 150 km. The model includes a two-way nesting capability, which makes it possible to obtain higher resolution over limited areas. Three nested domains are used in the model runs under NOVANA, where the first domain is covering Europe with a resolution of 50 km x 50 km. The second domain is covering Northern Europe with a resolution of 16.7 km x 16.7 km. The calculations of air quality in Denmark are carried out in a third domain with a horizontal resolution of 5.6 km x 5.6 km. In the vertical direction, the model is divided into 29 layers covering the lowest 15 km of the atmosphere. Of these, the lowest layers are relatively thin (20 m) while the upper layers are relatively thick (2,000 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 aggregated 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 2017 compiled by DCE (http://envs.au.dk/en/knowledge/air/emissions/) and international emission inventories for the year 2016 collected and distributed by EMEP (www.emep.int). Ship emissions around Denmark with very high resolution of 1 km x 1 km (Olesen et al., 2009) have been used after adjustments according to the regulation by 1 January 2015 that decreased the allowed content of sulphur in fuel used by ships in the Sulphur Emission Control Area (SECA: the North Sea and the Baltic Sea) from 1% to 0.1%. The new version of DEHM is since October 2019 also part of the Copernicus Atmospheric Monitoring Service providing quality-controlled operational air quality forecasts for Europe (https://atmosphere.copernicus.eu/).

The Urban Background Model, UBM (version 10.0), calculates the urban background air pollution with high spatial resolution (1 km x 1 km). For the calculations Danish emission inventories have been compiled for the same grid of 1 km x 1 km as the calculations are performed. In addition, the models uses other input data including meteorological data from the WRF model, and

air pollution initial concentrations and boundary conditions (concentrations at the edge of the calculation domain for UBM) obtained from DEHM . The UBM includes a Gaussian plume approximation for calculation of the dispersion and transport of the air pollutants to every receptor point at the local scale up to 25 km from each receptor point, and a simple chemical model accounting for the photochemical reactions of NO_x and O_3 (originally developed for OSPM). The basic principles of the model are described in Berkowicz (2000b). In recent years, UBM has undergone many improvements in the formulation of physical processes, and it now treats both area and point sources in a more physically consistent manner compared to previous versions of the model. These updates of UBM has improved the overall performance of the model; documented in comparisons with measurements. Furthermore, analyses of the results have shown that the model now provides a more realistic spatial distribution of concentrations around large point sources. The emissions used in the UBM model are obtained from the SPREAD model that spatially distributes national emissions from 2017 from all sectors on the 1 km x 1 km calculation grid for Denmark (more details on the SPREAD model can be found in Plejdrup et al., 2018). No calibration for NO_x /NO_2 for UBM has been carried out. For $PM_{2.5}$ and PM_{10} a small correction towards higher concentrations has been applied for the modelled values to compensate for a slight underestimation, for details see Appendix 3.

Finally, the street canyon model OSPM® (www.au.dk/ospm) is used to calculate the air quality at 2 m height at the sidewalks in selected streets. Data from the meteorological model WRF and air pollution concentrations from UBM are used as input to the model. The model calculates and apply emissions from traffic in the specific street. The OSPM includes a simple chemical model accounting for the photochemical reactions of NO_x and O_3 (it is this chemical modulet hat is also applied in UBM), and the model calculates the dispersion of air pollution in the urban street (accounting for the influence of meteorological conditions, turbulence induced by traffic and the specific street geometry).

The traffic data and street configuration data used as input for OSPM for the selected urban streets are generated using the AirGIS system. AirGIS is constructed around a GIS road network with traffic data, GIS foot-prints of buildings with building heights and a series of small calculation routines (Jensen et al., 2001; 2009; 2017; Khan et al., 2019; http://envs.au.dk/videnudveksling/luft/model/airgis/). We also refer to this model chain as DEHM/UBM/AirGIS.

Traffic data used in the OSPM calculations 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 is determined for the location of the calculation points. For Copenhagen, traffic data is constructed from manual counts performed annually or with 5-year intervals. Aalborg does not have a systematic traffic counting program like Copenhagen, and traffic data is therefore based on available data from manual and automatic traffic counts in combination with data from a traffic model. Based on information from Copenhagen and Aalborg municipalities, the Average Daily Traffic (ADT) and vehicle distribution for all streets in the calculations have been updated on basis of the most recent available traffic data. The vehicle distribution includes data for passenger cars, vans, trucks, and buses. In Copenhagen, 37 out of the 98

calculation points had updated traffic data for 2018. For Aalborg 15 out of 31 streets had updated traffic data.

Manual traffic counts are carried out annually for the street segments with the location of the measuring stations of H. C. Andersens Boulevard and Jagtvej in Copenhagen. Manual counts for the 2018 assessment originate from September 2018 in Copenhagen. In Aarhus, automatic traffic recording was carried out to estimate traffic volume and vehicle classification during three separate weeks in March, May, and November 2018. This method provides good estimates of traffic volume but only rough estimates of vehicle classification. One of the shortcomings is that the method cannot differentiate between passenger cars and vans as they have the same distance between axles. Hence, a manual count from 2015 was used for vehicle distribution.

Automatic traffic recording at Odense (Grønløkkevej) was carried out during three weeks in April 2018. Traffic volume and vehicle distribution were established based on this information assuming the same share of vans as the average of 98 streets in Copenhagen. In Aalborg (Vesterbro), the measuring station was not in operation during 2018 due to nearby building construction work, and the station has ultimo 2019 been moved to another location at the same street.

All the applied air pollution models are driven by meteorological data from the meteorological model WRF (Weather Research and Forecasting Model) that has been developed by National Center for Atmospheric Research, USA (NCAR). Details about WRF can be found in Skamarock et al. (2008).

The calculations were carried out in order to determine annual means of NO_2, $PM_{2.5}$ and PM_{10} concentration in 98 streets in Copenhagen and 31 streets in Aalborg. In previous years, calculations were only performed for NO_2, but since 2017 $PM_{2.5}$ and PM_{10} have been included.

2.2.1 Model calibration and validation

In the assessment for 2013, the model calculations with OSPM were improved through major revisions of the model. These included 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 3 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 2018 is similar to that of 2013 and onwards.

Before 2015, OSPM was calibrated against measurements at the street stations for the calculation year in question in order to ensure good correspondence between measured and modelled NO_2. Since the assessment of 2016, we are using available data from the last three years to avoid potential fluctuations that a single year approach may introduce. For some years, the street station at H. C. Andersen's Boulevard was not used in the calibration due to the about 8 $\mu g/m^3$ jump in concentrations since a change in street layout moved traffic closer to the station in 2010. The station was moved during October 2016 to compensate for the change in street layout, and hence the station has been in operation on this new location and included in the calibration from this time and onwards.

The comparison between modelled and observed NO_2 concentrations for 2018 are shown in table 2.2. For further details on the calibration and validation of the model system for the period 2016-2019 is presented in Appendix 3 that documents the good performance of the modelling.

The correlation between modelled and observed NO_2 concentrations for 2018 shows a good agreement for the street stations in Copenhagen but the model overestimates concentrations at the street stations in Odense and Aalborg. The DEHM/UBM models also overestimate urban and regional background concentrations.

Table 2.2. Comparison of modelled and measured annual means of NO_2 concentrations in 2018.

Unit: µg/m³	Measurements	Model results	Difference	Models used
Street:				
Copenhagen/HCAB/1103	39	39	-0.7%	DEHM/UBM/OSPM
Copenhagen/Jagtvej/1257	30	29	-5.4%	DEHM/UBM/OSPM
Aarhus/6153	25	33	24%	DEHM/UBM/OSPM
Odense/9156	17	27	62%	DEHM/UBM/OSPM
Urban Background:				
Copenhagen/1259	13	15	24%	DEHM/UBM
Aarhus/6160	12	18	46%	DEHM/UBM
Odense/9159	11	14	34%	DEHM/UBM
Aalborg/8159	11	10	-13%	DEHM/UBM
Hvidovre/2650	12	13	8%	DEHM/UBM
Rural:				
Risø/2090	8	12	66%	DEHM/UBM
Keldsnor/055	9	12	55%	DEHM/UBM
Anholt/6001	5	8	74%	DEHM/UBM

The comparison between modelled and observed $PM_{2.5}$ and PM_{10} concentrations for 2018 is shown in table 2.3 and table 2.4, respectively. The modelled particle concentrations are slightly underestimated in comparison with observed concentrations.

Table 2.3. Comparison of modelled and measured annual means of $PM_{2.5}$ concentrations in 2018.

Unit: µg/m³	Measurements	Model results	Difference	Models used
Street:				
Copenhagen/HCAB/1103	16	15	-11%	DEHM/UBM/OSPM
Copenhagen/Jagtvej/1257	14	13	-11%	DEHM/UBM/OSPM
Aarhus/6153	14	12	-15%	DEHM/UBM/OSPM
Urban Background:				
Copenhagen/1259	13	11	-16%	DEHM/UBM
Aarhus/6160	12	11	-7%	DEHM/UBM
Aalborg/8159	12	9	-22%	DEHM/UBM
Hvidovre/2650	12	11	-8%	DEHM/UBM
Rural:				
Risø/2090	12	11	-12%	DEHM/UBM

Table 2.4. Comparison of modelled and measured annual means of PM_{10} concentrations in 2018.

Unit: µg/m³	Measurements	Model results	Difference	Models used
Street:				
Copenhagen/HCAB/1103	31	25	-23%	DEHM/UBM/OSPM
Copenhagen/Jagtvej/1257	25	21	-18%	DEHM/UBM/OSPM
Aarhus/6153	22	21	-10%	DEHM/UBM/OSPM
Odense/9156	23	21	-12%	DEHM/UBM/OSPM
Urban Background:				
Copenhagen/1259	18	16	-15%	DEHM/UBM
Rural:				
Risø/2090	17	16	-10%	DEHM/UBM

2.3 Health impacts and external costs of air pollution

Model calculations of the health impacts and associated external costs related to air pollution have been included in the air quality monitoring programme since the revision of NOVANA in 2016. High-resolution assessment of health impacts from air pollution and related external costs have therefore been carried out for Denmark for the years 2016-2018 using the integrated EVA (Economic Valuation of Air Pollution) model system, version 5.2 (Brandt et al., 2015; 2016). A three-year average is applied in the assessment to smooth out variations in meteorological conditions between years. EVA is based on the impact-pathway methodology, where the site-specific emissions will result, via atmospheric transport and chemistry, in a concentration distribution, which together with detailed population data, is used to estimate the population-level exposure. Using exposure-response functions and economic valuations, the exposure is transformed into impacts on human health and related external costs (see figure 2.4).

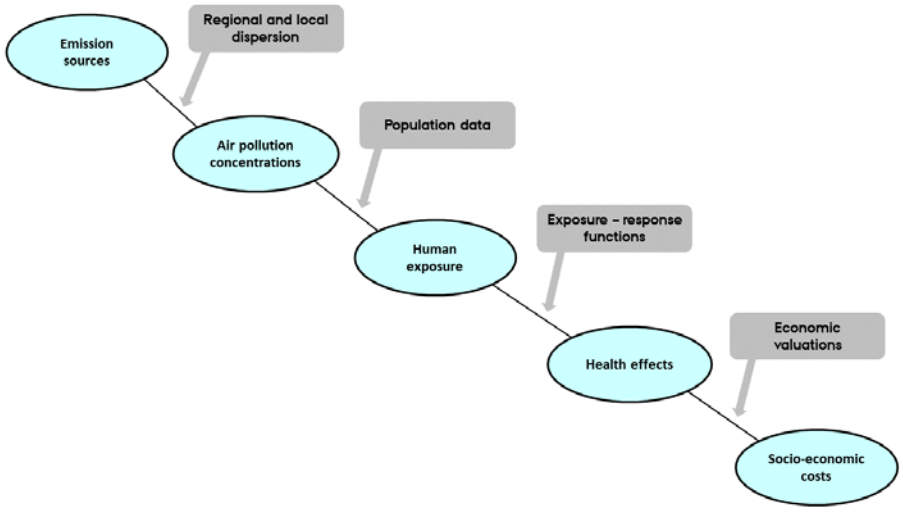


Figure 2.4. An illustration of the EVA model system, based on the impact pathway chain.

As described in Chapter 2.2 the air quality model calculations have been updated and this consequently introduces adjustments in the results for the calculated health impacts and external costs. In addition, important updates have been implemented in the EVA-system concerning the following aspects:

- *Exposure-response relationships*. The calculations include for the first time a direct impact of NO_2 on mortality and morbidity (Andersen et al., 2019). This increases the mortality and morbidity, and thereby external costs attributed to air pollution.
- *Economic valuation of a statistical life*. The new calculations are based on the updated value of a statistical life from the Danish Economic Council (2016) and Ministry of Finance (2017). This value is about twice as high as the value used in the previous model calculations.

These major changes, and a number of minor revisions, are described further in Ellermann et al. (2019) that also provides more details about the exposure-response functions used for the calculations. Further details are given in Andersen et al. (2019).

The air quality data used in the EVA system is based on calculations with the two chemistry transport models (DEHM and UBM) described above. Exposure to $PM_{2.5}$ is responsible for the majority of the health impacts from air pollution in Denmark. Table 2.3 shows a comparison between measured and calculated annual concentrations of NO_2 and $PM_{2.5}$ at the Danish measurements stations. The model results for $PM_{2.5}$ have been calibrated (addition of 2.3 $\mu g/m^3$) for this year's reporting to adjust for a bias between measurements and model calculations. No calibration was used for NO_2 and the other compounds used for the calculation of health impacts.

The population density for Denmark is based on the geographical distribution in the Civil Registration System (CPR data) from 2017. The individual health impacts in the EVA system have previously been documented in Brandt et al. (2013a;b) and reviewed in Bønløkke et al. (2011). The exposure-response relationships and economic valuation of the individual health impacts are described in Andersen et al. (2019), and the methodology for the economic valuation is documented in Andersen et al. (2004) and Bach et al. (2006). The updated exposure-response functions have been selected based on an international reviewed project funded by WHO (WHO, 2013). The EVA model system has previously been applied for assessment of future scenarios (Geels et al., 2015), and EVA has furthermore been compared with other health impact assessment systems (Anenberg et al., 2015).

3. Nitrogen oxides

The nitrogen oxides (NO, NO_2, NO_x) are measured at thirteen monitoring sites using gas monitors based on chemiluminescence. The concentrations are measured continuously throughout the year with a time resolution of minutes that is aggregated to hourly averages for this report.

3.1 Annual statistics

The annual statistics for 2018 for nitrogen dioxide (NO_2) and nitrogen oxides are shown in tables 3.1 and 3.2. There were no exceedances of the annual limit value for NO_2 of 40 µg/m^3 (EC, 2008). Further, there were no exceedances of the hourly limit value for NO_2 of 200 µg/m^3. This value must not be exceeded more than 18 times within a calendar year (see 19th highest hourly concentration in table 3.1). In 2018, there was no information to the public triggered by exceedance of the information threshold for NO_2 (three hours average must not exceed 400 µg/m^3). Installation problems regrettably resulted in the EU requirement of 7446 hours of hourly averaged values were not being upheld at the street station in Copenhagen, Copenhagen/1257 and the Rural station Keldsnor/9055, column two in tables 3.1 and 3.2.

Table 3.1. Nitrogen dioxide (NO_2) in 2018. All parameters are based on hourly averages.

Unit: µg/m^3	Number	Average	Median	98-percentile	19-highest
Street:					
Copenhagen/1257	6747	30	25	85	110
Copenhagen/1103	8234	39	36	93	117
Aarhus/6153	8275	25	22	66	88
Odense/9156	8206	17	14	49	73
Aalborg/8151 §	0	-	-	-	-
Urban Background:					
Copenhagen/1259	8040	13	10	43	63
Aarhus/6160	8195	12	9	42	63
Odense/9159	7871	11	8	35	54
Aalborg/8158	8146	11	9	39	62
Suburban:					
Hvidovre/2650	8125	12	9	45	69
Rural:					
Risø/2090	8129	8	5	28	49
Keldsnor/9055	7228	9	7	30	49
Anholt/6001	8124	5	3	20	30
Ulborg/7060	8001	4	3	16	27
Limit value 2010	>7446*	40			200

*) 90% data capture of number of hourly measurements in relation to total number of hourly measurements in 2018 excluding hours used for calibration.

§) For Aalborg/8151 (street) there is no data since the station was shut down due to construction work at the site. Measurements were not reinitiated in Aalborg (traffic) before ultimo 2019.

Table 3.2. Nitrogen oxides (NO_x=NO+NO_2) in 2018. All parameters are based on hourly averages.

Unit: µg/m³ (as NO_2)	Number	Average	Median	98-percentile	19-highest
Street:					
Copenhagen/1257	6747	56	40	209	358
Copenhagen/1103	8234	84	68	264	404
Aarhus/6153	8275	49	39	160	294
Odense/9156	8206	30	22	109	257
Aalborg/8151 §	0	-	-	-	-
Urban Background:					
Copenhagen/1259	8040	15	12	57	117
Aarhus/6160	8195	15	10	63	144
Odense/9159	7871	13	9	52	119
Aalborg/8158	8146	15	10	58	128
Suburban:					
Hvidovre/2650	8125	16	10	69	213
Rural:					
Risø/2090	8129	8	6	33	69
Keldsnor/9055	7228	9	6	38	67
Anholt/6001	8124	6	4	23	38
Ulborg/7060	8001	5	3	17	28

§) Aalborg/8151 (street) there is no data since the station has been shut down due to construction work at the site. Measurements were not reinitiated in Aalborg (traffic) before ultimo 2019.

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 concentrations of nitrogen oxides are due to national and international regulations of the emissions. The large emission reductions in the cities are achieved by improvement of the vehicles, for example mandatory use of catalytic converters.

For many years, the long-term trend in nitrogen dioxide showed a decrease much smaller than observed for NO_x. However, since around 2006, NO_2 has decreased at about the same rate as NO_x. The smaller decrease before 2006 was mainly due to an increase in the share of diesel cars and increase in the share of diesel cars with oxidative catalysts where up to about half of the emissions of NO_x consist of NO_2 (called direct NO_2). This increase in the direct emissions of NO_2 counteracted the decrease in the traffic emissions from vehicles. The amount of directly emitted NO_2 reached a maximum in 2009-2011 and has slightly decreased since then. This change in the amount of directly emitted NO_2 is believed to be one of the main reasons why NO_2 now decreases at a similar pace as NO_x.

At Odense street station and Aarhus urban background station there have been large decreases in NO_x and NO_2 since 2013. In Odense, there was a major permanent rearrangement of the traffic in Odense Centre that changed the traffic at the street station in Albanigade in two steps from a street with relatively high traffic intensity to a street with much reduced traffic intensity. Finally, the street was closed for traffic in 2015. These changes began on 28 June 2014. This is the reason for the large decrease of the NO_2 and NO_x values for Odense/9155 in 2014 and 2015. The station was shut down on 16 June 2015 and was relocated to Grønnelykkevej and was renamed Odense/9156 in June 2016. The large change at Aarhus/background from 2013 to 2014 is due to the relocation of the measurement site in January 2014 (Chapter 2.1) to an urban background area with lower concentrations compared to the old location.

