# THE DANISH AIR QUALITY MONITORING PROGRAMME

Annual Summary for 2017

Scientific Report from DCE - Danish Centre for Environment and Energy

No. 281

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AARHUS UNIVERSITY DCE - DANISH CENTRE FOR ENVIRONMENT AND ENERGY [Blank page]

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

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# Data sheet

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# Summary and Conclusion

This report presents the result from the Danish Air Quality Monitoring Programme in 2017. The monitoring programme is carried out by the DCE - Danish Centre for Environment and Energy (DCE) at Aarhus University. The core part of this programme consists of continuous measurements at eleven monitoring stations; nine stations situated in the four largest cities, two stations located in background areas and a station in a suburban area. These measurements are supplemented with model calculations using DCE's air quality models.

The aim of the program is to monitor air pollutants relevant to human health in accordance with the EU air quality directives. The programme includes measurements of sulphur dioxide (SO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>/NO<sub>2</sub>), particulate mass less than 10 (PM<sub>10</sub>) and 2.5 micrometers (PM<sub>2.5</sub>), particle number, benzene (C<sub>6</sub>H<sub>6</sub>), toluene (C<sub>7</sub>H<sub>8</sub>), carbon monoxide (CO), ozone (O<sub>3</sub>), 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<sub>3</sub>. The measurements and model calculations are used to evaluate the Danish air quality in relation to limit values as well as to follow trends. Further, the obtained data are used for determination of sources of the air pollutants, basis for evaluation of the impact of regulations of emissions and as basis for various research projects related to air quality.

The permitted number of exceedances in a year of the diurnal limit value of  $50 \ \mu g/m^3$  for PM<sub>10</sub> was not exceeded at any station in the measuring network. Likewise, there were no exceedances of the annual limit values for PM<sub>10</sub> (40  $\ \mu g/m^3$ ) and PM<sub>2.5</sub> (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<sub>2.5</sub> 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 and data for 2017 has to be regarded as preliminary. Therefore, the particle number represents the particle range from 41 to 559 nm. The particles in ambient air was about 5,000 particles per cm<sup>3</sup> as an annual average at the street station H.C. Andersens Boulevard. This is roughly a factor of 2 higher than in suburban and 2.5 higher than in urban and rural background, respectively. A significant reduction of more than 40% in particle numbers has been observed since 2002. This reduction has mainly been attained by reduction of traffic emissions (cleaner fuel, particle filters etc.).

The annual limit value for NO<sub>2</sub> ( $40 \ \mu g/m^3$ ) was not exceeded at any of the measurements stations in Denmark. Model calculations at selected streets in Copenhagen and Aalborg in 2017 indicate no exceedances of the limit values whereas model calculations of 98 streets in Copenhagen in 2016 indicated 6 exceedances.

The annual average  $O_3$  concentrations in 2017 were at the same level as in the previous years but the maximum 8-hours running mean concentration was smaller in 2017 compared to 2016. 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  $180 \ \mu g/m^3$  was not exceeded at any of the measurement stations in 2017. The target value for the maximum daily 8-hours mean  $O_3$  concentration of  $120 \ \mu g/m^3$  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 data for this has not been decided yet.

Measurements of VOCs at the urban background in Copenhagen showed concentration levels between 0.03  $\mu$ g/m<sup>3</sup> and 0.827  $\mu$ g/m<sup>3</sup> for the selected 17 different compounds. VOCs can act as O<sub>3</sub> precursors, and the aim of these measurements is to improve the general understanding of the O<sub>3</sub> formation at a European level. The formation of O<sub>3</sub> in Denmark is in general small due to moderate solar radiation. O<sub>3</sub> pollution in Denmark is to a 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 decreased 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 0.18 ng/m<sup>3</sup> and 0.29 ng/m<sup>3</sup> at H.C. Andersens Boulevard and Hvidovre, respectively. The target value for benzo[a]pyrene (1 ng/m<sup>3</sup>) was not exceeded in 2017.

Measurements of the chemical content in  $PM_{2.5}$  were due to minor revisions of the program only carried out at the rural background station at Risø. The results are almost unchanged compared to 2016.

Model calculations show that air pollution causes about 3,200 premature deaths in Denmark as average for 2014-2017 and a large number of other negative health effects. There is about 400 fewer premature deaths in 2017 compared to 2016. This decrease is due to a general reduction in emissions and extraordinary low peak O3 concentrations in 2017 due to the weather conditions during summer 2017. About 770 (24 %) of the premature deaths are due to Danish emission sources while the remaining premature deaths are caused mostly by European sources outside Denmark. The total health related external costs for Denmark have been calculated to 3.3 billion EUR (~25 billion DKK) as an average over the three years 2014-2017. The negative health effects and external costs have declined with about 40% since 1988-1990. The calculation of the health impacts and external costs are associated with considerably uncertainties among others because the model underestimates the concentrations of airborne particles compared to the measurements. Moreover, newer results indicate that there is an independent health impact due to nitrogen oxides and this impact has not been included in the model yet.

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

# Danish summary - Dansk resumé

Rapporten præsenterer resultater for 2017 fra Overvågningsprogrammet for luftkvalitet i danske byer. Programmet, som udføres af DCE - Nationalt Center for Miljø og Energi (DCE) ved Aarhus Universitet, er baseret på målinger ved ni målestationer placeret i de fire største danske byer samt ved to baggrundsmålestationer udenfor byerne og en station i et forstadsområde. Disse måleresultater suppleres med resultater fra modelberegninger udført med DCE's luftkvalitetsmodeller.

Formålet med programmet er at overvåge luftforurening af betydning for sundhed. Målingerne udføres i overensstemmelse med EU's luftkvalitetsdirektiver. I henhold til disse og øvrige danske behov måles koncentrationer af svovldioxid (SO<sub>2</sub>), nitrogenoxider (NO<sub>x</sub>/NO<sub>2</sub>), massen af partikler mindre end 10 (PM<sub>10</sub>) og 2,5 mikrometer (PM<sub>2,5</sub>), partikelantal, benzen (C<sub>6</sub>H<sub>6</sub>), toluen (C<sub>7</sub>H<sub>8</sub>), carbonmonoxid (CO), ozon (O<sub>3</sub>), 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<sub>3</sub>. 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 kilderne til luftforureningen i Danmark, vurdering af effekt af reduktionstiltag og som grundlag for en række videnskabelige undersøgelser, fx vurdering af små partiklers effekt på sundheden.

Der er fastsat grænse- og målværdier for flere af de målte stoffer. Grænseværdierne skal være overholdt fra 2005, 2010 eller 2015 alt efter hvilke stoffer, det drejer sig om. En detaljeret beskrivelse af gældende mål- og grænseværdier og deres gennemførelse 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 stilles krav om målinger af de fine partikler (PM<sub>2,5</sub>), og at der er indført en grænseværdi for PM<sub>2,5</sub>, som skulle overholdes fra 2015.

I 2017 blev grænseværdien for NO<sub>2</sub> som årsmiddelværdi ikke overskredet. Koncentrationerne af NO<sub>2</sub> målt på gadestationerne i 2017 var faldet i forhold til koncentrationerne målt i 2016. Modelberegninger viser et fald i koncentrationerne i NO<sub>2</sub> som følge af fald i udledningerne fra navnlig trafikken. Endvidere indikerer modelberegningerne, at der ikke var overskridelse af grænseværdien i 2017, mens der i 2016 var overskridelser på 6 ud af 98 beregnede gadestrækninger i København, men ikke på udvalgte gadestrækninger i Aalborg.

 $PM_{10}$  overholdt grænseværdien på 40 µg/m<sup>3</sup> 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<sup>3</sup> må ikke overskrides mere end 35 gange årligt) blev overskredet.

 $PM_{2,5}$  overholdt grænseværdien på 25  $\mu g/m^3$  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) 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 2017 foreløbige. Derfor angives antallet af partikler for 2017 i intervallet fra 41 – 550 nm. Antallet af partikler var omkring 5.000 partikler per cm<sup>3</sup> på gademålestationen H.C. Andersens Boulevard, hvilket er en faktor 2 højere end ved forstadsstationen Hvidovre samt en faktor 2,5 højere end både by- og land-baggrundsstationen hhv. HCØ og Risø. Siden 2002 har der været et fald på ca. 40% i antal partikle med diameter mellem 41 – 550 nm. Faldet er blandt andet sket som følge af indførelse af svovlfrie brændstoffer og krav om partikelfilter på alle nye dieselkøretøjer.

Ozonkoncentrationerne i 2017 var på niveau med tidligere år. Der er ikke fastsat egentlige grænseværdier for O<sub>3</sub>, men kun "målværdier" og "langsigtede mål" (hensigtsværdier). Der var i 2017 ingen overskridelser af målværdierne for beskyttelse af sundhed, mens de langsigtede mål (120  $\mu$ g/m<sup>3</sup>) blev overskredet på tre bybaggrundsstationer, København (ved H. C. Ørsted instituttet), Aarhus (lokaliseret ved den botaniske have) og Odense (på taget af Rådhuset). Målværdien for ozon trådte i kraft i 2010, mens de langsigtede mål endnu ikke er trådt i kraft og der er ikke taget beslutning om hvornår dette sker. Tærsklen for information af befolkningen om høje ozonniveauer (timemiddel 180  $\mu$ g/m<sup>3</sup>) blev ikke overskredet i 2017.

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

Målinger af partikelbundet PAH blev fortaget på H.C. Andersens Boulevard i København. Middelværdien for benz[a]pyren var 0,18 ng/m<sup>3</sup> og 0,29 ng/m<sup>3</sup> på henholdsvis H.C. Andersens Boulevard og ved målestationen i Hvidovre. Målværdien på 1 ng/m<sup>3</sup> var således ikke overskredet i 2017

Målinger af 17 udvalgte VOC'er i bybaggrund i København viser koncentrationsniveauer, som spænder fra 0,03  $\mu$ g/m<sup>3</sup> til 0,87  $\mu$ g/m<sup>3</sup> i 2017. Disse VOC'er bidrager til den kemiske dannelse af O<sub>3</sub> på europæisk plan, og målingerne skal først og fremmest understøtte den generelle forståelse af ozondannelsen i Europa. I Danmark skyldes størstedelen af O<sub>3</sub> langtransport af luftforurening fra centrale og sydlige dele af Europa.

Grundet revision i måleprogrammet blev målinger af det kemiske indhold i PM<sub>2,5</sub> i 2017 kun gennemført ved landbaggrundsmålestationen på Risø. Målingerne i 2017 er stort set uændrede.

Modelberegningerne af helbredseffekterne viser, at luftforureningen som gennemsnit for 2015-2017 er skyld i omkring 3.200 for tidlige dødsfald og en lang række andre negative helbredseffekter. Der er dermed omkring 400 færre for tidlige dødsfald sammenlignet med perioden 2014-2016. Årsagerne til dette er generelt faldende udledninger kombineret med lave ozonkoncentrationer i 2017, som følge af de meteorologiske forhold. Omkring 770 (24 %) 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 25 milliarder kr. (omkring 3,3 milliarder euro). De negative helbredseffekter og de eksterne omkostninger er faldet med omkring 40% siden 1988-1990. Beregningerne af helbredseffekterne er behæftet med betydelige usikkerheder bl.a. fordi modelberegningerne underestimerer partikelkoncentrationerne sammenlignet med målingerne og fordi nyere resultater indikerer, at der kan være en selvstændig sundhedseffekt af NO<sub>2</sub>, hvilket endnu ikke er implementeret i modelberegningerne.

# 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 Fødeministeriet, 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 these revisions were implemented from January 2017 except for the modelling part of the programme that has been extended, so that they now also include model calculations of the health impacts and the external costs of air pollution. Results from these calculations are therefore presented for the second time in this report.

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 programme and gradually installed at the PM-stations in the network to replace some of the older SM200 instruments that needed to be renewed. See introduction to Chapter 7 for an overview.
- A 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<sub>2.5</sub> by LVS, elementary (EC) and organic carbon (OC), particle number and nitrogen oxides (NO and NO<sub>2</sub>).
- The urban background measurement station in Aarhus was in January 2015 moved to another position (Chapter 2.1).

- The street station in Aalborg had to be temporarily closed down from September 2014 and onwards due to nearby construction work (Chapter 2.1).
- At the street station in Albanigade in Odense there was a large decrease in daily traffic intensity 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. The station is situated 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.

In the following chapters, the results from measurements and model calculations for 2017 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 still 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: <a href="http://dce.au.dk/en/authorities/air/">http://dce.au.dk/en/authorities/air/</a> (in Danish <a href="http://dce.au.dk/myn-digheder/luft/">http://dce.au.dk/myn-digheder/luft/</a>).



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

Location	Station type	Station number
Copenhagen		
H.C. Andersens Boulevard (HCAB)	Street	1103
Jagtvej	Street	1257
H.C. Ørsted Institute (HCØ)	Urban background	1259
Hvidovre, 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, Limfjordsbroen	Street	8151
Østerbro	Urban background 8150	
Rural		
Lille Valby/Risø*	Rural background	20901
Keldsnor	Rural background	9055

**Table 2.1.** Main stations used for monitoring of air quality in relation to health in 2017

\* 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 2014-2016 there were four major changes regarding the stations:

- The measurement station on Vesterbro at Limfjordsbroen in Aalborg was closed down temporarily 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 not been reestablished yet.
- 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 2.3). 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 for 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 2.2. The new street station at Grønløkkevej, Odense (left). The map shows the position of the street station (blue dot).



Figure 2.3. 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 2017:

- Nitrogen oxides (NO, NO<sub>2</sub> and NO<sub>x</sub> (= NO + NO<sub>2</sub>)) were measured at all stations.
- Particle mass (PM<sub>10</sub> and/or PM<sub>2.5</sub>) as 24-hour averages, were measured throughout the year at all stations except at Aalborg /street (PM<sub>2.5</sub>) were no data were measured in 2017 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 2017, 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<sub>10</sub> were measured at Copenhagen/street (HCAB), Copenhagen/urban background, Aarhus/street and the rural site Risø.
- Additionally, PM<sub>10</sub> and PM<sub>2.5</sub> were measured at both Copenhagen/street (HCAB) and Risø by means of TEOM (Tapered-Element Oscillating Microbalance) that measures on a half hourly basis making it possible to resolve the diurnal variation. Part of these measurements was carried out in a research project funded separately by the Danish EPA.