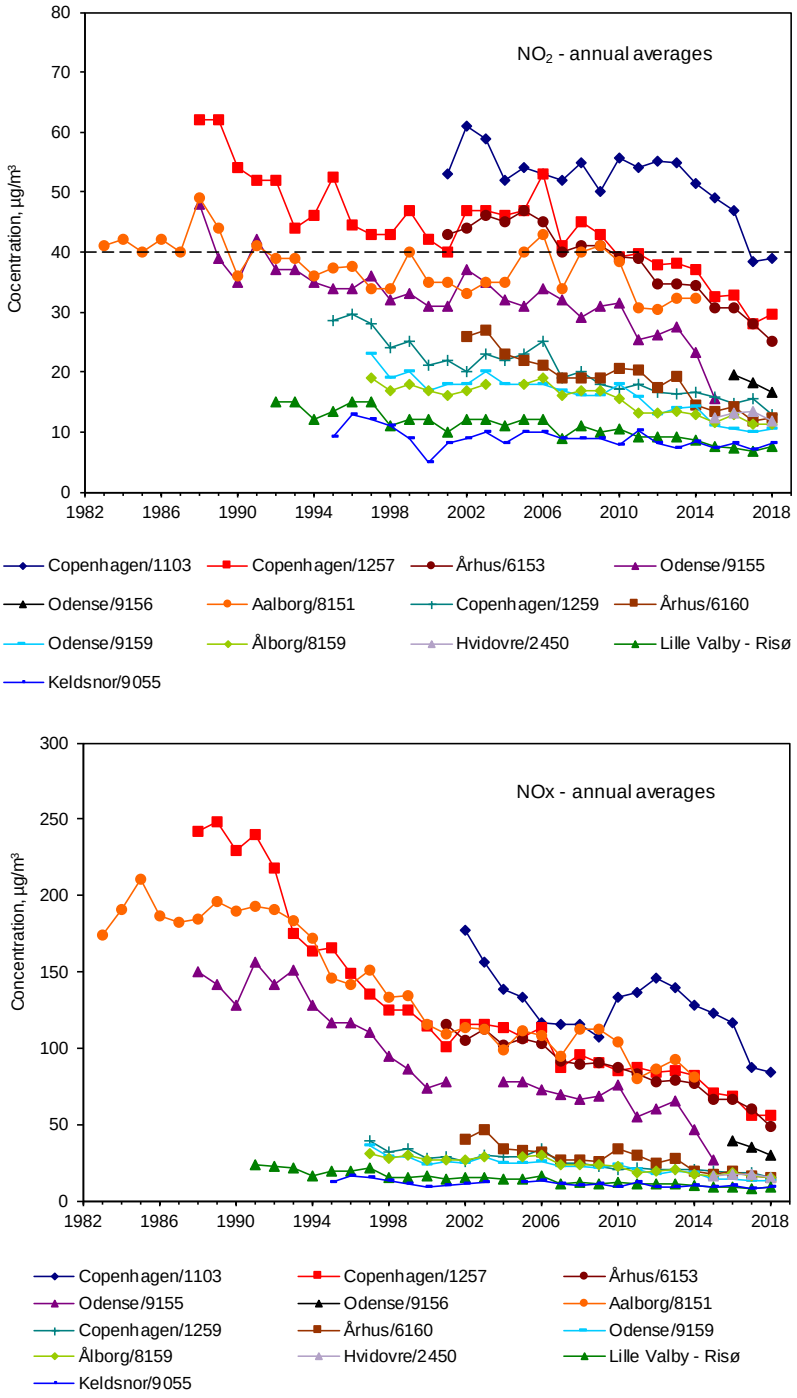


Figure 3.1. The graphs show the time series for the annual average values of NO_2 and NO_x. The dashed line on the upper graph shows the limit value that entered into force in 2010. On the same curve, results are shown from both the previous (6159) and the new background station (6160) in Aarhus.

During October 2016 the measurement station at H.C. Andersens Boulevard was moved 2.7 m (corresponds approximately to the width of a traffic lane) further away from the inner traffic lane. The aim of this relocation was to return to the same distance from the traffic lane as it was prior to 2010 (see Chapter 2.1 for further details). In 2010, the driving lanes were changed at the section of H. C. Andersens Boulevard where the measurement station (Copenhagen/1103) is located. This change moved the traffic closer to the measurement station and resulted in an increase in the annual average concentrations of NO_2 of about 8 µg/m³ in comparison to the levels measured before

the introduction of the new driving lanes. The data from 2017 and onwards shows the full impact of the relocation of the station on the annual average. The 8 μg/m^3 change in concentration of NO_2 from 2016 to 2017 is therefore partly due to the relocation of the measurement station and partly due to the general reduction of the emissions from traffic as seen on the other street stations (e.g. Jagtvej).

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). The selected streets represent busy streets and are mainly so-called street canyons. Concentrations are elevated in street canyons due to the restricted dispersion conditions. 98 streets are included for Copenhagen and 31 in Aalborg. ADT (Average Daily Traffic) was between 5,100 and 79,400 vehicles/day in Copenhagen and between 2,700 and 29,000 vehicles/day in Aalborg.

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

3.3.1 NO_2 model calculations for Copenhagen

The annual mean concentrations of NO_2 for streets in Copenhagen in 2018 are shown in figures 3.2 (bar chart) and 3.3 (map). The average of the NO_2 street concentrations at all 98 streets increased from 2017 to 2018 (1.7 μg/m^3), the average urban background concentrations also increased (2.3 μg/m^3) and similarly the regional background contribution increased (1.8 μg/m^3). The regional background concentrations are included in the urban background concentrations and the background concentrations are included in the street concentrations. Measurements at H. C. Andersens Boulevard and Jagtvej in Copenhagen also show a small increase in NO_2 concentrations from 2017 to 2018. The increase in street concentrations is a result of a combination of changes in traffic, emission factors, background concentrations and meteorology. There has been a very slight decrease in ADT (-0.4%) but the share of heavy-duty vehicles remained the same as in 2017 and travel speeds are assumed similar as in 2017. However, there have been some changes in ADT and heavy-duty share for a few of the streets included in the model calculations. Vehicle emission factors show a decrease due to the general replacement of the car fleet where the increase in Euro 6 vehicles with low emissions and replacement of older vehicles with higher emissions play a significant role. Furthermore, the fraction of directly emitted NO_2 has also slightly decreased, leading to slightly lower NO_2 concentrations. For 2018, the NO_2 fraction is 11% based on analysis of measurements of NO_x, NO_2 and O_3, whereas it was 12% in 2017. The combination of the similar traffic conditions and lower vehicle emissions factors would, other things equal, lead to lower concentrations. However, the increase in urban and regional background concentrations is not due to increase in emissions in Denmark or Europe, as emissions show a downward trend. Therefore, the increase in modelled concentrations is partly due to changes in meteorological factors from 2017 to 2018, as the observed levels also increase. However, part of the explanation is

also that the model overestimates the urban background concentrations as shown in section 2.2.1.

In 2018, the limit value for the annual mean concentration was only exceeded at one of the 98 selected streets in Copenhagen according to the model results (figure 3.2). No exceedances were calculated for 2017. However, the number of streets exceeding the limit value is sensitive to very small changes in concentrations, since a number of streets still are close to the limit value (figure 3.2).

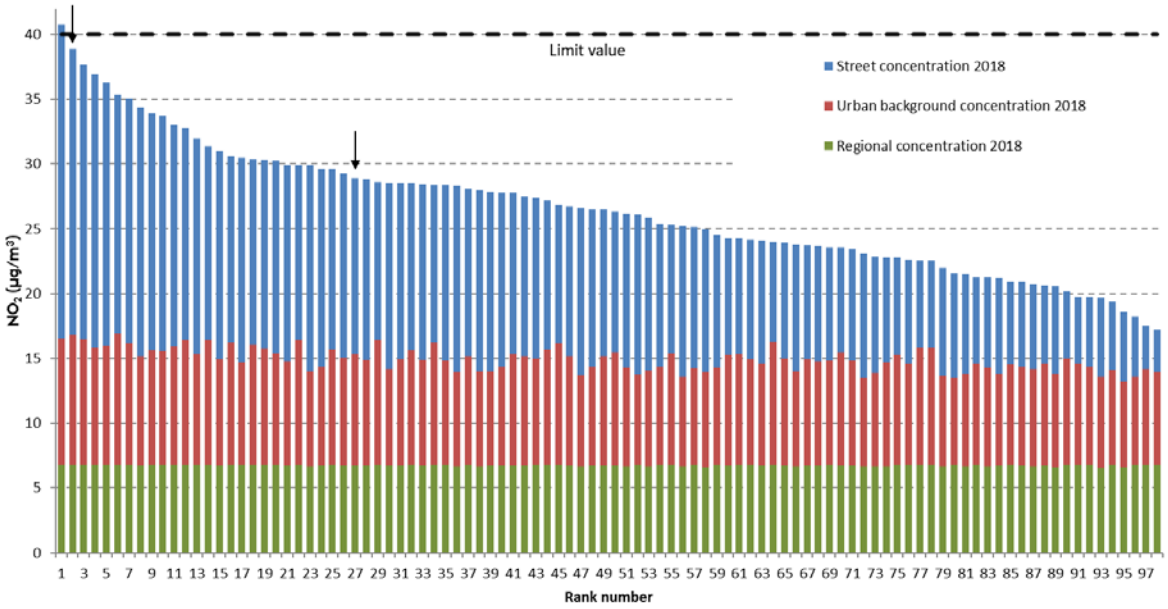


Figure 3.2. Annual mean concentrations of NO_2 in 2018 for 98 streets in Copenhagen according to model calculations. The contribution from traffic in the street canyons is based on the street canyon model OSPM® (blue colour). The urban background (reddish colour) is obtained from calculations with the urban background model UBM with input from the regional scale model DEHM (green colour). The value for a street segment is for the side of the street with the highest annual mean concentration of the two sides. However, for streets with a measuring station it is the side where the station is located. The names of the streets can be seen in table 3.3. Arrows indicate street segments with a measuring station.

The 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.

There have been minor changes in the ranking of streets according to NO_2 concentrations from 2017 to 2018, mainly due to small changes in traffic inputs. The highest modelled NO_2 concentration in 2018 is at H. C. Andersen's Boulevard (2) (40.8 µg/m^3). The second highest (38.9 µg/m^3) is where the measuring station is located (H. C. Andersens Boulevard (1)).

Table 3.3. Rank number and names for the street segments that are shown in figures 3.2 and 3.3. The streets are numbered (1-98) according to NO_2 levels in 2018 (1 = highest, 98 = lowest). The numbers in parentheses refer to different segments of the same street that has more than one model calculation. An asterisk (*) indicates a street segment with a measurement station.

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

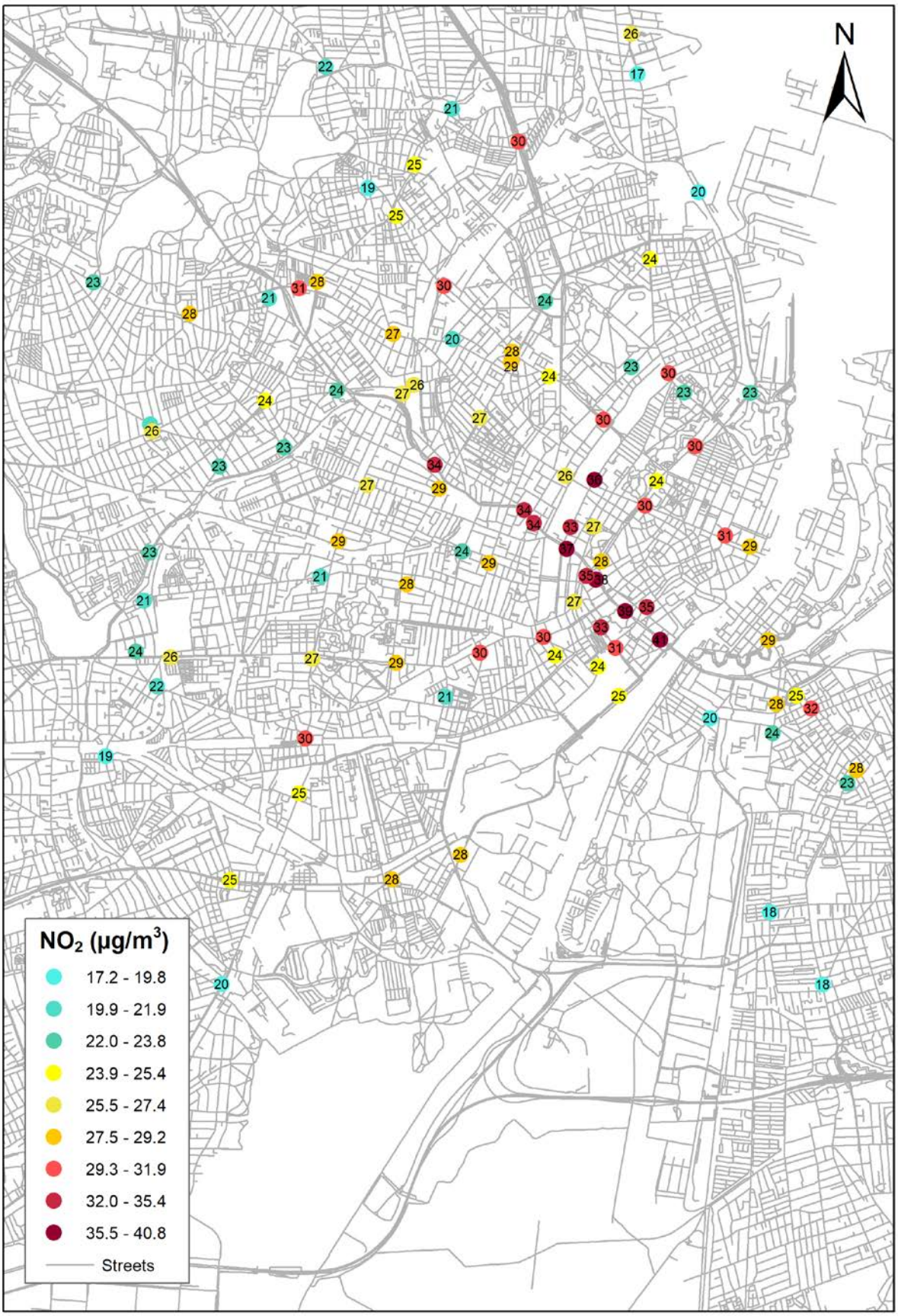


Figure 3.3. Map showing the locations of the selected streets in Copenhagen and the annual mean concentrations of NO_2 for 2018 visualized on top of the calculation point. The contribution from traffic in the street canyons is based on the street canyon model OSPM®. The urban background is obtained from calculations with the urban background model UBM with input from the regional scale model DEHM. The value for a street segment is for the side of the street with the highest annual mean concentration of the two sides. However, for streets with a measurement station it is the side where the station is located. The names and numbers for the streets are shown in table 3.3. The map can be viewed at a webGIS service, see https://arcg.is/qLSaD.

3.3.2 NO_2 model calculations for Aalborg

For Aalborg the modelled street concentrations show an average small increase of about 0.4 µg/m³ for NO_2 compared to 2017 when considering all 31 street segments. Measurements at H. C. Andersen's Boulevard and Jagtvej in Copenhagen also show a small increase in NO_2 concentrations from 2017 to 2018, whereas measurements from street station in Aarhus show a small decrease. Street measurements are not available from Aalborg. The same level is modelled for urban background concentrations, which is also consistent with measurements at the urban background station in Aalborg. The small increase in street concentrations is the result of a combination of several factors. On average ADT and heavy-duty share of vehicles were unchanged, and travel speeds were assumed to be unchanged. Vehicle emission factors show a decrease due to the general replacement of the car fleet where the increase in Euro 6 vehicles with low emissions and replacement of older vehicles with higher emissions play a significant role. Furthermore, the directly emitted NO_2 of NO_x emissions (NO_2 fraction) has also slightly decreased, leading to slightly lower modelled NO_2 concentrations. The combination of the same traffic conditions and lower vehicle emissions would, other things equal, lead to lower concentrations. However, street concentrations have shown a slight increase that most likely is due to changes in meteorological factors from 2017 to 2018.

According to the model calculations, the limit value for the annual mean concentration in 2018 was not exceeded at any of the 31 selected streets (figure 3.4 and figure 3.5). The order of some of the streets has changed slightly due to changes in traffic data.

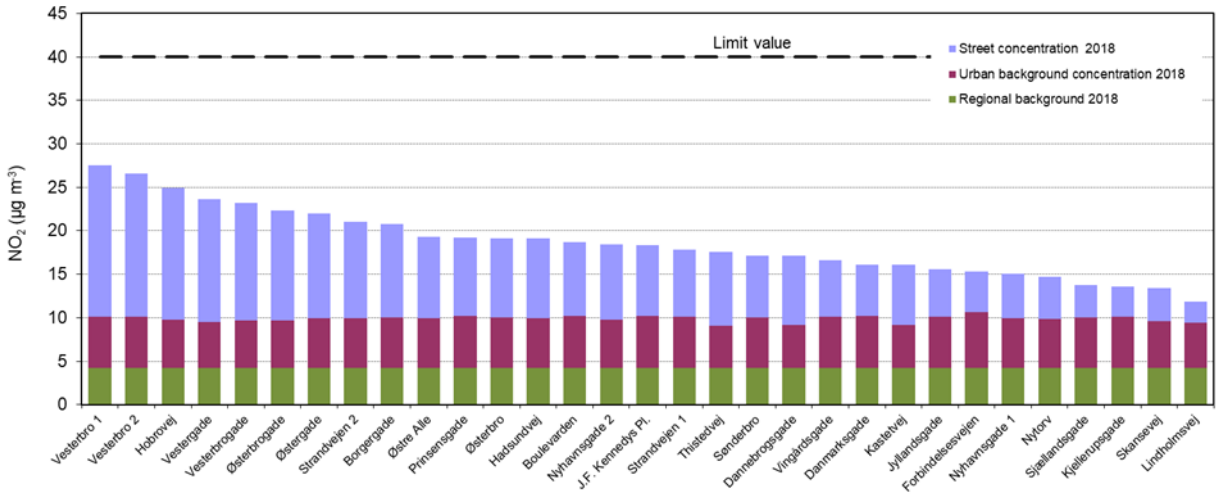


Figure 3.4. Modelled annual mean concentrations of NO_2 in 2018 for 31 streets in Aalborg. The contribution from traffic in the street canyons is based on the street canyon model OSPM® (blue colour). The urban background (dark red colour) is obtained from calculations with the urban background model UBM (reddish colour) with input from the regional scale model DEHM (green colour). The value for a street segment is for the side of the street with the highest annual mean concentration of the two sides. However, for streets with a measurement station it is the side where the station is located. Vesterbro 1 is the street segment where the measurement station is located. However, the station was not been operational during 2018 due to nearby building construction works.

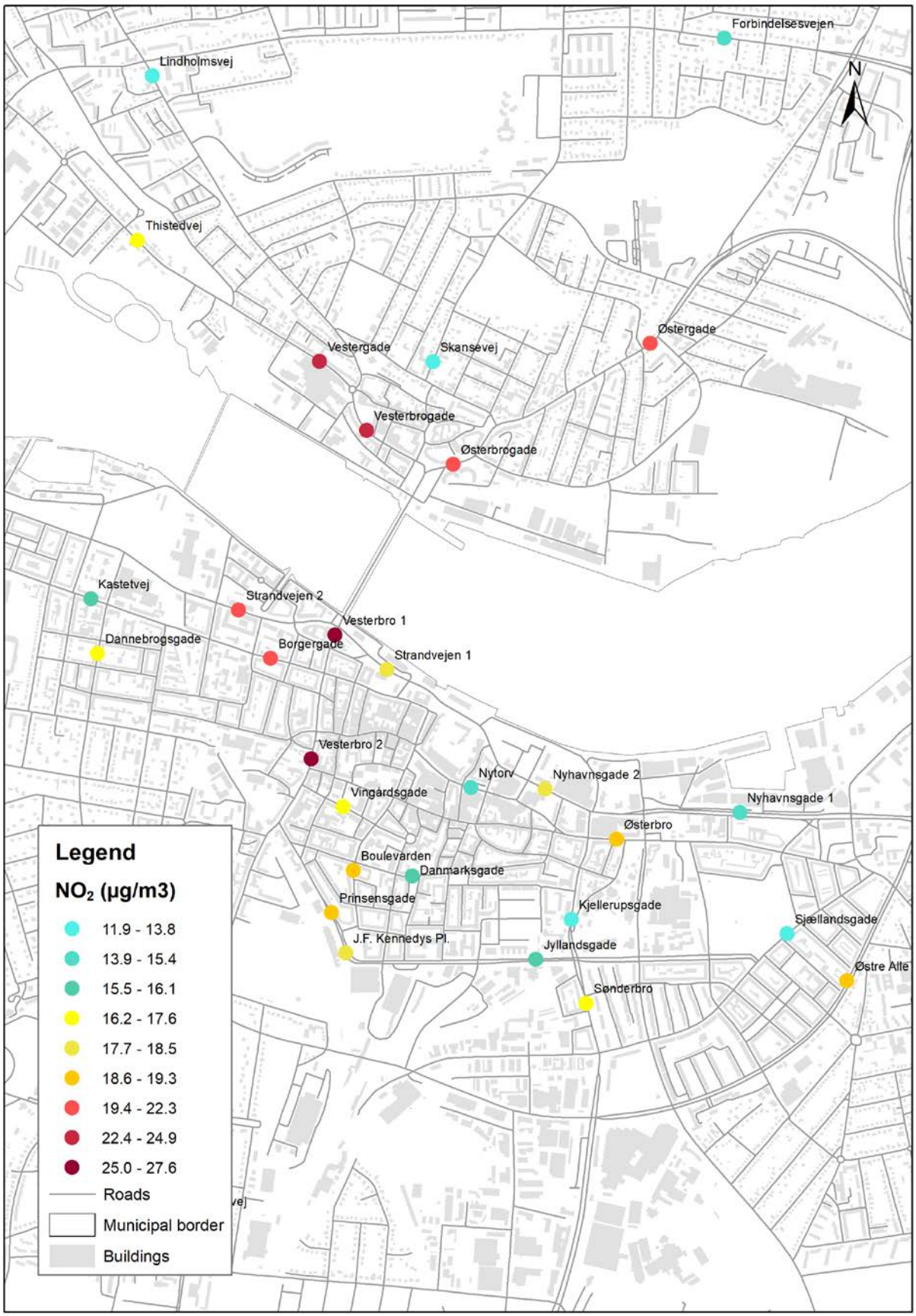


Figure 3.5. Map showing the location of the selected streets in Aalborg and the annual mean concentrations of NO_2 for 2018. The contribution from traffic in the street canyons is based on the street canyon model OSPM®. The urban background is obtained from calculations with the urban background model UBM with input from the regional scale model DEHM. The value for a street segment is for the side of the street with the highest annual mean concentration of the two sides. However, for streets with a measurement station it is the side where the station is located. Vesterbro 1 is the street segment with the measurement station, however, not operating in 2018 due to nearby building construction work. Map can be viewed at a webGIS service, see http://arcg.is/1Lf8CP

3.3.3 Trends in modelled exceedances of NO_2

In figure 3.6, the modelled trends in number of exceedances of annual mean of NO_2 are shown for Copenhagen and Aalborg. The limit value of 40 µg/m³ for annual mean of NO_2 had to be met in 2010, and in previous years, the limit value plus a margin of tolerance depending on the year in question had to be met.