- Particle number was measured at Copenhagen/street (HCAB), Copenhagen/urban background and Risø in cooperation with a particle research project funded separately by the Danish EPA. Additionally, measurements were started at a suburban site in Hvidovre in autumn 2015.
- Ozone (O<sub>3</sub>) was measured at all urban background and rural stations, and at the street stations Copenhagen/street (HCAB).
- Carbon monoxide (CO) was measured at all street stations except Jagtvej as well as at the urban background station, Copenhagen/urban background and the rural site Risø.
- Benzene and toluene were measured at Copenhagen/street (HCAB) and Copenhagen/urban background using passive sampling on a weekly basis.
- PAHs were measured at Copenhagen/street (HCAB) and at the suburban site in Hvidovre.
- Sulphur dioxide (SO<sub>2</sub>) was measured at Copenhagen/street (HCAB). The main purpose was to monitor episodic high concentrations.
- Elemental carbon (EC) and organic carbon (OC) were measured at Copenhagen/street (HCAB) and the rural site Risø in PM<sub>2.5</sub>. EC was measured at the suburban station in Hvidovre. In addition, the main inorganing ions in PM<sub>2.5</sub> 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<sub>x</sub>, NO<sub>2</sub>, O<sub>3</sub>, CO, SO<sub>2</sub>) and particle number were recorded as <sup>1</sup>/<sub>2</sub>-hour averages. Particle mass (PM<sub>10</sub> and PM<sub>2.5</sub>) were measured as 24-hour averages using LVS (gravimetric method) but also to a lesser extend as <sup>1</sup>/<sub>2</sub>-hour averages using TEOM. Elements in the particles as well as PAH were measured as 24-hour averages. EC and OC were measured as 24hour averages. Benzene and toluene were measured weekly by passive sampling. Furthermore, volatile organic compounds were sampled as 24-hour averages.

#### 2.1.1 Model calculations

In the monitoring programme, the measurements at the permanent measuring stations are supplemented with model calculations using the THOR modelling system. In the present report, model results are presented for  $NO_2$ ,  $PM_{2.5}$ , and  $PM_{10}$  in streets and for  $O_3$  at a national level.

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

The meteorological data for 2017 from MM5v7 is subsequently used to drive the air pollution models, including the Danish Eulerian Hemispheric Model,

DEHM (Christensen, 1997; Brandt et al., 2012), the Urban Background Model, UBM (Berkowicz, 2000b; Brandt et al., 2001) and the Operational Street Pollution Model, OSPM® (Berkowicz 2000a; Ketzel et al., 2012). DEHM is providing air pollution input data for UBM which again is providing air pollution input data to OSPM. Further details about the integrated THOR system can be found in Brandt et al. (2000; 2001 and 2003 or at <a href="http://www.au.dk/thor">http://www.au.dk/thor</a>). The same model setup is also used for a new 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 is carried out using DEHM (version 5.0), which is an Eulerian model where emissions, atmospheric transport, chemical reactions, and dry and wet depositions of air pollutants are calculated in a 3D grid covering the northern hemisphere with a resolution of 150 km x 150 km. The model includes a two-way nesting capability, which makes it possible to obtain higher resolution over limited areas. Three nested domains are used in the model runs under NOVANA, where the first domain is covering Europe with a resolution of 50 km x 50 km. The second domain is covering Northern Europe with a resolution of 16.7 km x 16.7 km. The calculations of air quality in Denmark are carried out in a third domain with a horizontal resolution of 5.6 km x 5.6 km. In the vertical direction the model is divided into 29 layers covering the lowest 15 km of the atmosphere. Of these, the lowest layers are relatively thin (20 m) while the upper layers are relatively thick (2000 m). The model includes a comprehensive chemical scheme designed for calculation of the chemical reactions in the lower part of the atmosphere. The emission inventories used in DEHM have a geographical resolution of 1 km x 1 km for Denmark 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 2016 compiled by DCE (http://envs.au.dk/en/knowledge/air/emissions/) and international emission inventories for the year 2015 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 adjustment 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 (North Sea and the Baltic Sea) from 1% to 0.1%.

The Urban Background Model, UBM (version v9N2017), calculates the urban background air pollution based on emission inventories with a spatial resolution of 1 km x 1 km and based on input data from DEHM concerning the regional background. UBM is suitable for calculations of urban or rural background concentrations on high resolution (1 km x 1 km). The model includes a Gaussian plume approximation for calculation of the dispersion and transport of the air pollutants to every receptor point and a simple chemical model accounting for the photochemical reactions of NO<sub>x</sub> and O<sub>3</sub>. 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 now treats both area and point sources in a more physically correct manner compared to earlier versions of the model. This has improved the overall performance of the model in comparison with measurements and provides a more realistic spatial distribution of concentrations around large point sources. The emissions used in the UBM model are based on the SPREAD model that spatially distributes national emissions from 2016 from all sectors on a 1 km x 1 km grid for Denmark (more details on the model can be found

in Plejdrup & Gyldenkærne, 2011). In previous years, UBM has been calibrated against measurements at all four urban background stations in order to ensure good correspondence between measured and modelled NO<sub>2</sub>. UBM was applied with the same calibration as in the previous years and no additional corrections were necessary in 2017. No calibration for NO<sub>x</sub> /NO<sub>2</sub> was made since the agreement with measurements at the urban background locations of Copenhagen and Aalborg - where calculations are made for selected streets - was good. For the first time, the annual report includes model calculations of PM<sub>2.5</sub> and PM<sub>10</sub>. Evaluation of the model results with the most reliable PM measurements using LVS instrumentation revealed that a calibration was nessesary for these pollutants; see Appendix 3 for further details.

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

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

The traffic data used as input for the calculations with OSPM is updated annually for average daily traffic and vehicle distribution for the selected streets based on information obtained from the municipalities of Copenhagen and Aalborg. Traffic data are estimated at the location of the calculation points. For Copenhagen, traffic data is based on manual counts performed annually or in 5-year intervals. Aalborg does not have a systematic traffic counting program similar to Copenhagen, and traffic data is based on available traffic data from manual and automatic counts together with data from a traffic model. Based on information from Copenhagen and Aalborg municipalities the Average Daily Traffic (ADT) and vehicle distribution on all streets have been updated with the most recent available traffic data. The vehicle distribution includes passenger cars, vans, trucks<32t, trucks>32t, and buses. In Copenhagen, 26 out of the 98 calculation points had updated traffic data for 2017. For Aalborg 10 out of 31 streets had updated traffic data.

Manual traffic counts are carried out annually for the street segments in front of the measuring stations of H.C. Andersens Boulevard and Jagtvej in Copenhagen. Manual counts for the 2017 assessment originate from September 2017 in Copenhagen. In Aarhus, automatic traffic recording was carried out to estimate traffic volume and vehicle classification during four seperate weeks in March, May, September and November 2017. This method provides good estimate of traffic volume but only rough estimate of vehicle classification. One of the shortcomings is that the method can't 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.

In Odense, the street (Albanigade) with the measuring station was closed in May 2015 due to construction work and traffic has in recent years decreased

considerably due to major changes in the overall traffic plan for Odense City. The station has been moved to a new location in Odense (Grønløkkevej) and started operation in June 2016. Automatic traffic recording was carried out during one week 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 2017 due to nearby building construction work, and work is in progress to move the station to another location at the same street.

The model calculations for 2017 for Copenhagen and Aalborg have been carried out using the full model calculation system based on the THOR system, including MM5v7, DEHM, UBM, and OSPM. The calculations were carried out in order to determine annual means of NO<sub>2</sub>,  $PM_{2.5}$  and  $PM_{10}$  concentration in 98 streets in Copenhagen and 31 streets in Aalborg. In previous years, calculations were only performed for NO<sub>2</sub> but for 2017  $PM_{2.5}$  and  $PM_{10}$  have been included.

#### 2.1.2 Model calibration and validation

In the assessment for 2013, the model calculations with OSPM were improved through major revisions. These included changes related to the general building height, revision of  $NO_x$  emission factors for Euro 5 and 6 for passenger cars, and use of new travel speeds for the traffic based on GPS data (Speed-Map, speedmap.dk/portal/) and subsequent recalibration. Appendix 3 in Ellermann et al. (2014) describes the changes and presents documentation for the improved input data for the model calculations. The model setup for the assessment for 2017 is similar to that of 2013 and onwards.

Before 2015, OSPM was calibrated against measurements at the street stations for a single year in order to ensure good correspondence between measured and modelled NO<sub>2</sub>. For the assessment of 2016 we used available data from the last three years to avoid potential fluctuations that a single year approach may introduce. The same calibration assumptions used in 2016 has also been used for 2017.The correlation between modelled and observed NO<sub>2</sub> concentrations for 2017 shows an overall good agreement with zero bias based on the same calibration as in previous years. The street station of H.C. Andersens Boulevard has not been used in the calibration due to the about 8  $\mu$ g/m<sup>3</sup> 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 for the entire year of 2017.

The comparison between modelled and observed  $NO_2$  concentrations for 2017 are shown in table 2.2. For further details on the calibration and validation see Appendix 3.

Unit: µg/m <sup>3</sup> Measurements Model results D		Difference	Models used	
Street:				
Copenhagen/HCAB/1103	38	38	-1.8%	DEHM/UBM/OSPM
Copenhagen/Jagtvej/1257	28	26	-6.1%	DEHM/UBM/OSPM
Aarhus/6153	28	27	-1.7%	DEHM/UBM/OSPM
Odense/9156	18	21	17%	DEHM/UBM/OSPM
Urban Background:				
Copenhagen/1259	16	13	-15%	DEHM/UBM
Aarhus/6160	12	13	9.2%	DEHM/UBM
Odense/9159	10	11	10%	DEHM/UBM
Aalborg/8159	11	10	-11%	DEHM/UBM
Hvidovre/2650	13	11	-19%	DEHM/UBM
Rural:				
Risø/2090	6.8	8.1	19%	DEHM/UBM
Føllesbjerg/9055	7.0	7.1	0.7%	DEHM/UBM
Anholt/6001	4.1	4.7	17%	DEHM/UBM
Ulborg/7005	3.6	4.1	14%	DEHM/UBM

Table 2.2. Comparison of modelled and measured annual means of NO<sub>2</sub> concentrations in 2017.

The comparison between modelled and observed  $PM_{2.5}$  and  $PM_{10}$  concentrations for 2017 is shown in table 2.3 and table 2.4, respectively. Without any adjustments of the original modelled particle concentrations underestimate the observed concentrations. To compensate for the underestimation the model results of  $PM_{2.5}$  and  $PM_{10}$  have been calibrated to fit measurements with a factor of 1.26 and 1.46 for  $PM_{2.5}$  and  $PM_{10}$ , respectively. The underestimation is most likely related to underestimation of the non-exhaust particles (road wear, tyre wear, brake wear and re-suspension) and/or underestimation of certain particle components as secondary organic aerosols (SOA) or the water content.

**Table 2.3.** Comparison of modelled and measured annual means of  $PM_{2.5}$  concentrations in 2017. A calibration factor of 1.26 has been applied for all modelled PM values to compensate for underestimation of  $PM_{2.5}$ .

Unit: µg/m³	Measurements	Model results	Difference	Models used	
Street:					
Copenhagen/HCAB/1103	14	15	5.0%	DEHM/UBM/OSPM	
Copenhagen/Jagtvej/1257	13	13	-0.9%	DEHM/UBM/OSPM	
Aarhus/6153	12	11	-4.7%	DEHM/UBM/OSPM	
Urban Background:					
Copenhagen/1259	11	11	-2.5%	DEHM/UBM	
Aarhus/6160	8.7	9.6	11%	DEHM/UBM	
Aalborg/8159	8.4	8.0	-4.7%	DEHM/UBM	
Hvidovre/2650	9.3	10	10%	DEHM/UBM	
Rural:					
Risø/2090	9.4	10	6.8%	DEHM/UBM	

Unit: µg/m³	Measurements	Model results	Difference	Models used	
Street:					
Copenhagen/HCAB/1103	28	28	1.5%	DEHM/UBM/OSPM	
Copenhagen/Jagtvej/1257	22	22	-2.8%	DEHM/UBM/OSPM	
Aarhus/6153	18	20	13%	DEHM/UBM/OSPM	
Odense/9156	20	22	10%	DEHM/UBM/OSPM	
Urban Background:					
Copenhagen/1259	17	15	-7.5%	DEHM/UBM	
Rural:					
Risø/2090	14	15	6.6%	DEHM/UBM	

**Table 2.4.** Comparison of modelled and measured annual means of PM<sub>10</sub> concentrations in 2017. A calibration factor of 1.46 has been applied for all modelled PM values to compensate for underestimation of PM<sub>10</sub>.

### 2.1.3 Health impact and external cost of air pollution

Model calculations of the health impact and external cost of air pollution has been included in the air quality monitoring programme as a consequence of the revision of NOVANA in 2016. High-resolution assessment of health impacts from air pollution and related external cost has therefore been carried out for Denmark for the years 2015-2017 using the integrated EVA (Economic Valuation of Air Pollution) model system, version 2.5 (Brandt et al., 2015; 2016). A three-year average is used in order to smooth out variations in the 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.

The air quality data used in the EVA system is based on a coupling of the two chemistry transport models (DEHM and UBM) described above. PM<sub>2.5</sub> is responsible for the majority of the health impact from air pollution in Denmark. Table 2.3 shows a comparison between the measured and model calculated annual concentrations of PM<sub>2.5</sub> at the Danish measurements stations after a calibration of the models with a factor 1.26 and here the agreement between model and measurements are good. However, it has not been possible to recalibrate the entire EVA-system for this reporting and the presented data on health impact and external cost is therefore likely to be underestimated with up to about 25%.

The population density for Denmark is based on the geographical distribution of the Civil Registration System (CPR data) from 2017. The individual health impacts in the EVA system is documented in Brandt et al. (2013a) and reviewed in Bønløkke et al. (2011). The economic valuation of the individual health impacts is from Andersen and Brandt (2014) and the methodology for the economic valuation is documented in Andersen et al. (2004) and Bach et al. (2006). The EVA model system has previously been used for assessment of future scenarios (Geels et al., 2015) and has been compared with other health impact assessment systems (Anenberg et al., 2015).