For Copenhagen, the number of exceedances has decreased from 58 in 2008 to 6 in 2016, and it decreased further to zero in 2017 whereas it showed one exceedance in 2018. The main reason for the increase in number of exceedances between 2007 and 2008 in Copenhagen from 32 to 58 is that the limit value plus margin of tolerance for the annual mean concentration of NO_2 decreased from 46 µg/m³ in 2007 to 44 µg/m³ in 2008 (EC, 2008). This decrease in margin of tolerance lead to a higher number of streets exceeding the limit value plus margin of tolerance in 2008 compared to 2007. If the limit value plus margin of tolerance had been 44 µg/m³ in 2007, the number of streets exceeding the limit value plus margin of tolerance would have been 53. Roughly the same level as in 2008. In Copenhagen, the analysis includes 138 streets during 2007 to 2010 and 98-99 the following years. The reduction in the number of included streets from 2011 and onwards was implemented to better match locations of selected streets with locations with manual traffic counts.

For Aalborg, the model results showed 3-4 exceedances in 2007-2009, and none since 2010. Here the analysis includes 32 streets from 2007 to 2010, and 31 streets from 2011 and onwards.

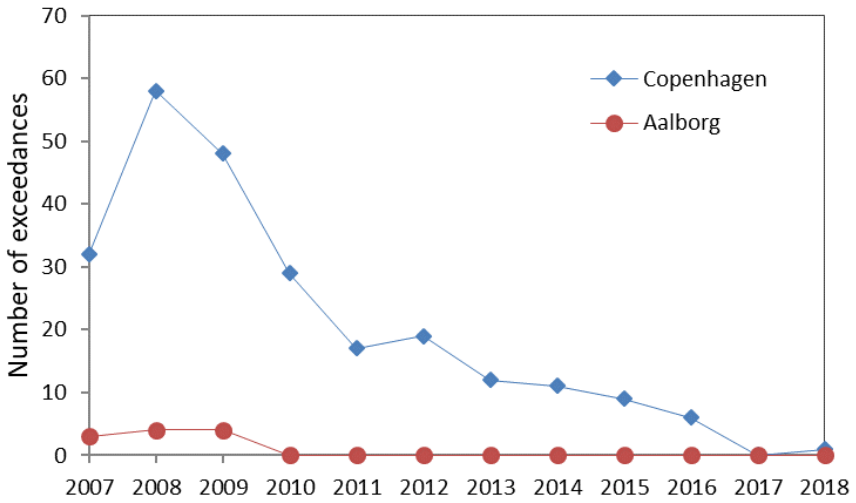


Figure 3.6. Trends in modelled exceedances of annual mean of NO_2 in Copenhagen and Aalborg.

4. Ozone

O_3 is measured at eight monitoring sites using gas monitors based on ultraviolet photometry. The concentrations are measured continuously throughout the year with a time resolution of minutes that is aggregated to hourly averages for the present report.

4.1 Annual statistics

The annual statistics for 2018 for O_3 are shown in table 4.1. The maximum 8-hour 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 were not exceeded for 2016-2018 at any of the stations. The long-term objective (maximum 8-hour daily mean value must not exceed 120 µg/m^3; table 4.1 column 5) was exceeded at all stations but the traffic station. However, the long-term objective has not entered into force.

In 2018, there were no exceedance of the information threshold (hourly average 180 µg/m^3).

Table 4.1. O_3 in 2018. All parameters are based on one-hourly average values. The 8-hourly values are calculated as a moving average based on hourly measurements. Days above target value is the number of days that the maximum running 8-hour average exceeds 120 µg/m^3 averaged over 2016-2018.

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	7800	61	61	144	8	153
Aarhus/6160	7752	58	58	130	7	144
Odense/9159	7535	61	61	148	14	169
Aalborg/8158	7931	59	59	131	8	159
Rural						
Risø/2090	7893	63	64	151	16	168
Keldsnor/9055	7635	61	61	143	10	161
Ulborg/7060	7765	61	61	153	9	160
Traffic						
Copenhagen/1103	7773	42	41	110	0	126
Target value*	-	-	-	-	25	-
Long term objective	-	-	-	120	-	-
Information threshold	-	-	-	-	-	180
Data capture**	>7446	-	-	-	-	-

*) As average over 3 years.
**) 90% data capture of number of hourly measurements in relation to total number of hourly measurements in 2018 excluding hours used for calibration.

4.2 Trends

The long-term trends in O_3 concentrations are shown in figure 4.1. The annual averages of O_3 have a slightly increasing slope. This trend is most likely due to a decrease in the local (Danish) NO pollution; NO reacts with the O_3 to form NO_2. The reaction can be indirectly observed in our online data (https://envs2.au.dk/Luftdata/Presentation/table/Copenhagen/HCAB) for H. C. Andersen Boulevard each rush hour where the O_3 level decreases. Thus, it appears that the local NO pollution has decreased more than the ozon pollution. The Danish and European reductions of the precursors to O_3 formation (NO_x, volatile organic compounds) have therefore not been sufficient to reduce the O_3 concentrations. The maximum concentrations of O_3 have been relatively constant for more than a decade, which is illustrated by the relatively constant maximum 8-hour average concentrations in figure 4.1.

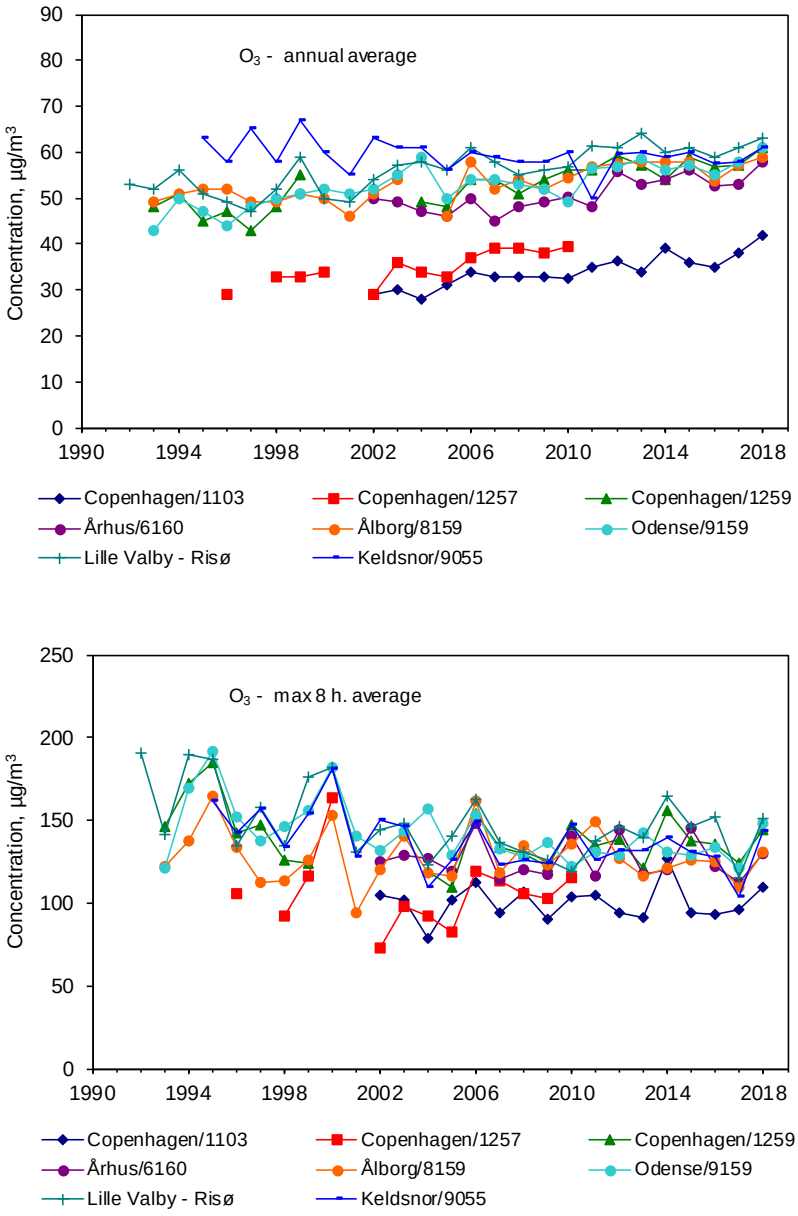


Figure 4.1. Annual average values and the max. 8-hour average value of O_3. The latter is calculated as 8-hourly running averages according to the provisions in the EU Directive (EC, 2008). Results from the previous (6159) and the new background station (6160) in Aarhus are shown on the same curve.

4.3 Results from model calculations

The annual mean concentration of O_3 is roughly on the same level throughout Denmark (figure 4.2). This is because the main production of O_3 takes place in the southern part of Europe and is subsequently long-range transported to Denmark. At the coasts, the concentrations are slightly higher than over the remaining land areas, since O_3 is deposited faster over land than over sea. In the cities, the concentrations are lower than the average for the monitoring stations, since O_3 is degraded by nitrogen oxide emitted from mainly traffic in the cities. This effect is seen for Copenhagen.

The target value for protection of human health is the running 8-hour mean concentration of O_3 that 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 objective is that the running 8-hour mean concentration of O_3 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 2018 show that the number of days with maximum daily 8-hour mean value above 120 µg/m^3 was well below the target value for the entire country in 2018. The target value that is determined as an average over three years (2016-2018), was not exceeded, since the number of days with exceedances in 2016 and 2017 were well below 25 as well (Ellermann et al., 2017, 2018).

The highest number of days with exceedance of 120 µg/m^3 was seen at coastal areas, where the maximum number of days typically was 4-5 days and with levels above 120 µg/m^3 (figure 4.3). For the main part of Denmark, the days with ozone levels above 120 µg/m^3 was 2-3. This is somewhat lower than observed in the measurements. The reason is that the model tend to underestimate the episodes with high ozone concentrations. This discrepancy is most likely a result of the model not including emissions of O_3 precursors from wild fires that are known to increase episodic O_3 concentrations.

In 2018 the long term objective were exceeded in all parts of Denmark. However, the long-term objective has not entered into force jet. In 2018, the highest 8-hour mean concentrations were observed at Sealand (figure 4.4).

According to the directive (EC, 2008), the public has to be informed when the 1-hour average concentration exceeds the information threshold of 180 µg/m^3. Neither measurements nor model calculations showed exceedances of the threshold in 2018 (figure 4.5).

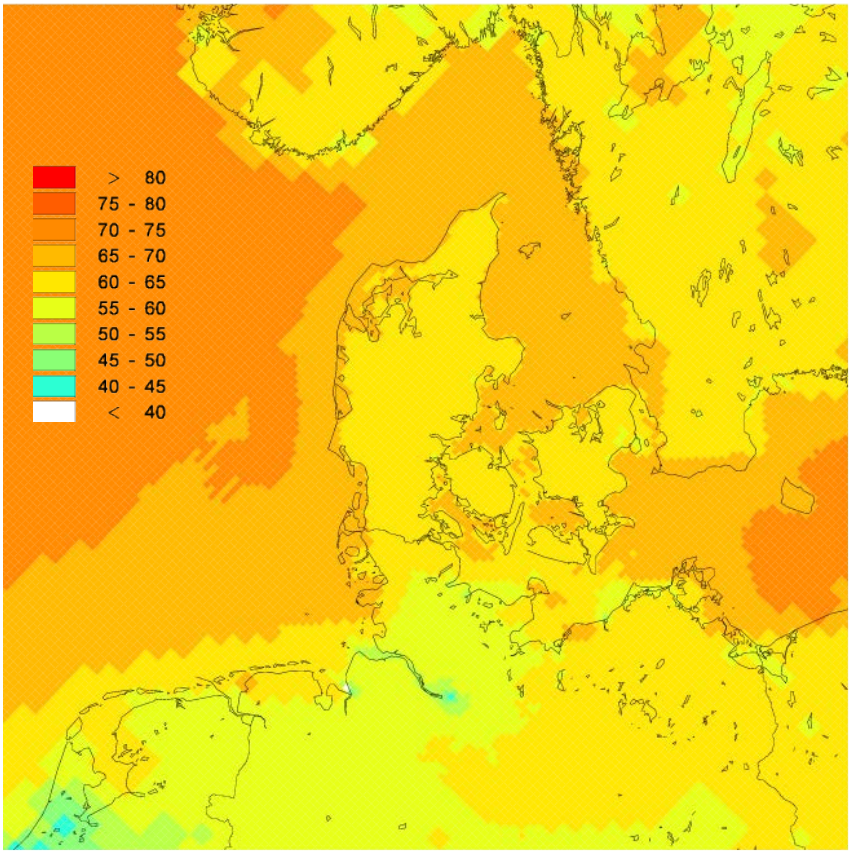


Figure 4.2. Annual mean concentrations of O_3 (µg/m^3) for 2018 calculated using DEHM. The figure shows the average concentrations for the 6 km x 6 km grid cells used in the model.

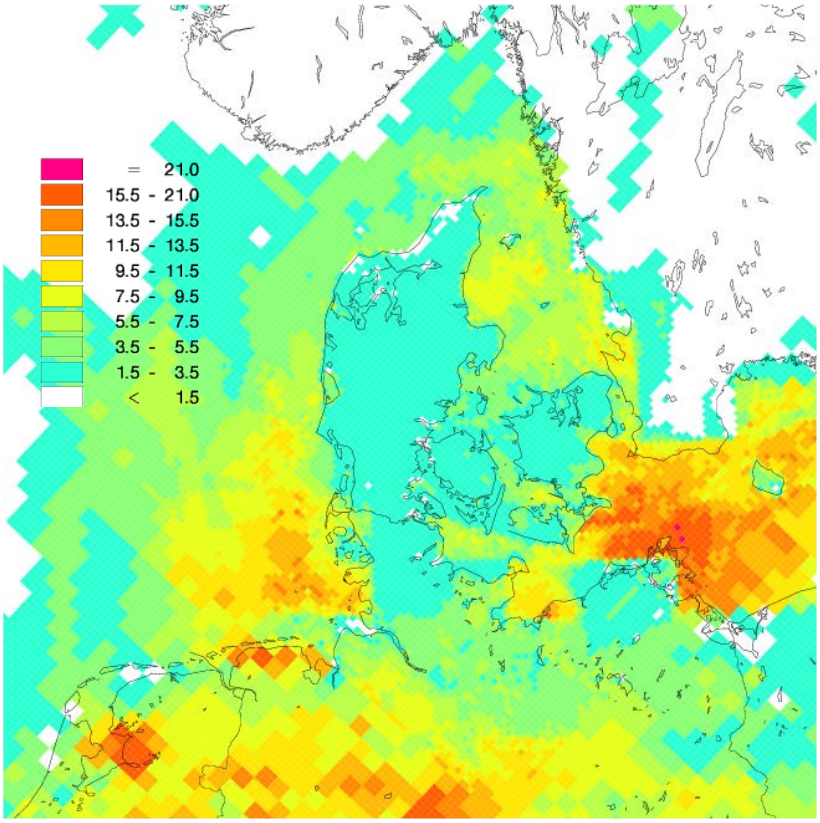


Figure 4.3. Number of exceedances of 120 µg/m^3 for 8-hour running mean concentrations of O_3 in 2018. The calculations were carried out using DEHM.

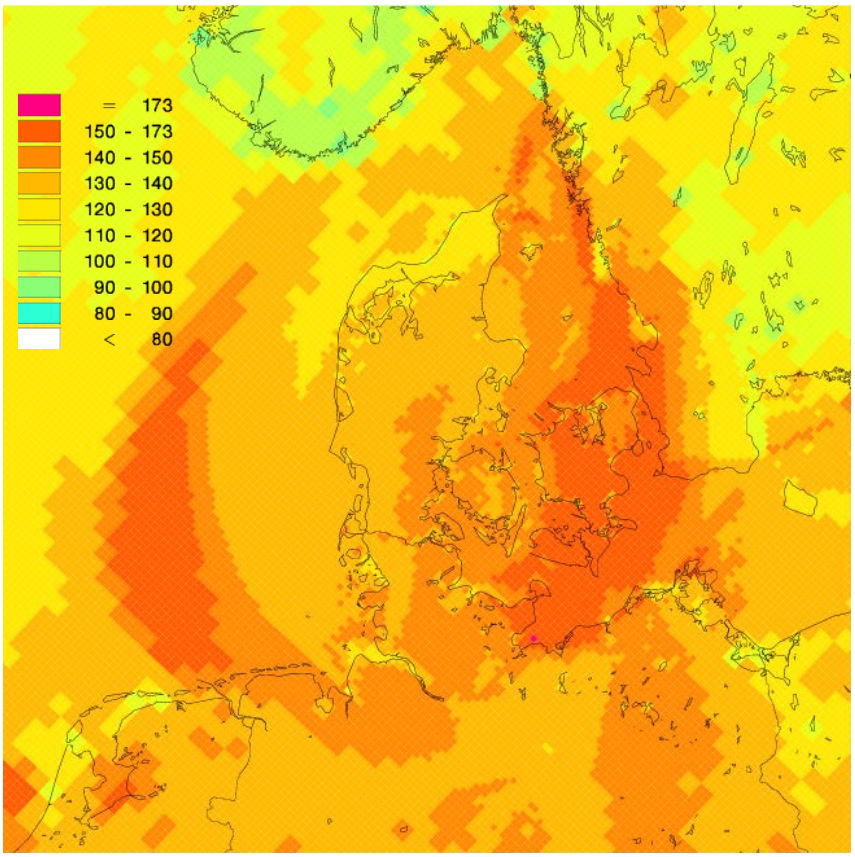


Figure 4.4. Maximum 8-hour running mean concentration (µg/m^3) of O_3 in 2018 calculated using DEHM.

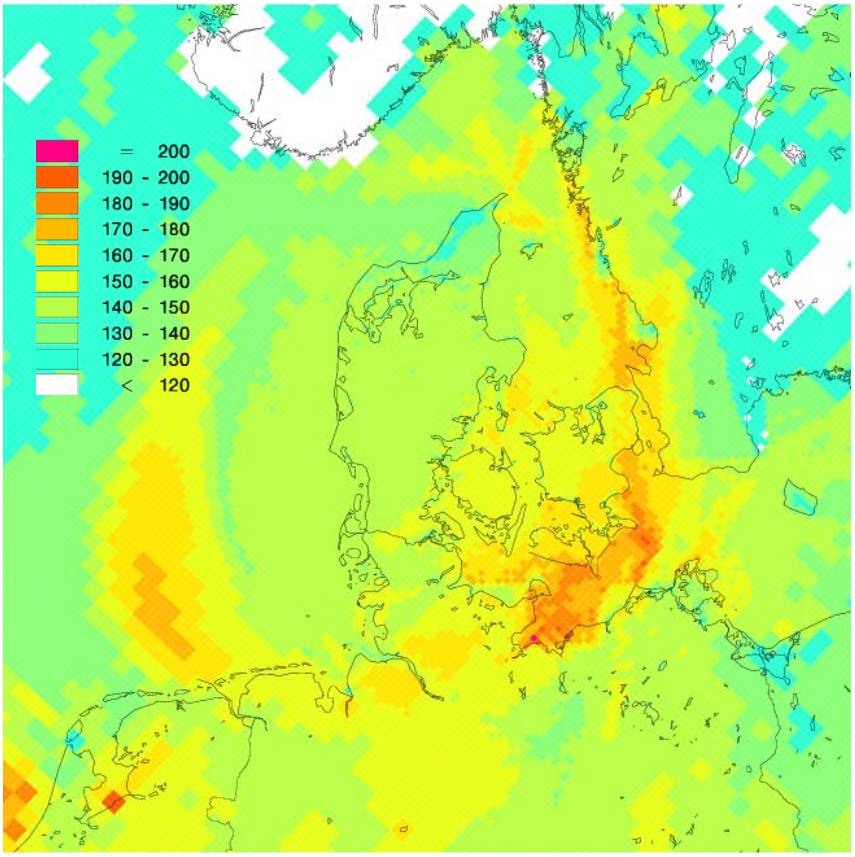


Figure 4.5. Maximum 1-hour mean concentration of O_3 (µg/m^3) in 2018 calculated using DEHM.