# 3. Nitrogen oxides

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

# 3.1 Annual statistics

The annual statistics for 2017 for nitrogen dioxide (NO<sub>2</sub>) and nitrogen oxides are shown in table 3.1 and 3.2. There was no exceedance of the annual limit value for NO<sub>2</sub> of 40  $\mu$ g/m<sup>3</sup> (EC, 2008). Further, there were no exceedances of the hourly limit value for NO<sub>2</sub> of 200  $\mu$ g/m<sup>3</sup>. This value must not be exceeded more than 18 times in a calendar year (see 19th highest hourly concentration in table 3.1). In 2017, there was no information to the public triggered by exceedance of the information threshold for NO<sub>2</sub> (three hours average must not exceed 400  $\mu$ g/m<sup>3</sup>). During the summer of 2017 there were quality problems with the monitors used for the measurements. Regretebly the problems resulted in the EU requirement of 7446 hours of hourly averaged values not being upheld at the street station in Aarhus, Aarhus/6153, column two in table 3.1 and 3.2.

Unit: µg/m³	Number	Average	Median	98-percentile	19-highest
Street:					
Copenhagen/1257	8092	28	23	79	108
Copenhagen/1103*	8085	38	35	89	121
Aarhus/6153	7244	28	25	72	92
Odense/9156 §§	8262	18	15	53	71
Aalborg/8151 §	0	-	-	-	-
Urban Background:					
Copenhagen/1259	8171	16	12	56	84
Aarhus/6160	7961	12	9	45	62
Odense/9159	8039	10	7	36	53
Aalborg/8158	8132	11	8	48	76
Suburban:					
Hvidovre/2650	7675	13	10	49	79
Rural:					
Risø	7763	7	4	30	58
Keldsnor/9055	7661	7	4	30	54
Limit value 2010	>7446*	40			200

Table 3.1. Nitrogen dioxide (NO<sub>2</sub>) in 2017. All parameters are based on hourly averages.

\*) 90% data capture of number of hourly measurements in relation to total number of hourly measurements in 2017 excluding hours used for calibration.

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

§§) The site in Odense/9155 (Albanigade) was affected by a major permanent rearrangement of the roads in Odense. The station changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on 28 June 2014. The station was shut down on 16 June 2015 and has been moved to a new position in summer 2016 and renamed Odense/9156.

Table 3.2. Nitrogen oxides	$(NO_x=NO+NO_2)$ in	2017. All parameters are based	l on hourly averages.
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Unit: µg/m³ (as NO <sub>2</sub> )	Number	Average	Median	98-percentile	19-highest
Street:					
Copenhagen/1257	8092	56	39	211	404
Copenhagen/1103	8085	88	70	281	516
Aarhus/6153	7244	61	44	225	428
Odense/9156 §§	8262	35	26	131	293
Aalborg/8151 §	0	-	-	-	-
Urban Background:					
Copenhagen/1259	8262	19	13	72	149
Aarhus/6160	7961	16	10	77	203
Odense/9159	8039	13	9	54	148
Aalborg/8158	8132	16	10	81	286
Suburban:					
Hvidovre/2650	7678	18	11	81	268
Rural:					
Risø	7763	8	5	35	95
Keldsnor/9055	7661	8	5	34	64

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

§§) The site in Odense/9155 (Albanigade) was affected by a major permanent rearrangement of the roads in Odense. The station changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on 28 June 2014. The station was shut down on 16 June 2015 and has been moved to a new position in summer 2016 and renamed Odense/9156.

#### 3.2 Trends

The long-term trends for  $NO_2$  and  $NO_x$  are shown in figure 3.1. For  $NO_x$  there are clear downward trends at all stations. The decreases in the concentrations of nitrogen oxides are due to the national and international regulations of the emissions. The large emission reductions in the cities are achieved by improvement of the vehicles, for example mandatory use of catalytic converters.

For many years the long-term trend for nitrogen dioxide has decreased much slower than observed for  $NO_x$ . However, since around 2006  $NO_2$  has decreased with about the same rate as  $NO_x$ . The slow 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 roads in Odense Centre that changed the traffic at the street station in Albanigade in two steps from a street with relatively high traffic intensity to a street with much reduced traffic intensity. Finally, the street was closed for traffic in 2015. These changes began on 28 June 2014. This is the reason for the large decrease of the NO<sub>2</sub> and NO<sub>x</sub> 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. Results from the previous (6159) and the new background station (6160) in Aarhus are shown on the same curve.

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 before 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<sub>2</sub> of about 8  $\mu$ g/m<sup>3</sup> in comparison to the levels measured before the introduction of the new driving lanes. The data from 2017 shows the full impact of the relocation of NO<sub>2</sub> 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<sub>2</sub> have been performed for selected streets in Copenhagen (capital) and Aalborg (fourth largest city). The selected streets represent busy streets and are mainly so-called street canyons. Concentrations are elevated in this type of streets due to the high emissions and restricted dispersion conditions. 98 streets are included for Copenhagen and 31 in Aalborg. ADT (Average Daily Traffic) was between 5,400 and 67,600 vehicles/day in Copenhagen and between 2,700 and 29,000 vehicles/day in Aalborg.

Model calculations have been carried out in order to determine the annual concentrations of NO<sub>2</sub> for comparison with the limit values. The airquality limit value for the annual mean is  $40 \ \mu g/m^3$ . 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  $\mu g/m^3$  since the limit value is given as an integer.

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

The annual mean concentrations of NO<sub>2</sub> for streets in Copenhagen in 2017 are shown in figure 3.2 (bar chart) and figure 3.3 (map). The average of the NO<sub>2</sub> street concentrations at all 98 streets decreased from 2016 to 2017 (-4.5 µg/m<sup>3</sup>) and the average urban background concentrations decreased slightly (-1.1  $\mu$ g/m<sup>3</sup>). However, the regional background contribution was similar in 2016 and 2017. The decrease in street concentrations is a result of a combination of changes in traffic, emission factors, urban background and meteorology. There has been no average change in traffic as the ADT and heavy-duty share remained the same as in 2016 and travel speeds are assumed to be the same as in 2016. 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 place a significant role. Further, the directly emitted NO<sub>2</sub> of NO<sub>x</sub> emissions (NO<sub>2</sub> fraction) has also decreased leading to lower modelled NO2 concentrations. Lower NOx emissions and lower NO<sub>2</sub> fraction are the main reasons for the modelled decrease in NO<sub>2</sub> street concentrations. For 2017 the NO2 fraction is 12% based on analysis of measurements of NO<sub>x</sub>, NO<sub>2</sub> and O<sub>3</sub> whereas for 2016 it was 24% based on information from the COPERT emission model. The latter is likely too high. The

lower urban background concentrations also contribute to lower street concentrations. Finally, the average wind speed was a little higher in 2017 compared to 2016 contributing to lower concentrations in 2017 compared to 2016.

In 2017, the limit value for the annual mean concentration was not exceeded at any of the 98 selected streets in Copenhagen according to the model results although the highest modelled concentrations are still close to the limit value (figure 3.2). This is the first time that no exceedances have been calculated since the beginning of the calculations in 2007. However, the number of streets exceeding the limit value is very sensitive to 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<sub>2</sub> in 2017 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<sup>®</sup> (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<sub>2</sub> concentrations from 2016 to 2017 mainly due to small changes in traffic inputs. The highest modelled NO<sub>2</sub> concentration in 2017 is at H.C. Andersens Boulevard (2) (38.2  $\mu$ g/m<sup>3</sup>). The second highest (37.7  $\mu$ g/m<sup>3</sup>) is where the measuring station is located (H.C. Andersens Boulevard (1)).