5. Carbon monoxide

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

5.1 Annual statistics

The annual statistics for 2018 for CO are shown in table 5.1. The limit value for CO is based on the maximum daily 8-hour average concentration that must not exceed 10,000 µg/m^3 (EC, 2008). This limit value was not exceeded at any of the stations.

Table 5.1. Annual statistics for CO in 2018. All parameters are based on hourly average. The 8-hour values are calculated as a moving average based on hourly results.

Unit: µg/m^3	Number	Average	Median	98-percentile	99.9-percentile	Max. 8-hours	Max. hour
Traffic:							
Copenhagen/1103	8047	326	309	619	926	1683	3611
Århus/6153	8322	243	230	477	726	691	1326
Odense/9156	8307	218	198	513	858	857	1385
Aalborg/8151 §	0	-	-	-	-	-	-
Urban Background:							
Copenhagen/1259	8131	189	180	344	548	551	582
Rural:							
Risø	8213	210	202	378	603	635	667
Data capture*	>7446	-	-	-	-	-	-
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 2018 excluding hours used for calibration.

§) For Aalborg/8151 (traffic) there is no data since the station has been shut down due to construction work at the site. Measurements are reinitiated at the street station in Aalborg ultimo 2019.

5.2 Trends

The long-term trends for CO concentrations 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 8-hour average concentrations. The reductions are due to national and international regulation of the emissions, among others by requirement of catalytic converters on all vehicles.

At the street stations in Odense/9155 (Albanigade) there was a larger reduction in CO from 2013 to 2015 than at the other stations. This is due to a major permanent rearrangement of the roads in Odense that resulted in a large reduction in the traffic intensity in Albanigade. The street station in Odense was therefore relocated to Grønløkkevej (Odense/9156) where measurements started in June 2016.

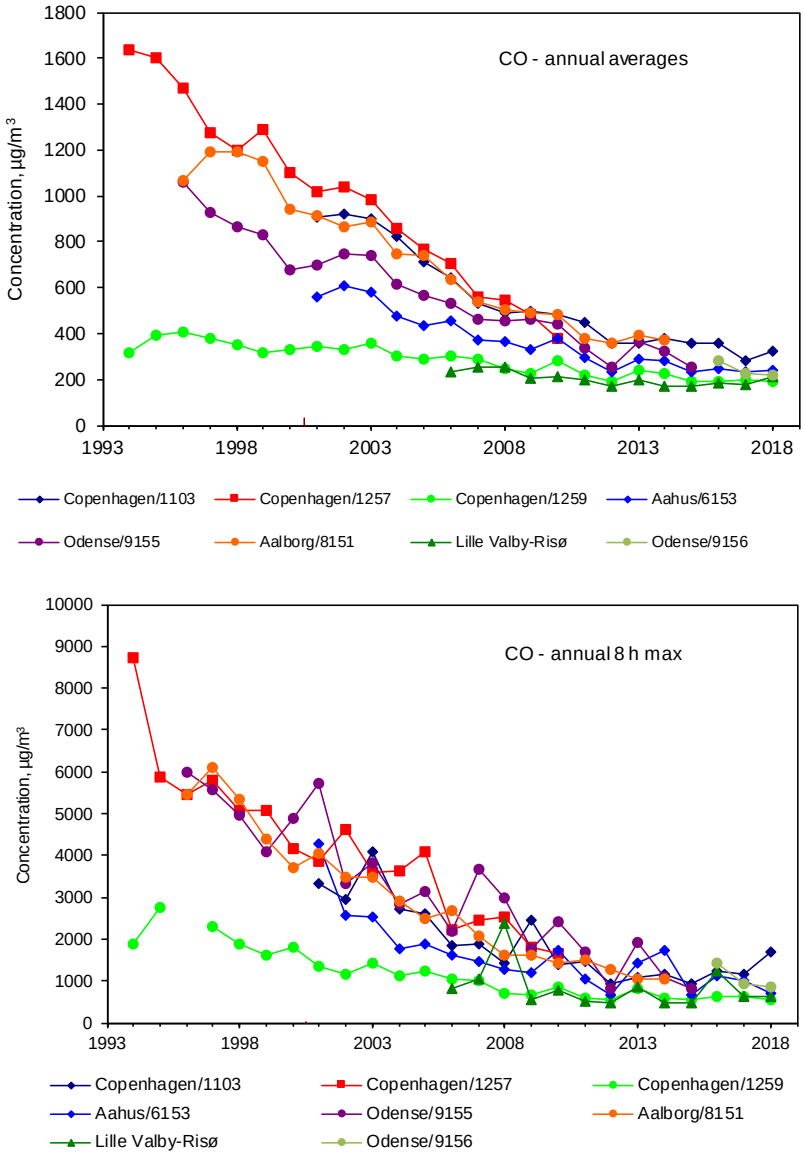


Figure 5.1. Annual average values and highest 8-hour values calculated based on an hourly moving average of CO. The site in Odense/9155 (Albanigade) was due to a major permanent rearrangement of the roads in Odense. It changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on 28 June 2014. A new street station was opened in Odense at Grønnelykkevej in June 2016.

6. Benzene and other Volatile Organic Compounds

This chapter presents the reults from measurements of ozone precursors in urban background as well as aromatic compounds at kerbside stations in the city of Copenhagen, all of which are Volatile Organic Compounds (VOC).

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

Benzene and toluene are additionally measured in urban background (Copenhagen/1259) along with 16 other potential O_3 precursor VOCs in diurnal time resolution. The focus is VOCs of anthropogenic origin, though isoprene which is mainly a biogenic compound emitted from deciduous trees is also included. Air is sampled and preconcentrated on Carbopack X adsorbent tubes and analyzed using Thermal Desorption Gas Chromatography Mass Spectrometry (TD-GC-MS).

6.1 Annual statistics and trends

Annual averages of benzene and toluene are listed in table 6.1 and 6.2 for 2018. Benzene is well below the EU-limit value of 5 µg/m^3 (EC, 2008), averaging 0.61 and 0.63 µg/m^3 at the kerbside stations 1257 and 1103, and 0.45 µg/m^3 in urban background. Thus, the local input of benzene from traffic amounts to 26 % of the concentration at the kerbside station 1257. For toluene, the local input is 38 %. Next to traffic exhaust, residential wood combustion is an important source of benzene, and for this reason the summer concentrations of benzene are lower even at kerbside stations. Both kerbside stations in Copenhagen report similar concentrations of anthropogenic aromatic compounds, including toluene and benzene (table 6.1), in spite of their differences with respect to traffic load and buildings close to the street. These VOCs decreased dramatically at the kerbside stations during 2004-2008 (figure 6.1) and has continued to do so, though at a slower yet comparable rate in the urban environment. Benzene has decreased by 54 % and 40 % at the kerbside station 1257 and urban background 1259, respectively, from 2010 to 2018. With respect to toluene, the corresponding decreases were 53 % and 29 %, respectively. Of the monitored VOCs at kerbside, toluene is by far the most abundant. Other aromatic compounds are comparable in abundance to benzene (table 6.1).

Table 6.1. Annual statistics for benzene, toluene, ethylbenzene and xylenes in 2018 based on weekly average concentrations (µg/m^3) at kerbside stations Jagtvej (1257) and H. C. Andersens Boulevard (1103) at 1 atm., 293 K. The limit value for benzene is 5 µg/m^3 (EU Directive 2008/50/EC).

Concentration µg/m^3	Copenhagen/1103	Copenhagen/1257	Number of results
Benzene	0.63	0.61	52; 51
Toluene	1.54	1.55	52; 51
Ethylbenzene	0.35	0.34	52; 51
m/p-Xylene	0.60	0.62	52; 51
o-Xylene	0.51	0.51	52; 51

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

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

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 are estimated to about 0.6-0.7 µg/m^3 for all the three street stations in Aarhus, Odense and Aalborg in 2018.

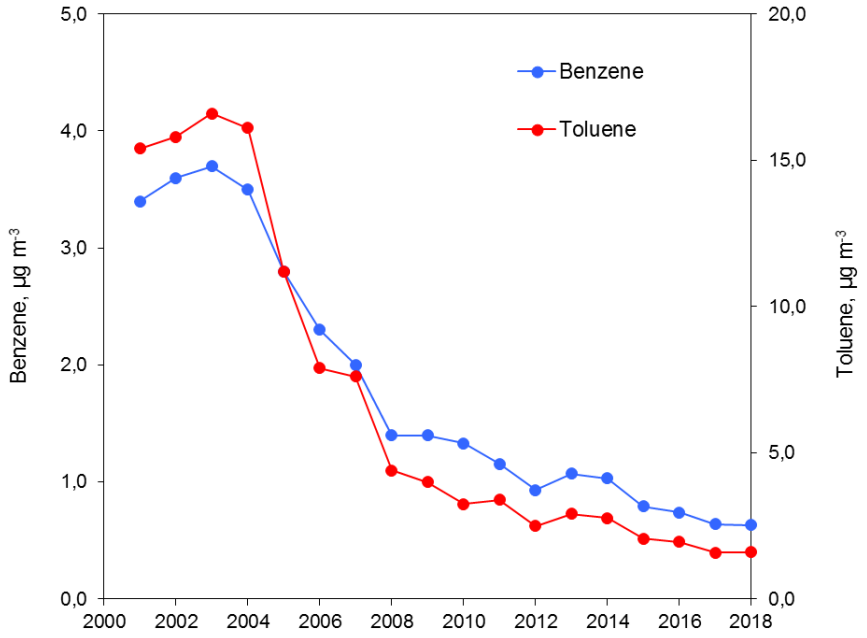


Figure 6.1. Trend in benzene and toluene (annual averages) on the kerbside station Jagtvej, Copenhagen/1257.

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

Table 6.2. Annual statistics for VOCs in urban background, Copenhagen (1259) for 2018 based on daily average concentrations (1 atm., 293 K).

Concentration (µg/m^3)	**Annual average 2010**	**Annual average 2018**	**Data coverage**
1-Pentene	0.04	0.04	73%
n-Pentane	0.53	0.59	73%
Trans-2-pentene	0.02	0.02	73%
Isoprene	0.03	0.08	73%
2-Methylpentane	0.31	0.19	73%
n-Hexane	0.19	0.13	73%
Benzene	**0.75**	**0.45**	73%
n-Heptane	0.28	0.18	73%
2,2,2-Trimethylpentane	0.10	0.04	73%
Toluene	**1.36**	**0.96**	73%
n-Octane	0.08	0.05	73%
Ethylbenzene	0.28	0.22	73%
m,p-Xylene	0.78	0.70	73%
o-Xylene	0.41	0.29	73%
1,3,5-Trimethylbenzene	0.10	0.01	73%
1,2,4-Trimethylbenzene	0.34	0.14	73%
1,2,3-Trimethylbenzene	0.09	0.04	73%
Sum of VOCs	5.68	3.76	

Measurements of mainly anthropogenic VOCs in urban background, which may act as O_3 precursors, were initiated in 2010 in the urban background. The major O_3 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 (table 6.2).

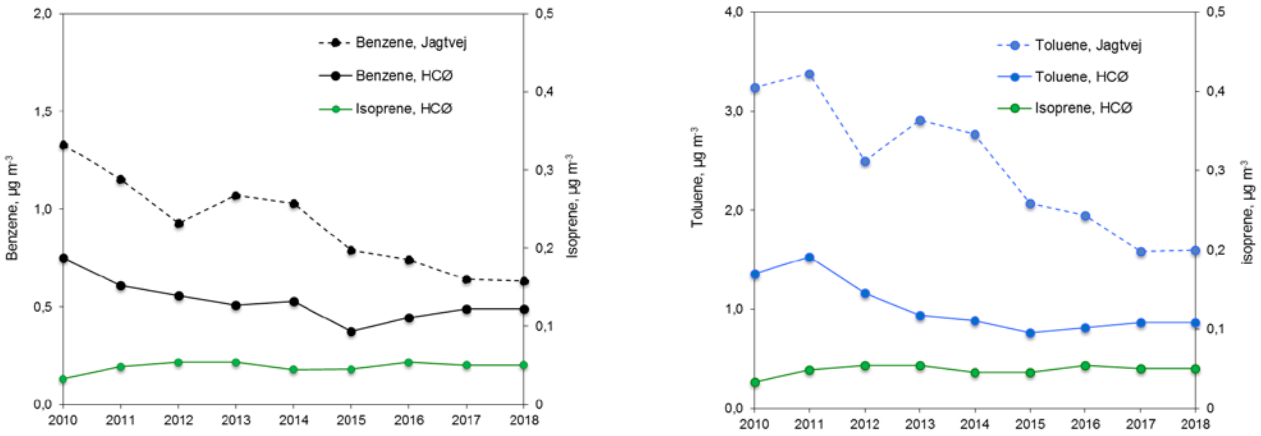


Figure 6.2. Annual average concentrations of benzene (left) and toluene (right) at the kerbside station at Jagtvej, Copenhagen/1257, and at urban background HCØ, Copenhagen/1259 for 2018. Isoprene - that is predominantly naturally emitted - is also shown for comparison

The annual isoprene concentration has remained fairly constant over the time period 2010 to 2017, but it increased markedly in 2018. Isoprene origins mainly from natural sources, e.g. terrestrial vegetation, and it peaks in the warmer summer months June, July and August with low concentrations in the winter months. On the contrary, the mainly anthropogenic compounds benzene and toluene have decreased in concentrations at comparable rates in both urban background and kerbside within this period (figure 6.2), though not as pronounced as from 2001-2008 (figure 6.1). Except for n-pentane, all anthropogenic VOCs either stayed constant during 2010-2018 or decreased.

The urban background ratio between toluene and benzene is somewhat smaller than the kerbside station 1257, i.e. 2.1 versus 2.5 reflecting the higher toluene/benzene ratio in traffic exhaust compared to e.g. the toluene/benzene ratio from biomass combustion in ambient air.

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

The measurements of particle mass (PM_{10} and $PM_{2.5}$) are today solely based on the EU's reference method (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 sampler (LVS) i.e. a flow of 2.3 m^3/hour on a diurnal basis with subsequent gravimetric determination of the sampled mass in the laboratory. Finally, the particle samples were analysed in the laboratory.

During the period from 2012 to 2016, the LVS-sampling method has gradually replaced the previously used SM200 beta (β) sampler (manufactured by OPSIS, Sweden). The LVS sampler collects particles on filters on a diurnal basis with subsequent determination of the sampled mass using β-absorption technique. This method is equivalent with the reference method. Comparison of the two methods have not documented any systematic deviation between the two measuring methods except for an improved reproducibility and data capture for the LVS instruments.

Additionally, PM is measured using TEOM (Tapered-Element Oscillating Microbalance) instruments at the Copenhagen street station HCAB (PM_{10} and $PM_{2.5}$), at the Aarhus street station (PM_{10}) and at the rural station at Risø (PM_{10}). The TEOM measurements have a time resolution of 30 minutes (table 7.3 and 7.4). During sampling, the collected particles are heated to 50°C. At this temperature, some of the semi-volatile particle constituents evaporate (mainly secondary aerosols and especially ammonium nitrate, NH_4NO_3). The loss depends on the actual composition of the aerosols. The measurements using TEOM has considerably uncertainty and they are therefore only used for near real time reporting of the data to the public.

Measurements of particle number concentrations have been carried out since 2001/2002 in cooperation between the monitoring programme and research projects funded by the Danish Environmental Protection Agency. The measurements have been performed using a Differential Mobility Particle Sizer (DMPS) that counts particles with mobility diameters between 6 and 700 nm. In 2015, additional measurements were initiated at the measurement station in Hvidovre using a Scanning Mobility Particle Sizer (SMPS) that counts particles with mobility diameters between 11 and 478 nm. In 2017, the instruments located at the street station at H. C. Andersen's Boulevard in Copenhagen, and at the regional background station Risø, were likewise replaced by two new SMPS systems. Subsequently it has been shown that the new SMPS instruments or the new inlets of the instruments cause problems with the measurements of the smallest particles. Intensive work has been carried out in 2017 and 2018 in order to solve these problems together with the manufacturer of the instruments. In 2019, it was found that the problems were due to technical issues with some of the new instruments and also in the delivered inlets. Data for the size range from 11 to 41 nm is therefore not presented for 2017 and 2018, and as a consequence, only data for particles larger than 41 nm is presented here.

In order to compare historical and new data together with investigating trends, only the size range from 41 – 550 nm (old systems) and 41 - 478 nm

(new systems) are presented and discussed in this report. The difference in the upper range for the two types of instruments do not influence the comparison between the two systems since the atmospheric particle numbers in the range from 478 (upper range on new systems) to 550 nm (upper range on old systems) are very low compared to the total number of particles in the range from 41-478 nm.

7.1 Annual statistics

In 2018, the permitted number of exceedances in a year of the diurnal limit value of 50 µg/m^3 for PM_{10} was not exceeded at any stations in the measuring network, even at stations where exceedances previously have occurred (the two traffic stations in Copenhagen (HACB/1103 and Jagtvej/1257)). Likewise, there were no exceedances of the annual limit value for PM_{10} (40 µg/m^3) and $PM_{2.5}$ (of 25 µg/m^3) 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 $PM_{2.5}$. In Denmark the average exposure indicator is measured in urban background at Copenhagen/1259, Aarhus/6159 and Aalborg/8158. For the years 2016-18 the AEI is determined to 10 µg/m^3 which is a decrease of about 30 % since 2010.

In 2018, the number of particles in ambient air in the range from 41- 478/550 nm was about 4,000 particles per cm^3 at the street station H. C. Andersens Boulevard (table 7.5). This is a factor of about two higher than at suburban, urban background and rural background.

Table 7.1. Annual statistics for PM_{10} in 2018. All parameters are given as diurnal averages at ambient temperature and pressure.

Unit µg/m^3	Number of results	Average (µg/m^3)	Median	Days above 50 µg/m^3	90-percentile	Max. day
Street						
Copenhagen/1103	364	31	29	22	46	104
Copenhagen/1257	351	25	23	8	39	94
Århus/6153	355	22	20	7	34	88
Odense/9156	339	23	21	9	37	88
Urban background						
Copenhagen/1259	361	18	16	4	29	84
Rural						
Risø	353	17	15	2	28	63
Keldsnor/9055	357	18	17	5	29	72
Limit value (2005)		40		35**		
90% data capture	>328*					

Measurements at all stations in 2018 were based on LVS with gravimetric determination of particle mass.

* 90% data capture of number of diurnal measurements in relation to the total number of days in 2018 (365).

**Permitted number of exceedances in a year of the diurnal limit value of 50 µg/m^3.

Table 7.2. Annual statistics for $PM_{2.5}$ in 2018. All parameters are given as diurnal averages at ambient temperature and pressure.

Unit µg/m³	Number of results	Average (µg/m³)	Median	90-percentile	Max. day
Street					
Copenhagen/1103	358	16	13	26	80
Copenhagen/1257	357	14	12	24	78
Aarhus/6153	359	14	11	26	70
Aalborg/8151*					
Suburban					
Hvidovre/2650	356	12	10	22	72
Urban background					
Copenhagen/1259	356	13	10	23	75
Aarhus/6159	354	12	10	21	66
Ålborg/8158	346	12	9	20	65
Rural					
Risø	352	12	10	23	58
Limit value (2015) (parenthesis gives proposed value for 2020)		25(20)			
90% data capture	>328**				

Measurements at all stations in 2018 were based on low volume sampling (LVS) with gravimetric determination of particle mass.

* No data from Aalborg/8151 (traffic site) in 2018 because the station is closed temporarily due to construction work.

**90% data capture of number of diurnal measurements in relation to the total number of days in 2018 (365).

Table 7.3. Annual statistics for PM_{10} measured in 2018 using TEOM. The values are based on ½-hourly averages. Total annual number of ½-hours is 17.520. Data are only used for near real time reporting to the public.

Unit: µg/m³	Number of results	Average
Street		
Copenhagen/1103	17283	31
Aarhus/6153	14495	18
Rural		
Risø	16963	14
Limit value		

Table 7.4. Annual statistics for $PM_{2.5}$ measured in 2018 using TEOM. The values are based on ½-hourly averages. Total annual number of ½-hours is 17.520. Data are only used for near real time reporting to the public.

Unit: µg/m³	Number of results	Average
Street		
Copenhagen/1103	17091	10
Limit value (2015) (parenthesis gives proposed value for 2020)		

Table 7.5. Annual statistics for particle number measured in 2018 in the range from 41 to 478/550 nm in diameter. All values are based on ½-hourly averages. Total annual number of ½-hours is 17,520. The low data capture at some of the stations are due to the technical problems with the equipment and all the tests that were applied to solve the problems. The difference in the upper range for the diameter has not significant influence on the particle number, since the number of particles in the range from 478-550 nm is very small.