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

**Table 3.3.** Rank number and names for the street segments that are shown in figure 3.2 and 3.3. The streets are numbered (1-98) according to  $NO_2$  levels in 2017 (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.



**Figure 3.3.** Map showing the locations of the selected streets in Copenhagen and the annual mean concentrations of  $NO_2$  for 2017 together with the rank number 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 <a href="https://arcg.is/1mmWHK">https://arcg.is/1mmWHK</a>.

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

For Aalborg the modelled street concentrations show an average decrease of about 5  $\mu$ g/m<sup>3</sup> for NO<sub>2</sub> compared to 2016 when considering all 31 street segments. This includes a decrease of about  $3 \mu g/m^3$  for urban background concentrations, and regional background concentrations are unchanged. The general decrease is a result of a combination of several factors. On average ADT decreased about 1.5% whereas the heavy-duty share of vehicles was unchanged, and travel speeds were assumed to be unchanged. This would - all other parameters equal - only slightly decrease concentrations due to these changes in traffic inputs. 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. Further, the directly emitted NO<sub>2</sub> of NO<sub>x</sub> emissions (NO<sub>2</sub> fraction) has also decreased leading to lower modelled NO2 concentrations. Lower NO<sub>x</sub> emissions and lower NO<sub>2</sub> fraction are the main reason for the modelled decrease in NO<sub>2</sub> street concentrations. For 2017 the NO<sub>2</sub> fraction is 12% based on analysis of measurements of NO<sub>x</sub>, NO<sub>2</sub> and O<sub>3</sub> whereas for 2016 it was 24% based on information from the COPERT emission model. The latter is likely too high. Finally, the average wind speed was a little higher in 2017 compared to 2016 contributing to lower concentrations in 2017 compared to 2016, other things equal.

According to the model calculations the limit value for the annual mean concentration in 2017 was not exceeded at any of the 31 selected streets which was also the case in 2016 (figure 3.4 and figure 3.5). The order of some of the streets has changed slightly due to changes in traffic data.



**Figure 3.4.** Modelled annual mean concentrations of NO<sub>2</sub> in 2017 for 31 streets in Aalborg. The contribution from traffic in the street canyons is based on the street canyon model OSPM<sup>®</sup> (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 2017 due to nearby building construction works, and plans are in place to move the station to a new location at the same street.



**Figure 3.5.** Map showing the location of the selected streets in Aalborg and the annual mean concentrations of NO<sub>2</sub> for 2017. The contribution from traffic in the street canyons is based on the street canyon model OSPM<sup>®</sup>. 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 2017 due to nearby building construction work. Map can be viewed at a webGIS service, see <a href="https://arcg.is/19yH5g">https://arcg.is/19yH5g</a>

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

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

For Copenhagen the number of exceedances has decreased from 58 in 2008 to 6 in 2016, and further to zero in 2017. The main reason for the increase in number of exceedances in Copenhagen from 32 in 2007 to 58 in 2008 is the following: The limit value plus margin of tolerance for the annual mean concentration of NO<sub>2</sub> decreased from 46  $\mu$ g/m<sup>3</sup> in 2007 to 44  $\mu$ g/m<sup>3</sup> 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  $\mu$ g/m<sup>3</sup> in 2007, the number of streets exceeding the limit value plus margin of tolerance had been 44  $\mu$ g/m<sup>3</sup> in 2007, the number of streets exceeding the limit value plus margin of tolerance had been 44  $\mu$ g/m<sup>3</sup> in 2007, the number of streets exceeding the limit value plus margin of tolerance had been 44  $\mu$ g/m<sup>3</sup> 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 3-4 exceedances were modelled in 2007-2009 and none since 2010. Here the analysis includes 32 streets from 2007 to 2010, and 31 streets from 2011 onwards.



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

# 4. Ozone

O<sub>3</sub> is measured at seven monitoring sites using gas monitors based on ultraviolet photometry. The concentrations are measured continuously throughout the year with a time resolution of minutes that is aggregated to hourly averages for the present report.

# 4.1 Annual statistics

The annual statistics for 2017 for  $O_3$  are shown in table 4.1. The maximum 8-hour daily mean value must not exceed 120 µg/m<sup>3</sup> more than 25 days per calendar year averaged over three years (EC, 2008). This target value were not exceeded for 2015-2017 at any of the stations. The long-term objective (maximum 8-hour daily mean value must not exceed 120 µg/m<sup>3</sup>; table 4.1 column 5) was exceeded at two of the stations. However, the long-term objective has not entered into force.

In 2017, there were no exceedance of the information threshold (hourly average 180  $\mu$ g/m<sup>3</sup>). There was also no exceedance of the alert threshold (hourly average 240  $\mu$ g/m<sup>3</sup>) for O<sub>3</sub>.

**Table 4.1.**  $O_3$  in 2017. All parameters are based on one-hour average values. The 8-hour values are calculated as a moving average based on hourly measurements. Days above target value is the number of days that the maximum running 8-hour average exceeds 120 µg/m<sup>3</sup> averaged over 2015-2017.

Unit: µg/m³	Number of results	Average	Median	Max 8-hours	Days above target value 8-hours	Max 1 hour
Urban Background:						
Copenhagen/1259	7728	57	58	124	1	133
Aarhus/6160	7450	53	55	112	0	118
Odense/9159	7838	58	59	121	1	132
Aalborg/8158	7775	57	60	110	0	116
Rural						
Risø/2090	7974	61	63	115	0	125
Keldsnor/9055	7765	58	59	104	0	116
Traffic						
Copenhagen/1103	7765	38	38	96	0	108
Target value*	-	-	-	-	25	-
Long term objective	-	-	-	120	-	-
Information thres- hold	-	-	-	-	-	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 2017 excluding hours used for calibration.

## 4.2 Trends

The long-term trends of  $O_3$  are shown in figure 4.1. The annual averages of  $O_3$  have increased slightly on most of the stations since 1992. The Danish and European reductions of the precursors to  $O_3$  formation (NO<sub>x</sub>, volatile organic compounds) have therefore not been sufficient to reduce the concentrations. However, the reductions of the precursors have decreased the maximum concentrations of  $O_3$ . This is illustrated by the decrease in the maximum 8-hour average concentrations.



**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, because  $O_3$  is deposited faster over land than over sea. In the cities the concentrations are lower than the average, because  $O_3$  is degraded by nitrogen oxide emitted from mainly traffic in the cities. This is clearly seen for Copenhagen. Model results are slightly lower in 2017 than in 2016 while the measured values were on the same level in 2016 and 2017.