Unit: particles per cm^3	Number of results	Average 41- 478/550nm)
Street		
Copenhagen/1103**	15363	4043
Urban Background		
Copenhagen/1259*	5857	2168
Suburban		
Hvidovre/2650**	10786	1969
Rural		
Risø**	12479	1956

* Measured with DMPS (41nm – 550 nm)

** Measured with SMPS (41nm – 478 nm)

7.2 Trends

Up to the year 2000, PM was measured as Total Suspended Particulate matter (TSP) corresponding to particles with a diameter up to around 25 µm (figure 7.1). The exact cut-off depends strongly on the wind velocity. From 2001 most of the measurements of particulate matter were changed from TSP to PM_{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 were initiated (figure 7.2). Although the measurements at HCAB (Copenhagen/1103) began five years later than most of the other PM_{10} measuring sites, the PM_{10} measurements at this station are also following a decreasing trend. However, this is mainly due to a major reduction (7 $\mu g/m^3$) 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 major permanent rearrangement of the roads in Odense. It changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on 28 June 2014. This has affected the measured PM_{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 display an increase in 2014 compared to 2013. In 2015, the road next to the measuring station was closed for traffic. PM_{10} measurements from Odense/9155 (Albanigade) for 2015 do not represent a traffic site but rather have character of an urban background site. In the process of relocating, the station the PM_{10} measurements were closed down the 15 June 2015. The PM_{10} measurements at the new traffic station in Odense/9156 (Grønløkkevej) were initiated 1 July 2016.

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 diurnal measurements of $PM_{2.5}$ until now. There seems to be a tendency towards a small reduction in $PM_{2.5}$, although this tendency is uncertain due to the relatively short period with measurements.

The AEI for $PM_{2.5}$ is determined as the average $PM_{2.5}$ measured at urban background in Copenhagen, Aarhus and Aalborg over a three-year period. Thus e.g. the 2010 AEI value represents the average of the years 2008-2010. The trend for AEI is shown in figure 7.4 and as seen for $PM_{2.5}$ itself, there is a small reduction in the AEI, although this tendency is uncertain due to the relatively short period with measurements, and the large inter-annually variation in $PM_{2.5}$ due to the natural variations in the meteorological conditions. Over the period 2010 to 2018 the AEI has been reduced with about 30 %.

The measurements show a significant reduction in the particle number concentrations for particles between 41 and 550 nm over the entire measuring period from 2002 to 2017 (figure 7.5). On the street station at H. C. Andersen's Boulevard, the number of particles in the range from 41 to 550 has decreased by more than 40 % during the period 2002 - 2017 in the presented size range. At the urban background station in Copenhagen, a similar trend is observed for the same period. A decrease was also observed at the rural background station at Risø though the decrease is much smaller. Trends at the suburban background station in Hvidovre cannot be investigated yet, as the time series started in 2015 and is hence too short to make reasonable conclusions.

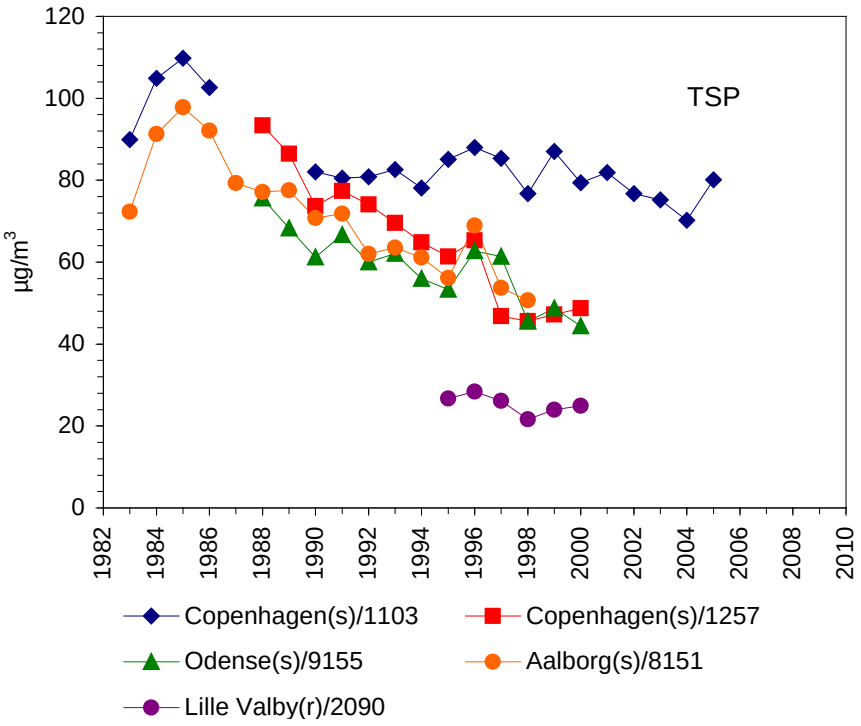


Figure 7.1.
Annual averages for TSP measured at street stations (s) and at a rural background station (r).

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 in method. Data is 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. Since 2017, all PM_{10} measurements are based on the LVS reference method.

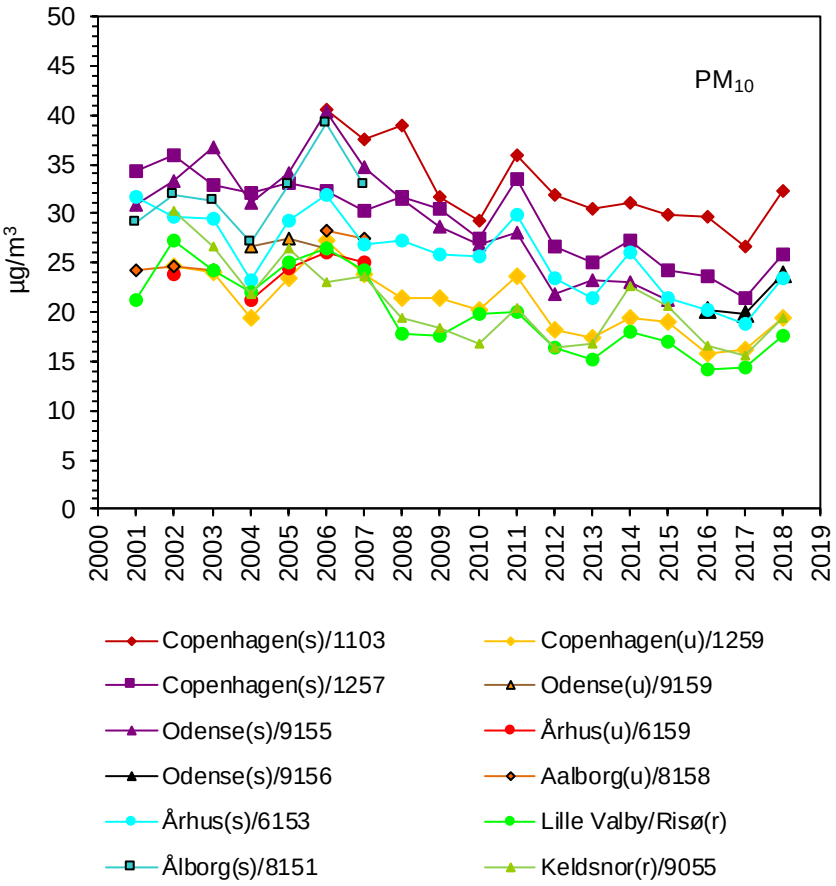


Figure 7.3.
Annual averages for $PM_{2.5}$ measured at street (s), suburban (sub), urban background (u) and at a rural background station (r). Only annual averages covering more than 2/3 of the years are shown except for the newly established suburban station at Hvidovre (began in 17 June 2015) and Aalborg(s) for 2014 (data covering the period 1/1 - 7/9). Data is 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. Since 2016, all $PM_{2.5}$ measurements are based on the LVS reference method.

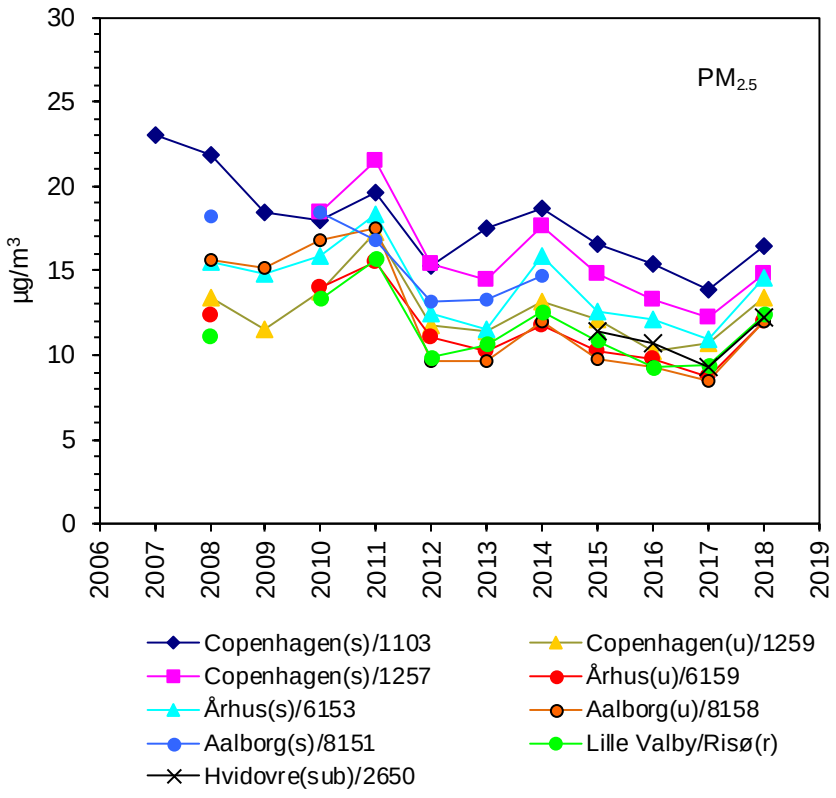


Figure 7.4.
The trend for AEI for $PM_{2.5}$. AEI is determined as the average $PM_{2.5}$ measured at urban background in Copenhagen, Aarhus and Aalborg averaged over a three years period. Data is given at ambient temperature- and pressure conditions. The value shown for 2010 corresponds to the average of the concentrations for 2008 to 2010 and likewise for the other years.

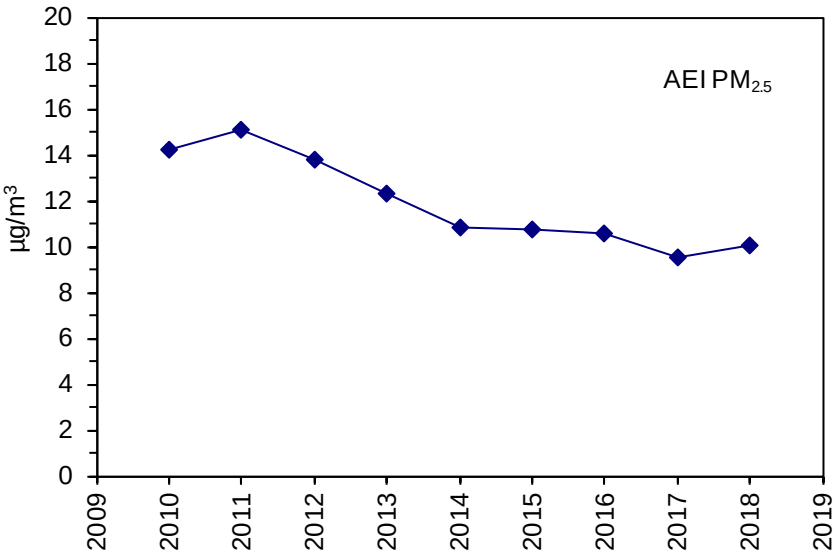
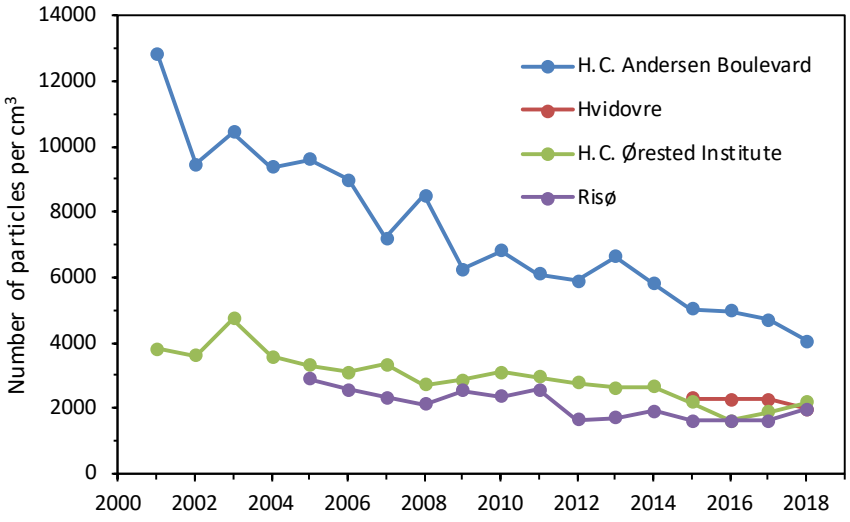


Figure 7.5. Annual averages for number of particles per cm^{-3} in the range from 41 to 478/550 nm at the street station at H. C. Andersens Boulevard, urban background station at H. C. Ørsted Institut, suburban station in Hvidovre and rural background station at Risø. At Hvidovre the numbers represent particles in the range from 41-478 nm measured with the new instrument type. At H. C. Ørsted Institut only the old instrument type has been used and these numbers represents particles in the range from 41 – 550 nm. At H. C. Andersens Boulevard and Risø measurements have been carried out with the old instrument type (41-550 nm) up to 2017 and in 2017 the new instrument type 41-478 nm) has been used. The difference in upper cut of range for the particle size do no change the values measured since the number of particles in the range from 478 – 550 nm is very small.



7.3 $PM_{2.5}$ and PM_{10} modelled concentration for Copenhagen and Aalborg

Model calculations of $PM_{2.5}$ and PM_{10} for selected streets in Copenhagen (capital) and Aalborg (fourth largest city) have been reported within the Danish Air Quality Monitoring Program since the start of 2017. 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 are included for Copenhagen and 31 for Aalborg. ADT (Average Daily Traffic) was between 5,100 and 79,400 vehicles/day in Copenhagen and between 2,700 and 29,000 vehicles/day in Aalborg.

Model calculations have been carried out in order to determine the annual concentrations of $PM_{2.5}$ and PM_{10} for comparison with the limit values. The air quality limit value for the annual mean is 25 and 40 µg/m^3 for $PM_{2.5}$ and PM_{10}, respectively (EC, 2008).

Modelled $PM_{2.5}$ and PM_{10} concentrations for Copenhagen are shown in figures 7.6 and 7.7, respectively. The rank numbers from the ranking of NO_2 is maintained and street numbers are shown in table 7.6.

Concentrations are well below the annual mean limit value for $PM_{2.5}$ of 25 µg/m^3 and the annual mean limit value for PM_{10} of 40 µg/m^3.

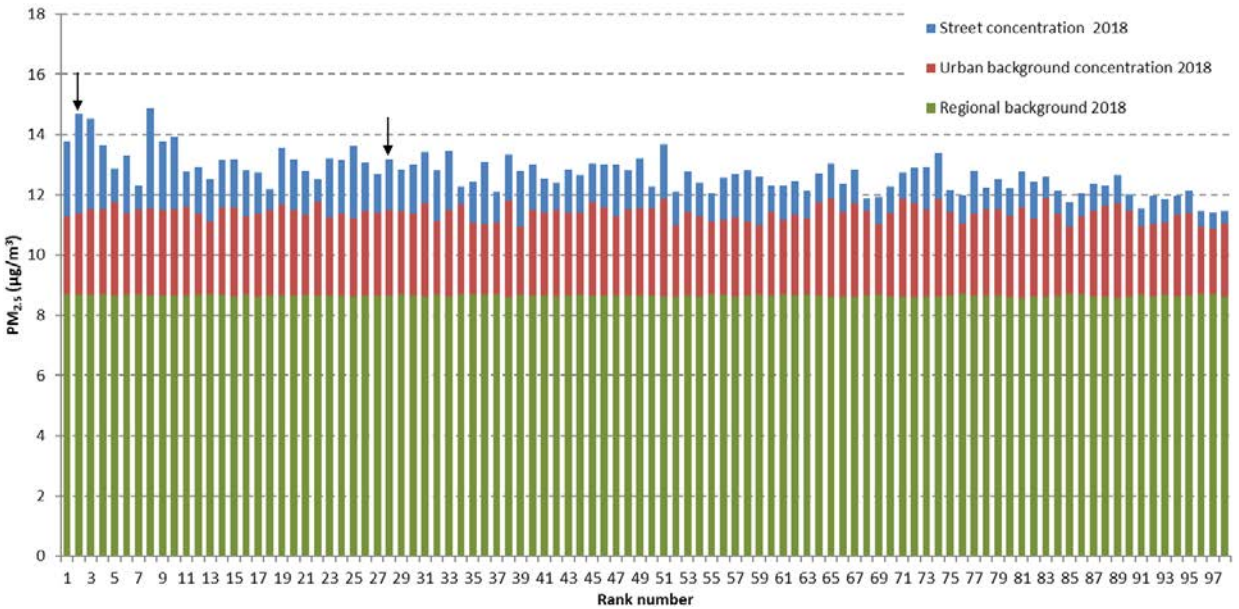


Figure 7.6. Modelled $PM_{2.5}$ concentrations for Copenhagen in 2018. The streets are ranked according to the concentrations of NO_2 (Chapter 3.3). Arrows indicate street segments with a measuring station.

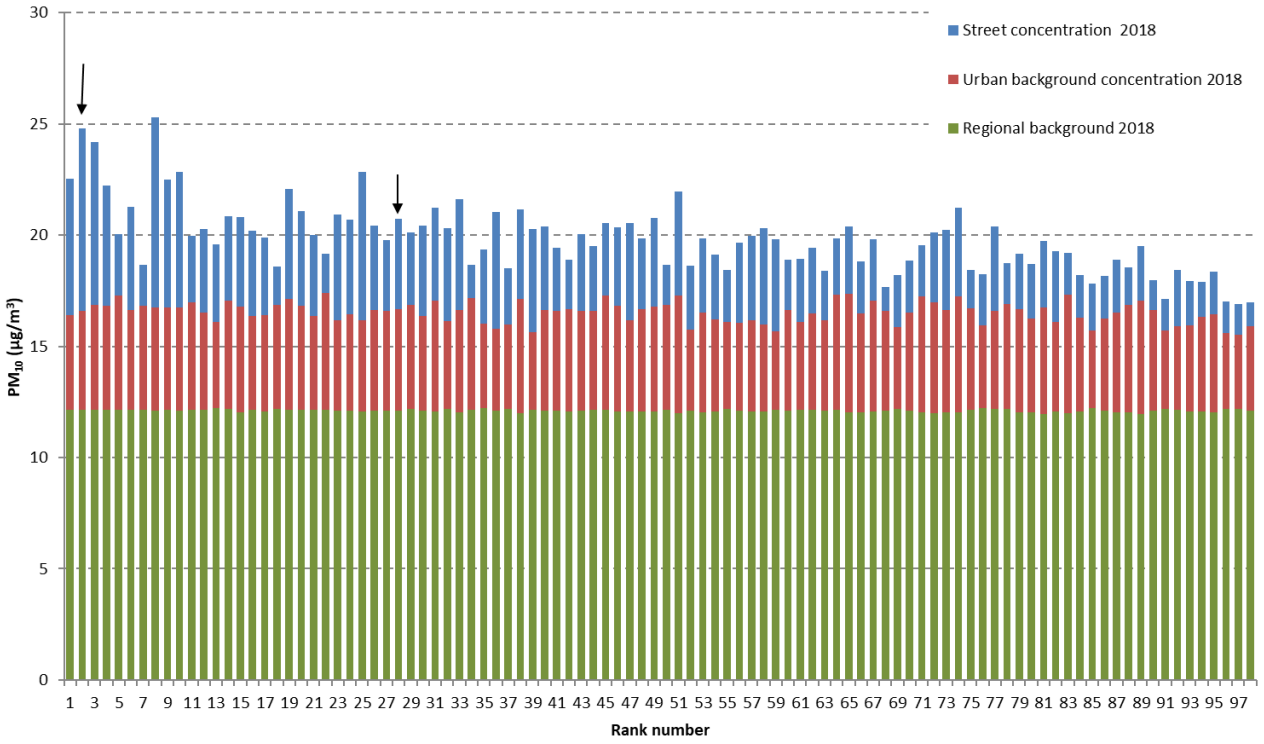


Figure 7.7. Modelled PM_{10} concentrations for Copenhagen in 2018. The streets are ranked according to the concentrations of NO_2 (Chapter 3.3). Arrows indicate street segments with a measuring station.