The target value for protection of human health is the running 8-hour mean concentration of  $O_3$  must not exceed 120 µg/m<sup>3</sup> more than 25 times during a calendar year calculated as an average over three years. The long-term objectives are that the running 8-hour mean concentration of  $O_3$  must not exceed 120 µg/m<sup>3</sup>. The target value and long-term objective are given in the EU Directive (EC, 2008). Results from the model calculations for 2017 show that the number of days with maximum daily 8-hour mean value above 120 µg/m<sup>3</sup> was well below the target value for the entire country in 2017. The target value that is determined as an average over three years (2015-2017), was not exceeded since the number of days with exceedances in 2015 and 2016 were well below 25 as well (Ellermann et al., 2016, 2017).

The highest number of days with exceedance of  $120 \ \mu g/m^3$  was seen at coastal areas in south western Jutland where the maximum number of days reached only two days above  $120 \ \mu g/m^3$  (figure 4.3) and for the main part of Denmark the days above  $120 \ \mu g/m^3$  was zero. This is considerably less than in 2016 were the maximum numbers of days with exceedances was 13. The large decrease in the maximum daily 8-hour mean value was due to the rainy and cold summer weather in 2017 that decreased the number of episodes with high concentrations of O<sub>3</sub>. As a consequence of the relatively large decrease in the maximum daily 8-hour mean values there were for most of the country no exceedance of the long-term objective (figure 4.4). The highest 8-hour mean concentrations were observed at coastal areas due to slow deposition over sea and long-range transport of O<sub>3</sub>.

According to the directive (EC, 2008) the public has to be informed if the 1-hour average concentration exceeds the information threshold at  $180 \ \mu g/m^3$ . Both measurements and model calculations showed that there was no exceedance of the threshold in 2017 (figure 4.5). This difference is because model calculations underestimate the maximum 1-hour mean concentration with about 10-20%. One of the reasons for this discrepancy is most likely that the model does not include emissions of O<sub>3</sub> precursors from wild fires that are known to increase episodic O<sub>3</sub> concentrations.


**Figure 4.2.** Annual mean concentrations of  $O_3$  ( $\mu$ g/m<sup>3</sup>) for 2017 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  $\mu$ g/m<sup>3</sup> for 8-hour running mean concentrations of O<sub>3</sub> in 2017. The calculations were carried out using DEHM.



Figure 4.4. Maximum 8-hour running mean concentration ( $\mu$ g/m<sup>3</sup>) of O<sub>3</sub> in 2017 calculated using DEHM.



Figure 4.5. Maximum 1-hour mean concentration of  $O_3~(\mu g/m^3)$  in 2017 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 2017 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  $\mu$ g/m<sup>3</sup> (EC, 2008). This limit value was not exceeded at any of the stations.

**Table 5.1.** Annual statistics for CO in 2017. All parameters are based on hourly average. The 8-hour values are calculated as a moving average based on hourly results.

Unit: µg/m³	Number	Average	Median	98- percentile	99.9- percentile	Max. 8-hours	Max. hour
Traffic:							
Copenhagen/1103	8253	282	258	641	711	1176	2133
Århus/6153	7860	232	211	546	1062	996	1696
Odense/9156 §	8221	226	201	557	1083	947	1759
Aalborg/8151 §§	0	-	-	-	-	-	-
Urban Background:							
Copenhagen/1259	8156	201	188	422	636	630	774
Rural:							
Risø	8005	178	167	403	638	633	727
Data capture*	>7446	-	-	-	-	-	-
EU Limit value	-	-	-	-	-	10 000	-
WHO Guideline values	-	-	-	-	-	10 000	30 000

\*) 90% data capture of number of hourly measurements in relation to total number of hourly measurements in 2017 excluding hours used for calibration.

§) The site in Odense/9155 (Albanigade) was affected by a major permanent rearrangement of the roads in Odense. It changed from a traffic site with relatively high traffic intensity to a site with much reduced traffic intensity. This change took place on 28 June 2014. The station was shut down on 16 June 2015 and has been moved to a new position during the summer of 2016 and was renamed to Odense/9156.

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

## 5.2 Trends

The long-term trends for CO 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 and benzene and toluene, all of which are Volatile Organic Compounds (VOC).

Benzene, toluene, ethylbenzene and xylenes are monitored on two kerbside stations in Copenhagen in weekly 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) with 16 other potential  $O_3$  precursor VOCs in diurnal time resolution. The focus is VOCs of anthropogenic origin, though isoprene which is typically emitted from deciduous trees is included. Air is sampled and preconcentrated on Carbopack X adsorbents 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 2017. Benzene is well below the EU-limit value of 5  $\mu$ g/m<sup>3</sup> (EC, 2008), averaging 0.64 and 0.68  $\mu$ g/m<sup>3</sup> at the kerbside stations 1257 and 1103, and 0.49  $\mu$ g/m<sup>3</sup> in urban background. Thus, the local input of benzene from traffic amounts to 24% of the concentration at the kerbside station 1257. For toluene, the local input is as high as 45%. Other than 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 show similar concentrations of anthropogenic aromatic compounds, including toluene and benzene (table 6.1), in spite of their differences with respect to traffic load and buildings close to the street. These VOCs decreased dramatically at the kerbside stations during 2004-2008 (figure 6.1) and has continued to do so, though at a slower yet comparable rate in the urban environment. In fact, benzene has decreased by 52% and 35% at the kerbside station 1257 and urban background 1259, respectively, from 2010 to 2017. With respect to toluene, the corresponding decreases were 51% and 36%, 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 2017 based on weekly average concentrations ( $\mu$ g/m<sup>3</sup>) at kerbside stations Jagtvej (1257) and H.C. Andersens Boulevard (1103) at 1 atm., 293 K. The limit value for benzene is 5  $\mu$ g/m<sup>3</sup> (EU Directive 2008/50/EC).

Concentration µg/m <sup>3</sup>	Copenhagen/1103	Copenhagen/1257	Number of results
Benzene	0.68	0.64	44, 52
Toluene	1.63	1.59	44, 52
Ethylbenzene	0.31	0.32	44, 52
m/p-Xylene	0.67	0.69	44, 52
o-Xylene	0.37	0.40	44, 52

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

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

Benzene = 0.0044 · CO - 0.37

where benzene and CO are in units of  $\mu g/m^3$ . Based on this and the concentrations of CO (table 5.1) the annual average concentrations of benzene are estimated to about 0.6  $\mu g/m^3$  for all the three street stations in Aarhus, Odense and Aalborg in 2017.



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

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

Concentration (µg/m <sup>3</sup> )	Annual average 2010	Annual average 2017	Data coverage
1-Pentene	0.04	0.03	82%
n-Pentane	0.53	0.61	84%
Trans-2-pentene	0.02	0.02	85%
Isoprene	0.03	0.05	85%
2-Methylpentane	0.31	0.30	78%
n-Hexane	0.19	0.18	86%
Benzene	0.75	0.49	86%
n-Heptane	0.28	0.15	85%
2,2,2-Trimethylpentane	0.10	0.07	85%
Toluene	1.36	0.87	86%
n-Octane	0.08	0.04	86%
Ethylbenzene	0.28	0.14	86%
m,p-Xylene	0.78	0.44	86%
o-Xylene	0.41	0.16	86%
1,3,5-Trimethylbenzene	0.10	0.03	85%
1,2,4-Trimethylbenzene	0.34	0.13	85%
1,2,3-Trimethylbenzene	0.09	0.03	86%
Sum of VOCs	5.68	3.76	

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

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<sub>5</sub>-C<sub>7</sub> alkanes: pentane, 2-methylpentane hexane and heptane. The more reactive unsaturated compounds are less abundant (table 6.2).