Modelled $PM_{2.5}$ and PM_{10} concentrations for Aalborg are shown in figure 7.8 and figure 7.9, respectively. The figures contain the same ranking as for NO_2 and street names are shown in table 7.6.

Concentrations are well below the annual mean limit value for $PM_{2.5}$ of 25 µg/m³ and the annual mean limit value for PM_{10} of 40 µg/m³.

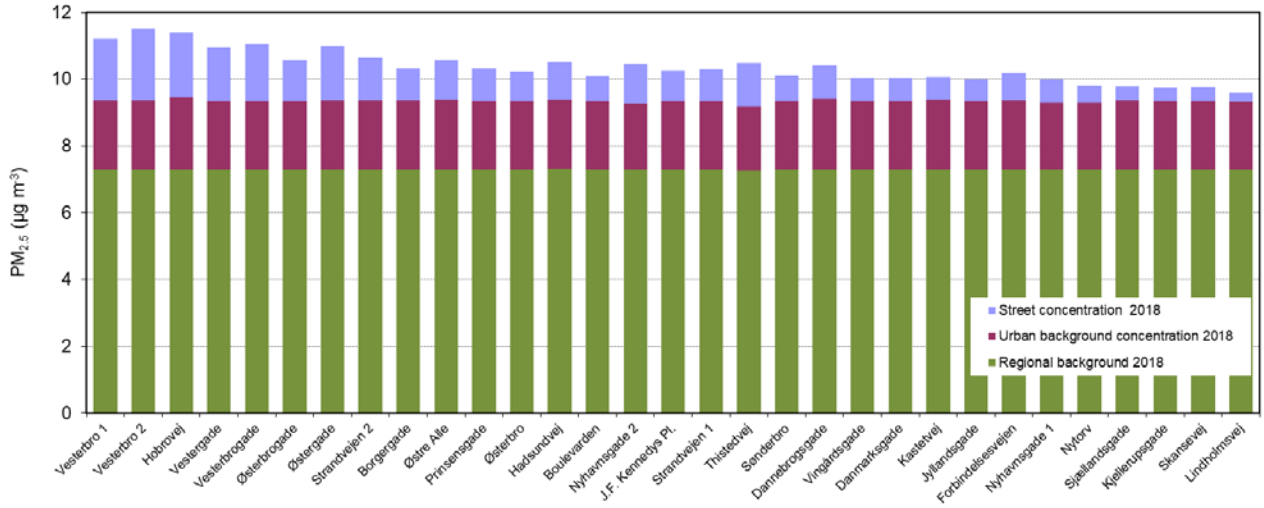


Figure 7.8. Modelled $PM_{2.5}$ concentrations for Aalborg in 2018

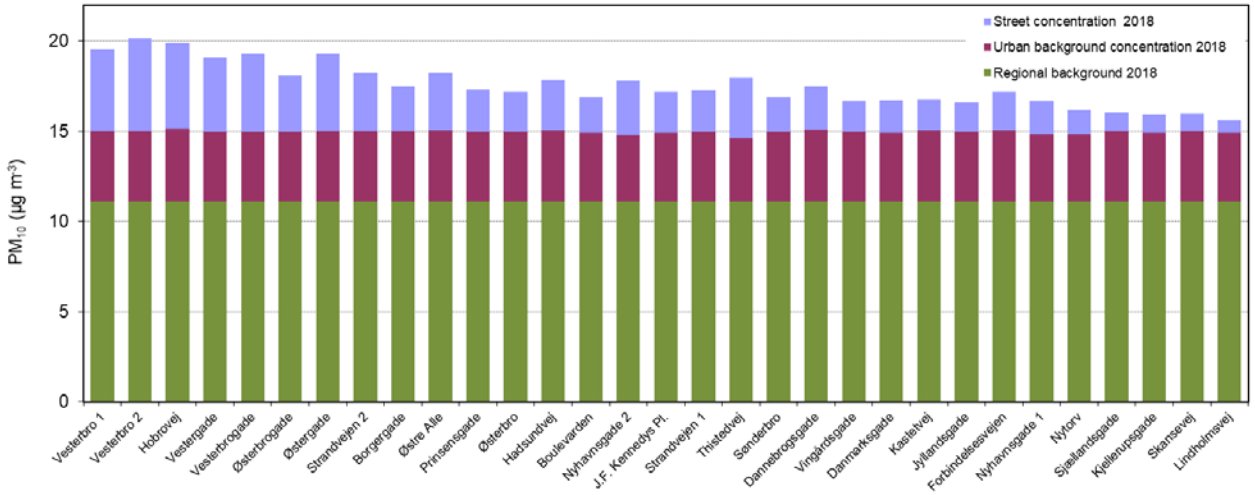


Figure 7.9. Modelled PM_{10} concentrations for Aalborg in 2018.

Table 7.6. Rank number and names for the street segments that are shown in figure 7.7 and 7.8. The streets are numbered (1-98) according to NO_2 levels in 2018 (1 = highest, 98 = lowest) (See chapter 3). The numbers in parentheses refer to different segments of the same street that has more than one model calculation. An asterisk (*) indicates a street segment with a measurement station.

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

8. Heavy metals

The EU Directives 2004/107/EC (EC, 2005) requires that the concentrations of arsenic (As), cadmium (Cd) and nickel (Ni) have to be measured in PM_{10}, and the directives lay down target values for these compounds. Similarly the EU Directive 2008/50/EC (EC, 2008) requires measurements of lead (Pb) and lay down a limit value for Pb.

In accordance with the directives, metals in PM_{10} are measured by collection of PM_{10} on filters that are analyzed by ICP-MS (Inductively Coupled Plasma Mass Spectrometry) for their content of the four regulated metals and 6 additional metals (vanadium (V), chromium (Cr), manganese (Mn), cupper (Cu), zink (Zn), and selenium (Se)) The heavy metals in PM_{10} are measured at two street stations (H. C. Andersens Boulevard (HCAB), Copenhagen; Banegaardsgade; Aarhus) and at the urban background station in Copenhagen (HCØ). These compounds are also measured at the rural background station Risø in total suspended particulate (TSP). The content of these metals in PM_{10} and TSP are to a good approximation equal at the rural measurement station Risø since these metals are mainly found in particles with diameter below 10 μm.

The EU directive 2004/107/EC (EC, 2005) requires furthermore that mercury (Hg) has to be measured. However, these measurements can be carried out in cooperation with neighbouring countries since the spatial variation in mercury is very small. 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.

8.1 Annual statistics

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

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 2018. For comparison, the table also includes results for these metals measured in TSP at the rural background station Risø.

Unit ng/m^3	V	Cr	Mn	Ni	Cu	Zn	As	Se	Cd	Pb
PM_{10}, Street										
Copenhagen/1103	2.4	7.9	22	1.7	75	43	0.7	0.5	0.09	3.5
Aarhus/6153	1.4	3.1	6.7	0.5	19	15	0.5	0.4	0.06	2.0
PM_{10}, Urban background:										
Copenhagen/1259	2.1	2.4	4.5	0.9	9	11	0.5	0.5	0.06	2.0
TSP, Rural background										
Risø	1.3	0.6	3.7	0.5	4.1	11	0.4	0.4	0.06	1.9
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 from EU Directive 2008/50/EC (EC, 2008).

**) The guidelines and lifetime 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 Hg in 2018 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)	1.4	0.0015

8.2 Trends

The long-term trends for six of the heavy metals are shown in figure 8.1. For Pb, As, Ni and manganese (Mn) there are clear reductions in the concentrations due to national and international regulations of the emissions. The reduction is most pronounced for Pb where removal of Pb from gasoline has resulted in large reductions of the concentrations. For Cu there has not been any clear long-term change in concentration. Emissions in Denmark show a slight increase during the period from 1990 to 2013 (DCE, 2017).

The long-term trend of Mn behaves differently at H. C. Andersen's Boulevard as compared to the other Danish stations. High Mn concentrations in the asphalt used at H. C. Andersen's Boulevard during the period 1991 - 2008 is believed to be the main reason for this. In 2008 the asphalt was replaced by a new type of asphalt. The abrupt decrease in concentration from 2005 to 2006 at HCAB is ascribed to a change in sample inlet. Annual averages up to 2005 is based on TSP, whereas annual averages are based on PM_{10} from 2006 and the change in particle fraction lead to the lower concentrations since Mn is found in large particles from the asphalt.

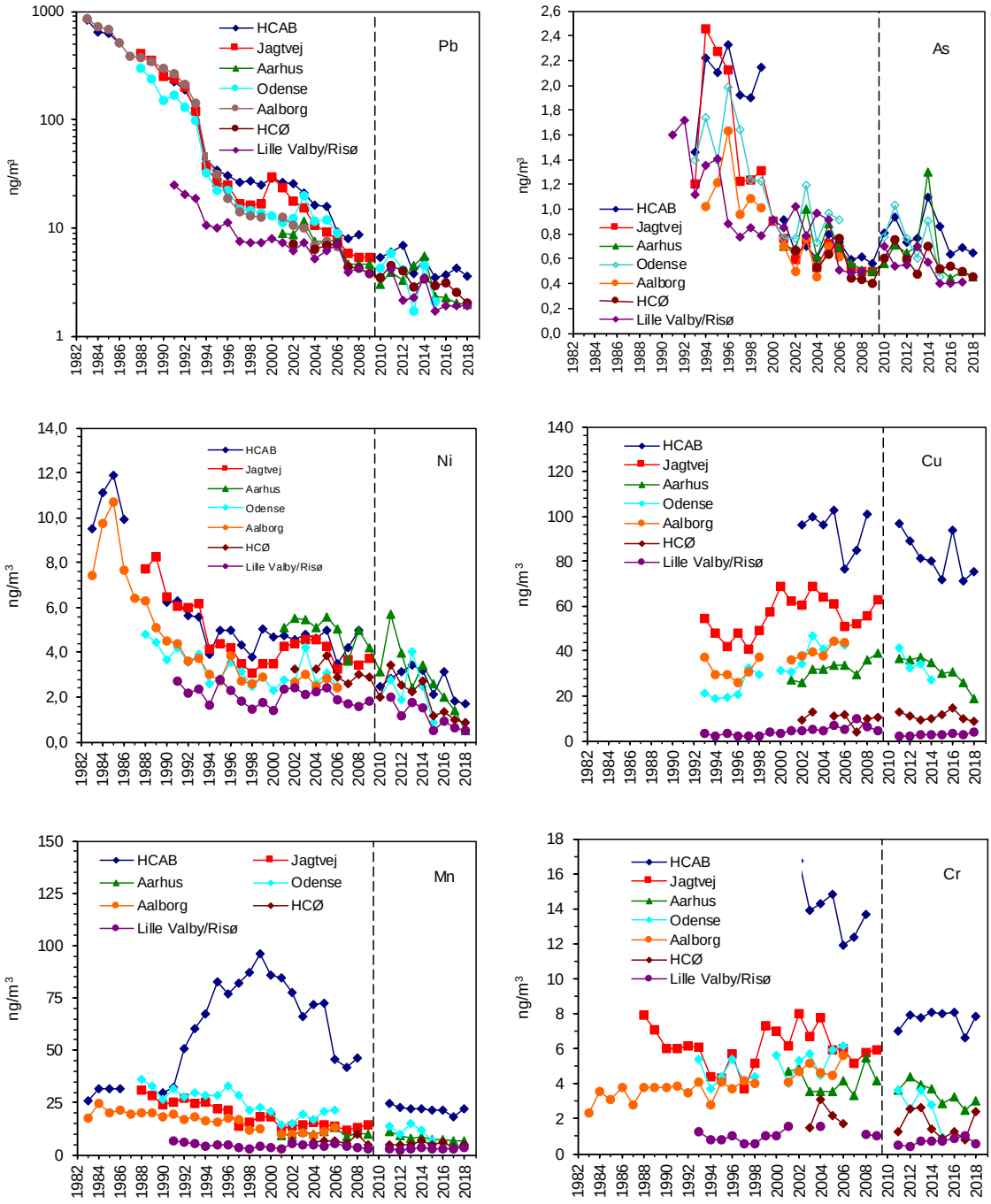


Figure 8.1. Annual averages from selected stations for some heavy metals in particulate matter. Until 2000 in TSP and later in PM_{10} – except for Copenhagen/1103 where PM_{10} replaced TSP from the beginning of 2006. The heavy metals are usually found in fine particles, which make the TSP and the PM_{10} values comparable. An exception is road dust and especially for Mn the values found in TSP is higher than in PM_{10}. Note that the scale for Pb is logarithmic. The dashed line indicates that the analysis method has been changed from 2009 to 2010.

9. Sulphur dioxide

The concentration of sulphur dioxide (SO_2) has reached very low levels in Denmark, and it is therefore only necessary to perform a limited monitoring of the concentrations; both with respect to the number of stations and the quality of the measurements. Hence, this is only measured at two traffic stations (Copenhagen and Aalborg) with focus on episodes with high concentrations of SO_2. It is measured using gas monitors based on ultraviolet fluorescence. The concentrations of SO_2 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 of minutes that is aggregated to hourly averages for this report.

9.1 Annual statistics

The annual statistics for 2018 for SO_2 are shown in table 9.1. None of the limit values (EU, 2008) were exceeded in 2018. In 2018, there was no information to the public due to exceedance of the alert threshold for SO_2 (one-hour average 500 $\mu g/m^3$).

Table 9.1. Annual statistics for SO_2 in 2018. All parameters are calculated based on hourly average. The detection limit for the monitors is a few $\mu g/m^3$, which makes the average and median values encumbered with high relative uncertainties.

Unit: $\mu g/m^3$	Number of results	Average year	Average winter	Median	98-percentile	Max. Hour	4th highest diurnal mean
Traffic:							
Copenhagen/1103	8001	1.0	1.3	0.7	4.5	12.2	5.1
Aalborg/8151 §	0	-	-	-	-	-	-
Limit values	>7446*	20	20			350	125

*) 90% data capture of number of hourly measurements in relation to total number of hourly measurements in 2018 excluding hours used for calibration.

§) Aalborg/8151 (traffic) there is no data since the station has been shut down due to construction work at the site. Measurements are not reinitiated at the street station in Aalborg in 2018 before ultimo 2019.

9.2 Trends

The long-term trends for SO_2 concentrations are shown in figure 9.1. Since the beginning of the 1980s, the annual concentrations have decreased by a factor of ten or more due to effective national and international regulations of the emissions. The emission reductions are due to use of effective cleaning technologies in combination with the decrease of the sulphur content in fuel.

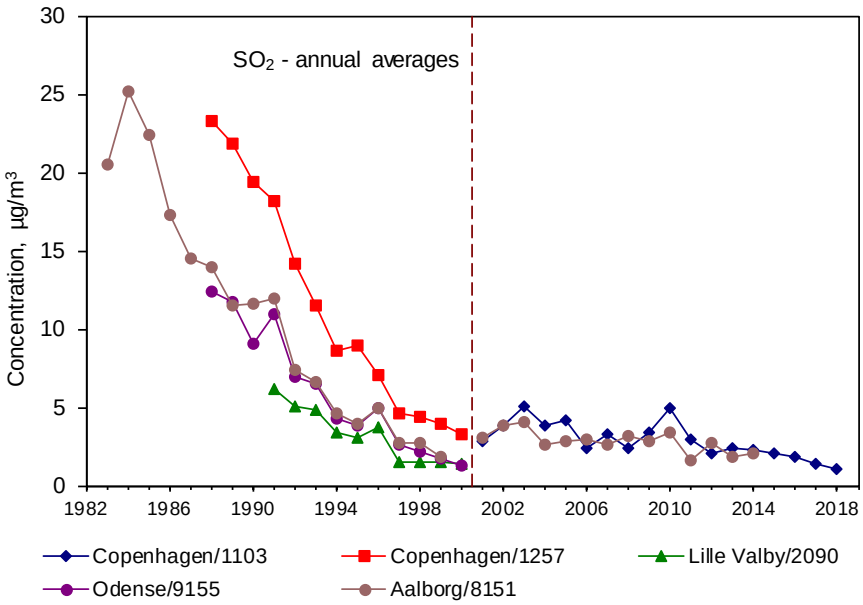


Figure 9.1. Annual averages for SO_2. Until 2001 the results were obtained using potassium hydroxide impregnated filters for collection of SO_2. These measurements ceased in 2000 and after 2000 the SO_2 measurements have been carried out using SO_2 monitors in order to monitor episodic results. The detection 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. The station in Aalborg (traffic) has temporarily been shut down due to construction work at the site. There is therefore no data from Aalborg from 2015 until 2018, which is the reporting year for this report.

10. Polyaromatic hydrocarbons

Following the EU Directive 2004/107/EC (EC, 2005), measurements of atmospheric concentrations of benzo[a]pyrene and other particle bound polyaromatic hydrocarbons (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.

10.1 Annual statistics

The average concentration of benzo[a]pyrene in 2018 was 0.25 ng/m^3 and 0.28 ng/m^3 at the street station on HCAB and the suburban station in Hvidovre, respectively. The average concentration of benzo[a]pyrene at HCAB has slightly increased with respect to the value in 2017 (0.18 ng/m^3), while the concentrations of all the other PAHs are substantially unchanged at both stations. Overall, it can be concluded that the target value for benzo[a]pyrene of 1 ng/m^3 was not exceeded in 2018.

Table 10.1 shows the average annual concentrations of benzo[a]pyrene and the other five PAHs listed in the EU Directive. There are no target values for these five compounds.

Table 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	0.25	0.28
Benzo[a]anthracene	0.17	0.20
Benzo[b]fluoranthene	0.28	0.41
Benzo[j+k]fluoranthenes	0.44	0.50
Indeno[1,2,3-cd]pyrene	0.32	0.34
Dibenzo[a,h]anthracene	0.04	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. The seasonal variation also seems to vary between the two measurements stations (table 10.2). The winter concentrations at Hvidovre are higher than at HCAB in 2013-2018 while the summer concentrations are at the same low level for most of the years. This is because the sources of benzo[a]pyrene in Hvidovre is largely wood burning for residential heating while the sources at HCAB are both wood burning and traffic. The seasonal variation in the emissions from traffic is small compared to that of wood burning.

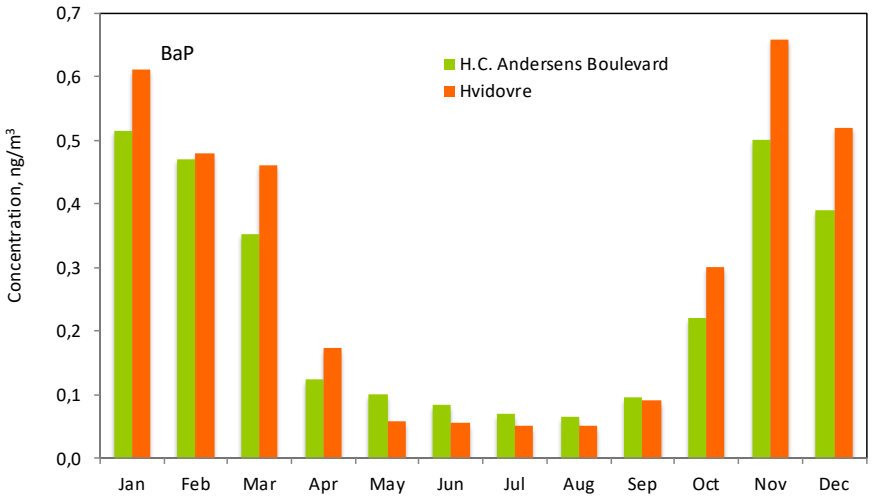


Figure 10.1. Monthly average concentrations of benzo[a]pyrene at H. C. Andersens Boulevard and Hvidovre in 2018.

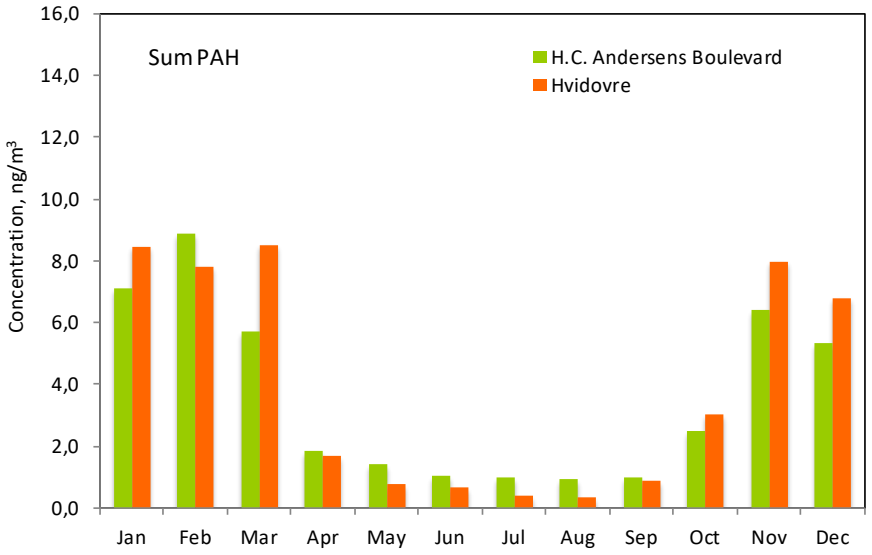


Figure 10.2. Monthly average concentrations of the sum of all analyzed PAHs at H. C. Andersens Boulevard and Hvidovre in 2018.

Table 10.2. Winter, summer and annual average concentrations of benzo[a]pyrene for 2013-2018.

	Hvidovre						HCAB					
	2013	**2014**	**2015**	**2016**	**2017**	**2018**	**2013**	**2014**	**2015**	**2016**	**2017**	**2018**
Winter	0.53	0.73	0.42	0.42	0.49	0.51	0.38	0.50	0.44	0.33	0.26	0.41
Summer	0.12	0.10	0.06	0.04	0.09	0.08	0.11	0.10	0.12	0.08	0.09	0.09
Annual	0.34	0.38	0.25	0.23	0.29	0.29	0.24	0.29	0.29	0.20	0.18	0.25

10.2 Trends

The annual averages of benzo[a]pyrene since 2008 at the street station on HCAB are shown in figure 10.3 together with five years of data from the sub-urban station in Hvidovre. A decrease in the annual averages of benzo[a]pyrene at HCAB is observed since 2008, and there is also a downward trend at Hvidovre since 2013. The variation from year to year is to a large extend due to the natural variation in the meteorology that have impact on both the need for residential heating and the dispersion of the emissions from the sources.

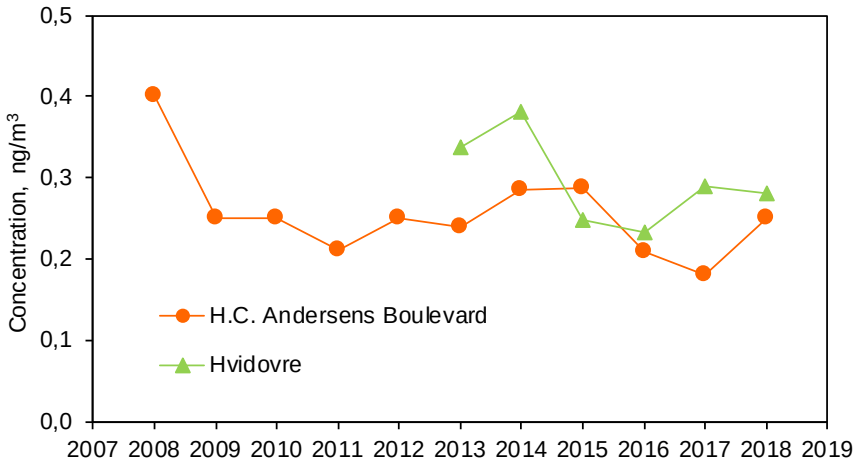


Figure 10.3. Annual average concentrations of benzo[a]pyrene at H. C. Andersens Boulevard and Hvidovre.

11. Organic carbon and elemental carbon

Ambient concentrations of particulate Organic Carbon (OC) and Elemental Carbon (EC) are monitored at four sites in Denmark with a time resolution of 24 hours. A kerbside station is located at H. C. Andersen's Boulevard/1103 in Copenhagen. EC in urban background is monitored in Copenhagen on H. C. Ørsted Instituttet/1259. OC and EC are measured at the semi-rural station Risø/2090 north of Roskilde, and at Hvidovre/2650 (suburban site, Hvidovre), which is considered to be a hotspot for residential wood burning. $PM_{2.5}$ is sampled on two filters in tandem, i.e. quartz-behind-quartz, to correct for positive artifacts from adsorption of volatile and semi-volatile organic compounds, which are not particulate material. The filters are analysed for OC and EC by a thermal/optical method according to the European EUSAAR2 temperature protocol (Cavalli et al., 2010) using a carbon analyser from Sunset Laboratories.

11.1 Annual statistics and trends

OC and EC have been measured in $PM_{2.5}$ since 2010. During this relatively short period, the annual averages of semi-rural OC has oscillated between 1.1 and 1.8 µg/m^3. Since biogenic sources are expected to account for the majority of the OC in $PM_{2.5}$ a constant trend biased by natural variation is expected. OC covariates at the kerbside station HCAB and the semi-rural site with an increment largely explained by the traffic source at HCAB (figure 11.1). The 2018 average EC in rural background (0.29 µg/m^3) has decreased by 36 % of its 2010 concentration. The kerbside station (1.1 µg/m^3), which is largely impacted by local traffic, has experienced a 53 % decrease in EC in the same period. In 2018, Copenhagen urban background (0.35 µg/m^3) and the suburban site in Hvidovre (0.40 µg/m^3) experienced EC concentrations 22 and 39 % higher than the semi-rural site. The ratio of EC to total carbon (TC) differs significantly between rural background (0.17) and the kerbside station in Copenhagen (0.34). While the EC/TC ratio has decreased most years from 2010 to 2017 at HCAB, EC/TC shows a nearly constant trend at Risø (figure 11.1).

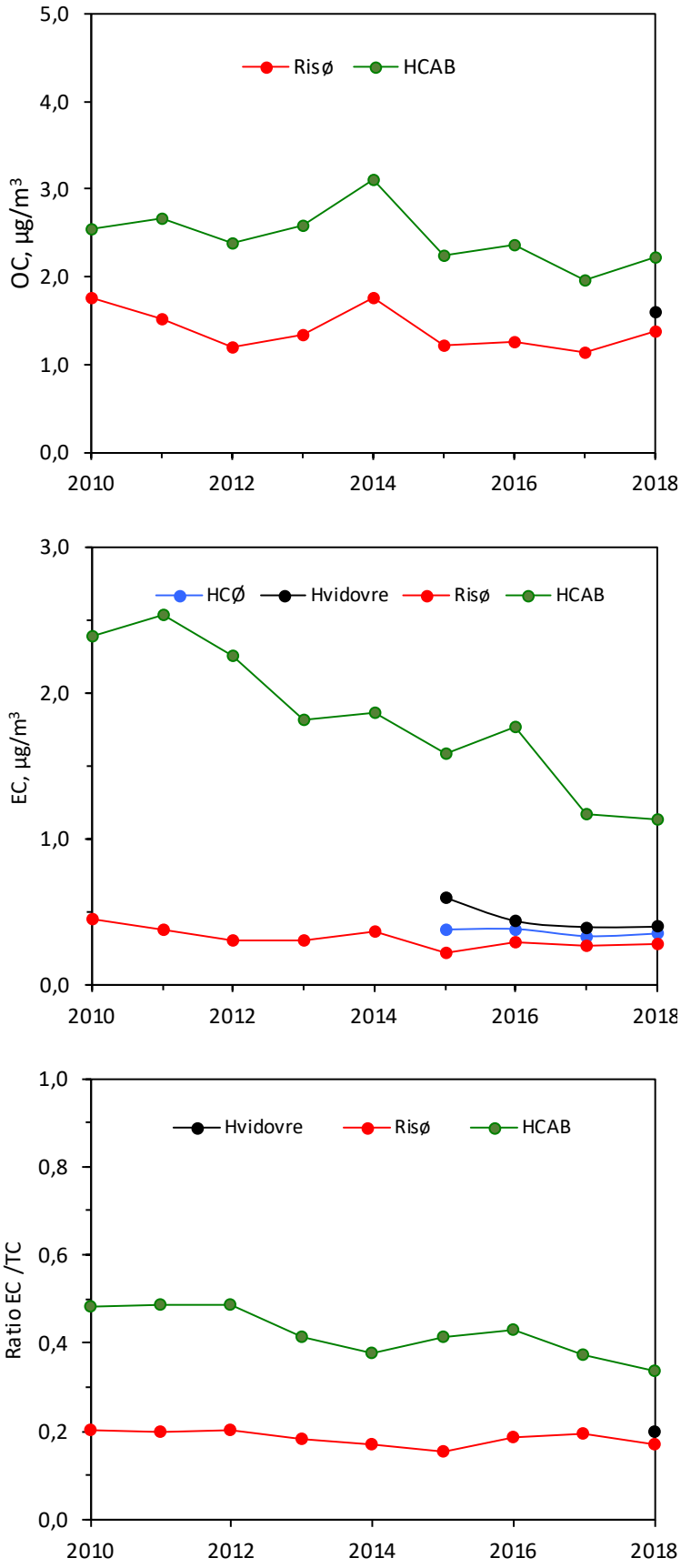
A clear seasonal pattern was observed for EC and OC at the rural and urban background with minimum summer concentrations and higher winter concentrations. EC and OC showed less seasonal variation at the kerbside station.

Table 11.1. Annual statistics for OC in 2018. The values are based on daily averages of Copenhagen kerbside and semi-rural background 30 km west of Copenhagen.

Concentration µg/m^3	Data capture	OC, average
Copenhagen/1103	99%	2.2
Risø/2090	95%	1.4

Table 11.2. Annual statistics for EC in 2018. The values are based on daily averages of Copenhagen kerbside and urban background, semi-rural background 30 km west of Copenhagen and at a suburban site southwest of Copenhagen.

Concentration µg/m^3	Data capture	EC, average
Copenhagen/1103	91%	1.1
Copenhagen/1259	92%	0.35
Risø/2090	93%	0.29
Hvidovre/2650	89%	0.35



Figur 11.1. OC, EC and the ratio of EC to total carbon (EC/TC) at kerbside (H.C. Andersen's Boulevard, HCAB), semi-rural background (RISØ), urban background (H.C. Ørsted Institut, HCØ) and at a suburban site (Hvidovre).

12. Chemical composition of $PM_{2.5}$

In addition to the measurements of elemental and organic compounds, also measurements of the main inorganic compounds in $PM_{2.5}$ (ammonium (NH_4^+), sodium (Na^+), potassium (K^+), calcium (Ca^{2+}), magnesium (Mg^{2+}), chloride (Cl^-), nitrate (NO_3^-), sulfate (SO_4^{2-})) have been conducted at the rural measurements station Risø. $PM_{2.5}$ is responsible for the majority of the health impacts from air pollution and determination of the chemical constituents in $PM_{2.5}$ are important. These measurements are carried out on the basis of the air quality directive from 2008 (EC, 2008). The method is chemical analysis of the daily $PM_{2.5}$ particle filters sampled using Low Volume Sampling.

12.1 Results

Examples of the daily variations of the concentrations are shown in figure 12.1 together with the variation of $PM_{2.5}$. The annual contributions to $PM_{2.5}$ of the different compounds are shown in figure 12.2. 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 Risø in 2018.

Table 12.1. Annual average contributions and relative distribution of the chemical composition of $PM_{2.5}$ at Risø in 2018. Organic matter (OM) has been estimated from the measured concentrations of OC by multiplication of OC with a factor of 2.1 for the OM at Risø that has undergone some chemical transformations in the atmosphere (Turpin and Lim, 2001). This is done in order to account for the contribution of hydrogen, oxygen, nitrogen etc. to the mass of the organic compounds.

Components	**µg/m^3**	**Distribution %**
$PM_{2.5}$	12	100
Na	0.18	1.5
Cl	0.23	2.0
Mg	0.02	0.2
NH_4	1.1	9,2
NO_3	2.5	21
SO_4	1.3	11
K	0.10	0.8
Ca	0.02	0.2
EC	0.29	2.5
OM	2.9	25
Unknown mass	3.1	26

13. Health effects of air pollution in Denmark

According to WHO, air pollution is now considered the world's largest single environmental health risk. Around 3.7 million people died prematurely in 2012 as a result of exposure to outdoor air pollution (WHO, 2014). This high impact of air pollution on human health is the reason for inclusion of model calculations of the health impacts and associated external costs of air pollution in Denmark in the Air Quality Monitoring Program under NOVANA.

The model calculations are carried out with the model system EVAv5.2. EVAv5.2 is an integrated part of a multi-scale model system that is capable of describing the contribution from intercontinental, regional, national and local sources on air pollution and hence also on the impact of air pollution on human health. For further details of the EVAv5.2-system, see chapter 2.3.

The health effects are associated with $PM_{2.5}$, NO_2, SO_2 and O_3. Of these, $PM_{2.5}$, NO_2 and O_3 are the most extensively used in studies of external costs, as their effects are dominant compared to the other species. Atmospheric particles are considered responsible for mortality and morbidity, primarily via cardiovascular and respiratory diseases. A review from Hoek et al. (2013) includes the most comprehensive analysis of cardio-respiratory impacts in long-term studies and concludes that the long-term relative risk for total mortality is 6.2 % per 10 µg/m^3 increase in $PM_{2.5}$, which is the relative risk applied in EVA.

The model calculations of the health effects and external costs related to air pollution in Denmark have been updated on several aspects. This has resulted in an adjustment of the numbers for the health impacts and the external costs. The results presented in this report are therefore not directly comparable to the results presented in the annual report for 2017. Chapter 2 describes the changes in the model calculations.

13.1 Status and trend for health effects

Table 13.1 presents the number of cases for the different health outcomes due to the total air pollution calculated using the EVA model system as a mean over the three years 2016-2018. The table presents the impact from short-term exposure to $PM_{2.5}$, NO_2, SO_2 and O_3 and long-term exposure to $PM_{2.5}$ and NO_2 as well as morbidity allocated to the different species.

The number of cases of premature deaths due to long-term exposure is calculated from the Years of Life Lost (YOLL) using an average number of life years lost (10.6 years, see Brandt et al., 2013a). The total annual number of premature deaths due to the total air pollution levels in 2016-2018 is estimated to around 4,200 cases in Denmark. Health impacts due to exposure to NO_2 are recently included in the EVA system and contributes with around 360 cases of premature deaths.

The main driver for the health impacts is $PM_{2.5}$, which in these calculations includes the total primary emissions of $PM_{2.5}$, including mineral dust, fresh and aged black carbon (BC), organic matter (OM), sea salt from sea spray, as well as the secondary inorganic aerosols (SIA) and the secondary organic aerosols (SOA). $PM_{2.5}$ accounts for about 90 % of all premature deaths, NO_2 for

about 8,5 %, O_3 for about 1,6 % and SO_2 for around 0.1 % (as a mean over the three years 2016-2018).

The risk of premature death resulting from exposure to $PM_{2.5}$, NO_2, O_3 and SO_2 is rather homogeneously distributed over Denmark, however with a gradient from south to north and higher risks in the major cities. The explanation is that the majority of premature deaths is related to $PM_{2.5}$, and the geographical variation in the concentration of $PM_{2.5}$ is fairly small. This is due to the large contribution to $PM_{2.5}$ that originates from long-range transport of air pollution mainly from the northern parts of the European continent.

Model calculations with the EVAv5.2 system have been carried out in order to calculate the development of the health impacts for the period 1990-2018. Figures 13.1 and 13.2 present the total number of premature deaths due to $PM_{2.5}$, NO_2, O_3 and SO_2 in Denmark as annual averages due to the total air pollution. The total number of premature deaths has decreased from around 7,200 cases/year in 1990 to around 4,500 cases/year in 2018 – a reduction of 38 % over this period (note that these numbers of premature deaths are means over a single year). The variations from year to year are due to natural variations in the meteorological conditions and the general development in emissions in Denmark and Europe.

Recent results for Europe (Brandt et al., 2013a; 2013b) show that outdoor air pollution caused about 570,000 premature deaths in 2011. For 2016-2018, the number of premature deaths in Europe is calculated to app. 400,000, however, this is for the countries in the European Union only.

Model calculations with the DEHM and UBM models based on an emission reduction scenario have been conducted in order to estimate the contribution from emissions in foreign countries to air pollution across Denmark (in this case all natural and anthropogenic emissions in the Northern Hemisphere) and the contribution from anthropogenic emissions in Denmark to the number of premature deaths, as calculated with the EVA model system, see table 13.2. The contribution from foreign countries to premature deaths in Denmark is estimated to about 3,000 (71 % of the total number of cases in Denmark), while the contribution from Danish emissions is about 1,220 premature deaths in Denmark (29 %). The contribution from Danish emissions to the number of premature deaths in Europe (excl. Denmark) is estimated to about 1,810 cases/year. The "import" of air pollution related health impacts from foreign sources is therefore about 40 % larger than the "export" of health impacts to foreign countries from Danish sources. It is also seen that Danish emissions cause about 33 % more premature deaths in foreign countries (~1,810) than they do in Denmark (~1,200).

Table 13.1 The number of cases for the different health outcomes in the EVAv5.2 model system due to the total air pollution concentrations as a mean over the three years 2016-2018 for the whole of Denmark.

	Number of cases				
Health outcome	**SO_2**	**O_3**	**NO_2**	**$PM_{2.5}$**	**Total**
Premature deaths (short-term exposure)	6	66	359	771	1,200
Premature deaths (long-term exposure)			1	3,020	3,020
Premature deaths (total)	6	66	360	3,790	4,220
Respiratory Hospital Admissions		91		1,470	3,240
Cerebrovascular Hospital Admissions		297		1,030	972
Cough Children				356	356
Chronic Bronchitis (adults)				3,000	3,000
Chronic Bronchitis (children)				19,700	19,700
Work loss days				1,250	1,250
Restricted Activity Days				3,700,000	3,700,000
Minor restricted Activity Days		777		0	777
Lung Cancer				56	56
Infant mortality				1	1

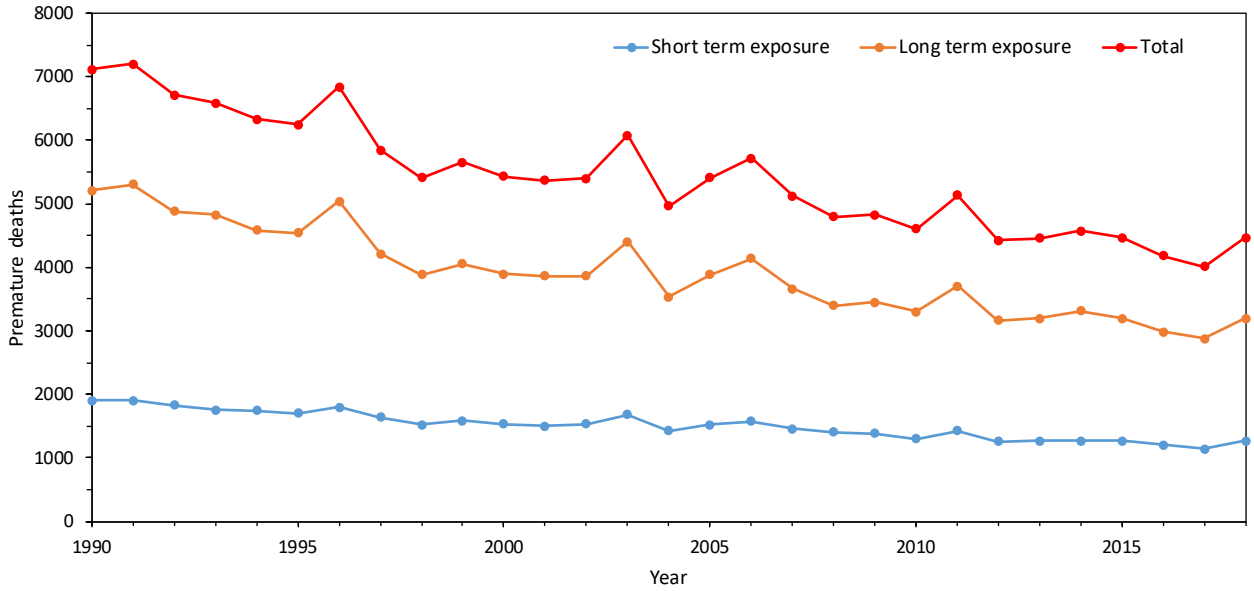


Figure 13.1. Total annual number of premature deaths due to the total air pollution of $PM_{2.5}$, O_3, NO_2 and SO_2 in Denmark from short-term and long-term exposure as well as the total number of premature deaths. Calculations are carried out using the EVAv5.2 model system.

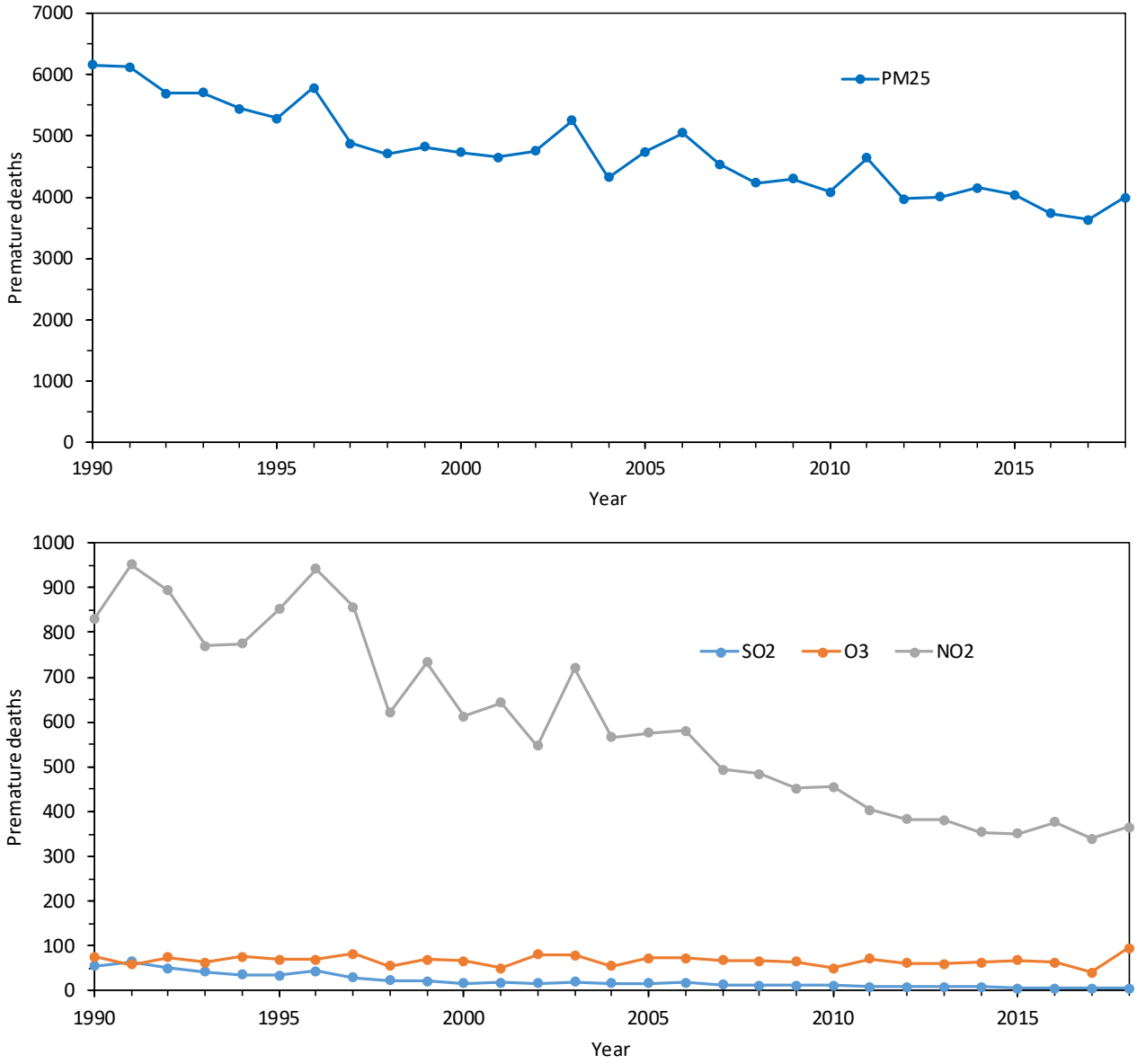


Figure 13.2. Annual number of premature deaths in Denmark due to exposure to the air pollutants $PM_{2.5}$ (top panel) and NO_2, O_3 and SO_2 (bottom panel) Calculations are carried out using the EVAv5.2 model system.

Table 13.2. Contribution from emissions in foreign countries to Denmark and the contribution from emissions in Denmark to the number of premature deaths, calculated with the EVAv5.2 model system as a mean over the three years 2016-2018.

Contributions	Number of premature deaths	% of total
Total air pollution in Denmark	4,220	100
Foreign contribution to Denmark	3,000	71
Denmark's contribution to Denmark	1.220	29
Denmark's contribution to Europe incl. Denmark	3,030	100
Denmark's contribution to Europe excl. Denmark	1,810	60

13.2 Status and trend for external costs of health impacts

An external cost occurs when the production or consumption of a good or service imposes a cost upon a third party, as e.g. activities leading to increased air pollution concentrations, which result in impacts on health, nature or climate. In the EVAv5.2 system, the external costs related to health impacts from air pollution are calculated.

The total health related external costs for Denmark have been calculated to about 79 billion DKK as an average over the three years 2016-2018 using the economic valuation of the individual health outcomes in Andersen et al. (2019) in 2016 prices. The trend in the total external costs is similar to the development of the total number of premature deaths and is therefore not shown here. The total health related external costs as an average over the years 1988-1990 is about 131 billion DKK and has therefore decreased by about 38 % since then.

The contribution from emissions from foreign countries to air pollution and associated health effects and socio-economic costs in Denmark, and the similar contribution from emissions in Denmark, calculated by the EVA model system, is given in table 13.3. The contribution from foreign countries to Denmark is estimated to about 55 billion DKK (70 % of the total health related external costs in Denmark), while the contribution from Danish emissions contributes with about 24 billion DKK in Denmark (30 %). The contribution from Danish emissions to the total health related external costs in Europe excluding Denmark is about 30 billion DKK.

Table 13.3. Contribution from emissions from foreign countries to Denmark and the contribution from emissions in Denmark to the total health related external costs, calculated with the EVAv5.2 model system as a mean over the three years 2016-2018.

Contributions	Billion DKK	% of total
Total air pollution in Denmark	79	100
Foreign contribution to Denmark	55	70
Denmark's contribution to Denmark	24	30
Denmark's contribution to Europe incl. Denmark	58	100
Denmark's contribution to Europe excl. Denmark	30	58

13.3 Adjustments

The model calculations of the health impacts and external costs for air pollution have been updated in connection with this reporting and it is therefore not possible to make a direct comparison between the numbers presented in this report for 2018 and the numbers presented in the latest report covering 2017.

The average number of premature deaths was in the reporting for 2017 calculated to be about 3,200 for the average of 2015-2017. The same number in the present report is calculated to be about 4,200 as an average for 2016-2018. This change is mainly due to two factors:

Inclusion of an exposure-response function for the direct impact of NO_2 on mortality. This accounts for 360 extra premature deaths in 2016-2018 compared to 2015-2017.

Improvements in the model calculations for $PM_{2.5}$ (see Chapter 2 for further details) have increased the level of the model calculated $PM_{2.5}$. Moreover, the present calculations have also been calibrated upwards in order to obtain good agreement with the measurement results (Appendix 3). In the reporting for 2017 the concentrations of $PM_{2.5}$ were about 26 % lower compared to measurement results. The underestimation of the health impacts from $PM_{2.5}$ in the reporting for 2017 corresponds to about 800 premature deaths. The better model performance of the results from this report compared to the reporting for 2017 is therefore the other main reason for the higher number reported for the health impacts this year.

The special weather conditions in 2018 lead to a significant increase in the measured $PM_{2.5}$ of around 25 % in 2018 compared to 2017 (Chapter 7). However, the numbers for the health impacts are given as average for three years and hence, the year-to-year variations due to changing meteorological conditions have been smoothed out. As a consequence the special weather conditions in 2018 have only given rise to small changes in the health impacts from 2015-2017 to 2016-2018.

There has also been a major update in the procedure for assessment of external costs. In the report for 2017, the total external costs were calculated to 25 million DKK while the same number has been calculated to about 79 million DKK in this reporting for 2018. This quite large adjustment is mainly a consequence of changes in the valuation of a statistic life. The new model calculations have been based on the Danish Economic Councils updated value of a statistic life (about 32 million DKK) (DØRS 2016; Ministry of Finance, 2017) and this value is about a factor of two higher than the value used in model calculations for 2017. This update has led to nearly a doubling of the external costs. Moreover, the adjustment of the model calculations of $PM_{2.5}$ that resulted in a higher number of premature deaths is also one of the major reasons for the higher numbers presented in the report.

13.4 Uncertainties

There are considerable uncertainties associated with the calculation of health impacts and external costs related to air pollution. Lelieveld and coworkers (2019) have estimated that their calculation of health impacts from air pollution is associated with an uncertainty of ± 50 %. DCE estimates that the uncertainty associated with the calculations presented in this report is of similar magnitude.

A significant part of the uncertainty relates to the exposure-response relationships and especially to the exposure-response relationships implemented for NO_2. For the chronic mortality of NO_2, WHO recommends the application of a threshold of 20 $\mu g/m^3$, so that it is only concentrations above this threshold that contribute to the impact of NO_2 on chronic mortality. This threshold is therefore implemented in DCE's calculations of the health impact of NO_2. However, there is considerable uncertainty connected with this threshold and some research (Héroux et al., 2015) indicates that the threshold is too high or that it should be removed all together. This will have a significant influence on the results from the calculations and a decrease in the threshold will lead to a significant higher number of premature deaths attributable to NO_2.

There are also large uncertainties related to the exposure-response relationships for O_3, where recent research indicates that O_3 at lower concentrations also has large impacts on human health. Until now, it has generally been accepted that the health impact of O_3 originates from exposure to O_3 at high concentrations and this is the background for use of exposure-response relationships that are based on the parameter SOMO35 that sums up all the O_3 concentrations above 35 ppb (= 70 $\mu g/m^3$). New research indicates that O_3 concentrations as low as 10 ppb (= 20 $\mu g/m^3$) can have significant impact indicating the use of a lower threshold (SOMO10). This will lead to a higher impact from O_3 on health. At present, it is the recommendation from Danish experts on health impacts from air pollution that DCE continues with the use of SOMO35 (Ellermann et al., 2019).

There is today very solid documentation for the health impacts related to $PM_{2.5}$. However, there is still a lack of scientific knowledge on which particular chemical constituents of $PM_{2.5}$ are responsible for the health impact. The recommendation from WHO is still to use the same exposure-response function for the different chemical constituents of $PM_{2.5}$ and the results from the EVA-model system is based on this assumption. Changes in this assumption will potentially lead to changes in the magnitude, sources and spatial distribution of the health impacts and external costs from air pollution.

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Appendix 1

Replacement of the station at H. C. Andersens Boulevard

On 3 October 2016, the station at H. C. Andersen's Boulevard was closed and replaced with a new station (2.3). The majority of the measurements were initiated on 19 October 2016. The new station was located 2.7 m further away from the inner traffic lane in order to compensate for the road change in 2010 (figure A.1 and A.2). Moreover, the station was moved about 2 m further away from a tree close to the station. The EU directive (EC, 2008) specifies measurements to be carried out several meters from trees in order to avoid influence from the trees on the measurements.

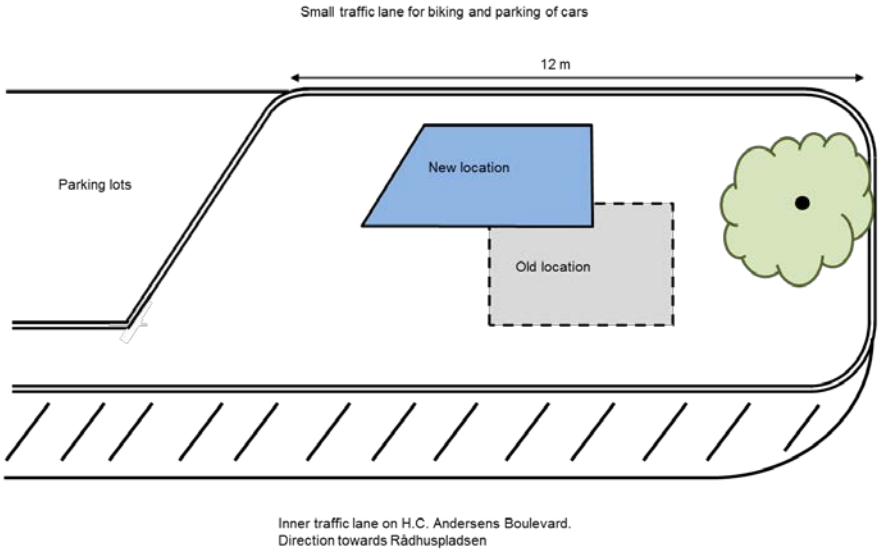

Figure A.1. Sketch of the old and new location of the measurement station at H. C. Andersens Boulevard.

Figure A.2. Aerial photo of the location of the measurement station (red circle) at H. C. Andersens Boulevard.

Appendix 2

Pollutants measured in the 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 NO by 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 NO_x and VOCs. The VOCs 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-fueled 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 VOCs are present in the air. Several of these are emitted by incomplete combustion in e.g. engines and wood burning stoves. Several of the VOCs are carcinogenic. A "target value" is implemented through an EU Council Directive in 2004 for benzo[a]-pyrene as indicator for PAH (poly aromatic hydrocarbones). PAH in PM_{10} 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 analyzed with the method.

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 is 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 monitoring program:

- The Beta method: The particles are collected on filters for 24 hours intervals. The mass on the filters is automatic determined by measurements in the instrument of 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 PM_{10} or $PM_{2.5}$ 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. HMs may also be emitted from traffic due to wear on engines, tires and brake pads. Several HMs are toxic even in low concentrations and a few also carcinogenic. A limit value is implemented for lead. Target 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 HMs.

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 in 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 1980s 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.

Appendix 3

Details on the calibration of OSPM and validation of model results

In section *2.2.1 Model calibration and validation* there is a description of the calibration procedure used for OSPM. No calibrations are carried out for NO_x/NO_2 in DEHM and UBM.

For $PM_{2.5}/PM_{10}$ a small calibration of all final model results was necessary since a comparison with measurements showed a noticeable underestimation. The observed underestimation was similar over all types of stations (rural, urban, kerbside) and therefore seems to affect all models in our modelling chain (DEHM, UBM, OSPM). The reason for the underestimation seems to be the lack or underestimation of some particle components in the model e.g.: Secondary Organic Aerosol (SOA), water content in PM, non-exhaust emissions. The calibration resulted in an increase in PM concentrations of about 2.3 µg/m³. In the figures below the calibrated $PM_{2.5}/PM_{10}$ model results are shown.

In the following, we present a number of scatter plots to characterize the correlation between measurements and model calculations. All data shown are from 2016 to 2018 for all available stations: street, urban background and rural. A period of 3 years was chosen in order to have the model calibration and performance evaluated for a more significant number of observations and to smoothen out some effects of variations in meteorology that might distort the picture of only a single year.

The different measuring stations and their corresponding name and identification number are shown in the figure legends.

In Figure A.3 the correlation between modelled and observed annual levels of NO_2 is shown for all stations for years 2016 to 2018. There are 36 observations and the average observed concentration is 16.7 µg/m³, and the modelled is 19.2 µg/m³. The Pearson and Spearman correlations (R p/s) are very high (0.96 and 0.92) and the Normalized Mean Bias (NMB %) is acceptable low (15.1 %).

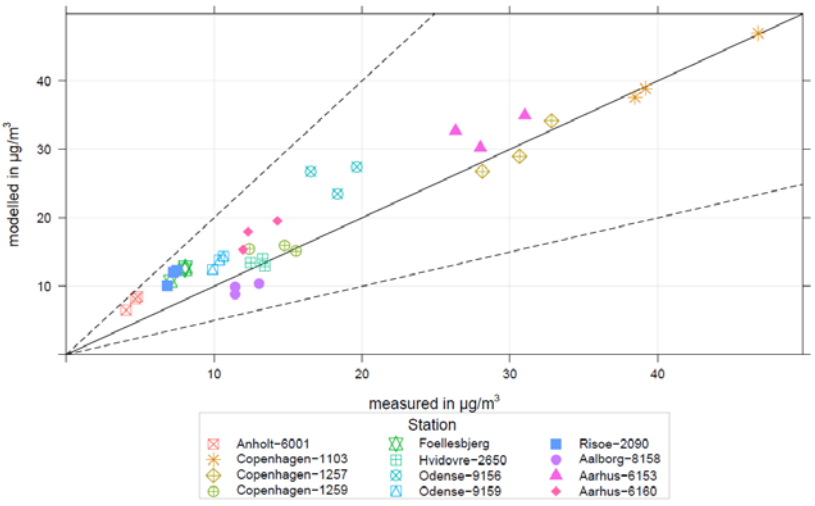

Figure A.3. Correlation between modelled and observed annual levels of NO_2 for all stations for 2016-2018. The solid line represent the 1:1 line and the dashed lines 1:2 lines.

In Figure A.4 the correlation between modelled and observed annual levels of $PM_{2.5}$ is shown for all stations for 2016-2018. There are 24 observations and the average observed concentration is 11.7 µg/m³, and the modelled is 11.3 µg/m³. The Pearson and Spearman correlations (R p/s) are very high (0.88 and 0.88) and the Normalized Mean Bias (NMB %) is low (-3.8 %).

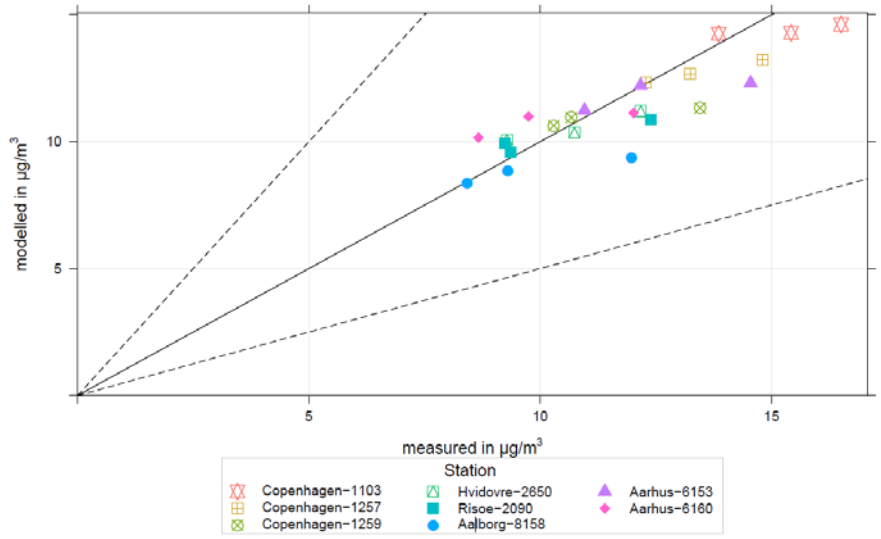

Figure A.4. Correlation between modelled and observed annual levels of $PM_{2.5}$ for all stations for 2016-2018. The solid line represent the 1:1 line and the dashed lines 1:2 lines.

In Figure A.5 the correlation between modelled and observed annual levels of PM_{10} is shown for all stations for 2016-2018. There are 18 observations and the average observed concentration is 21.3 µg/m³, and the modelled is 19.8 µg/m³. The Pearson and Spearman correlations (R p/s) are very high (0.92 and 0.92) and the Normalized Mean Bias (NMB %) is low (-7 %).

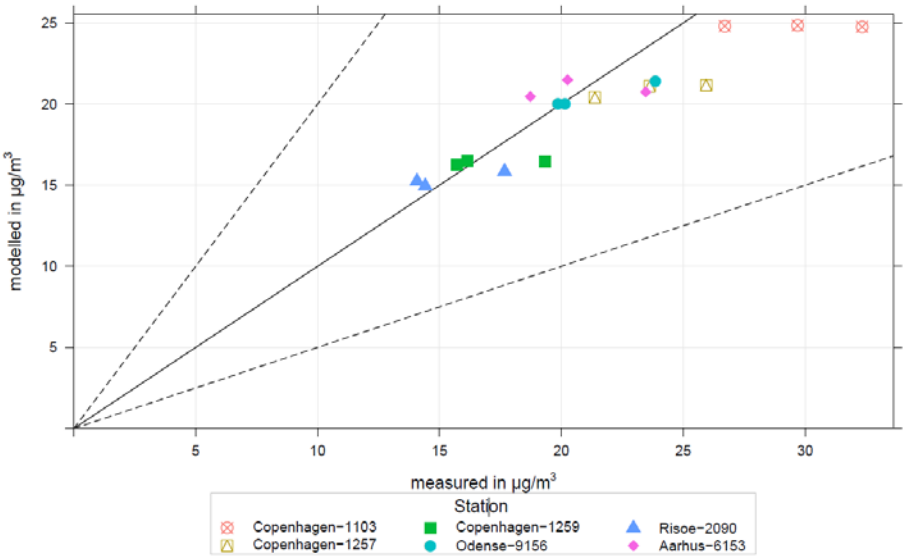

Figure A.5. Correlation between modelled and observed annual levels of PM_{10} for all stations for 2016-2018. The solid line represent the 1:1 line and the dashed lines 1:2 lines.

THE DANISH AIR QUALITY MONITORING PROGRAMME

Annual Summary for 2018

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 2018 the air quality was measured in four Danish cities and at two background sites. In addition, model calculations of air quality and the impact of air pollution on human health and related external costs were carried out. For 2018, no exceedances of the NO_2 EU limit values were observed. Model calculations were carried out for 98 streets in Copenhagen and 31 in Aalborg. Only one exceedance of the limit value for the annual average of NO_2 was modelled for a busy street in Copenhagen. Annual averages of PM_{10} and $PM_{2.5}$ were below limit values at all stations and the average exposure indicator ($PM_{2.5}$ in urban background) has decreased with about 30 % since 2010. The concentrations for most pollutants have been decreasing during the last decades.

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