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

The annual isoprene concentration has remained fairly constant from 2010-2017. Isoprene origins mainly from natural sources, e.g. terrestrial vegetation and 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 2010-2017 or decreased.

The urban background ratio between toluene and benzene is somewhat smaller than the kerbside station 1257, i.e. 1.8 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<sub>10</sub>, PM<sub>2.5</sub> and particle number)

The measurements of particle mass ( $PM_{10}$  and  $PM_{2.5}$ ) are today solely carried out using 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<sup>3</sup>/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 old SM200 beta ( $\beta$ ) sampler (manufactured by OPSIS, Sweden) that collects particles on filters on a diurnal basis with subsequent determination of the sampled mass using  $\beta$ -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 using the LVS instruments.

Additionally, PM is measured using a TEOM (Tapered-Element Oscillating Microbalance) instrument at the Copenhagen street station HCAB (PM<sub>10</sub> and PM<sub>2.5</sub>), at the Aarhus street station (PM<sub>10</sub>) and at the rural station at Risø (PM<sub>10</sub>). The TEOM measurements have a time resolution of 30 minutes (table 7.3 and 7.4) and enable near real time reporting of the data to the public. During sampling, the collected particles are heated to 50°C. At that temperature some of the semi-volatile particle constituents evaporate (mainly secondary aerosols and especially ammonium nitrate, NH<sub>4</sub>NO<sub>3</sub>). The loss will depend of the actual composition of the aerosols. The European Commission has accepted that measurements of PM using TEOM could be applied with a default correction factor of 1.3. However, the correction factor depends e.g. on the specific measurement site and seasonality and correction of TEOM measurements of PM using a correction factor of 1.3 gives considerable uncertainty on the corrected values.

Measurements of particle number concentrations have been carried out since 2001/2002 in cooperation between the monitoring programme and research projects financed 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. Andersens Boulevard in Copenhagen and at the regional background station Risø were also replaced by two new SMPS systems. Subsequently is has been shown that the new SMPS instruments or the new inlets of the instruments cause problems with the measurements of the smallest particles in the size range from 11 to 41 nm. Data from this size range will therefore not be presented for 2017. Intensive work has been carried out to solve these problems together with the manufacturer of the instruments and this work is still ongoing. The data presented in this report for particle number for 2017 shall therefore be regarded as preliminary.

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 2017, the permitted number of exceedances in a year of the diurnal limit value of  $50 \ \mu\text{g/m}^3$  for  $\text{PM}_{10}$  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  $\text{PM}_{10}$  ( $40 \ \mu\text{g/m}^3$ ) and  $\text{PM}_{2.5}$  (of 25  $\ \mu\text{g/m}^3$ ) 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<sub>2.5</sub>. In Denmark the average exposure indicator is measured in urban background at Copenhagen/1259, Aarhus/6159 and Aalborg/8158. For the years 2015-17 the AEI is determined to 10  $\mu$ g/m<sup>3</sup> which is a decrease of about 30% since 2010.

In 2017, the number of particles in ambient air was about 13,000 particles per cm<sup>3</sup> at the street station H.C. Andersens Boulevard (table 7.5). This is a factor of about 3.5 higher than suburban and 4.5 higher than in urban background and rural background, respectively.

Unit µg/m³	Number of results	Average (µg/m³)	Median	Days above 50 μg/m³	90- percentile	Max. day
Street						
Copenhagen/1103	356	25	23	15	40	90
Copenhagen/1257	342	21	17	10	35	88
Århus/6153	332	17	15	2	29	64
Odense/9156	347	19	16	3	32	90
Urban background						
Copenhagen/1259	358	15	13	6	27	68
Rural						
Risø	345	14	11	3	24	65
Keldsnor/9055	351	15	12	3	27	65
Limit value (2005)		40		35**		
90% data capture	>328*					

Table 7.1. Annual statistics for PM<sub>10</sub> in 2017. All parameters are given as diurnal averages at ambient temperature and pressure.

Measurements at all stations in 2017 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 2017 (365).

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

Table 7.2. Annual statistics for PM<sub>2.5</sub> in 2017. 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	346	13	11	22	68	
Copenhagen/1257	360	12	9	21	68	
Aarhus/6153	346	11	9	21	57	
Aalborg/8151*						
Suburban						
Hvidovre/2650	333	9	7	17	61	
Urban background						
Copenhagen/1259	345	10	8	19	62	
Aarhus/6159	352	8	6	16	51	
Ålborg/8158	349	8	6	13	49	
Rural						
Risø	351	9	6	17	61	
Limit value (2015) (parenthesis gives proposed value for 2020)		25(20)				

 90% data capture
 >328\*\*

 Measurements at all stations in 2017 were based on low volume sampling (LVS) with gravimetric determination of particle mass

\* No data from Aalborg/8151 (traffic site) in 2017 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 2017 (365)

**Table 7.3.** Annual statistics for  $PM_{10}$  measured in 2017 using TEOM. The values are based on  $\frac{1}{2}$ -hourly averages. Total annual number of  $\frac{1}{2}$ -hours is 17.520.

Unit: µg/m³	Number of results Average		Average x 1.3
Street			
Copenhagen/1103	17170	26	34
Aarhus/6153	15930	15	19
Rural			
Risø	15835	12	15
Limit value			40

**Table 7.4.** Annual statistics for  $PM_{2.5}$  measured in 2017 using TEOM. The values are based on ½-hourly averages. Total annual number of ½-hours is 17.520.

Unit: µg/m³	Number of results Averag		Average x 1.3
Street			
Copenhagen/1103	17358	10	13
Limit value (2015) (parenthesis gives proposed value for 2020)			25 (20)

**Table 7.5.** Annual statistics for particle number measured in 2017. All values are based on ½-hourly averages. Total annual number of ½-hours is 17.520.

Unit: particles per cm <sup>3</sup>	Number of results	Average 41- 550/478nm)
Street		
Copenhagen/1103**	12994	5168
Urban Background		
Copenhagen/1259*	6705	1895
Suburban		
Hvidovre/2650**	13159	2595
Rural		
Risø**	12557	1977

\* 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  $\mu$ m (figure 7.1). The exact cut-off depends strongly on the wind velocity. From 2001 most of the measurements of particulate matter were changed from TSP to PM<sub>10</sub> according to the EU directive adopted in 1999 (EC, 1999) and PM<sub>10</sub> measurements were started at all stations except Copenhagen/1103 where the TSP measurements were continued to the end of 2005. The TSP is on the average 30-80% higher than PM<sub>10</sub> at the street stations, while the difference is less at urban background and rural sites.

The measurements show a tendency for a decrease in  $PM_{10}$  at all the measurement stations since 2001, where the measurements began (figure 7.2). Although the measurements at HCAB (Copenhagen/1103) began later, there is also a decrease in  $PM_{10}$  at this station. However, this is mainly due to a major reduction (7 µg/m<sup>3</sup>) 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 2008-10. 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 interannually variation in  $PM_{2.5}$  due to the natural variations in the meteorological conditions. Over the period 2010 to 2017 the AEI has been reduced with about 30%.

The measurements show a significant reduction of the particle 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. Andersens Boulevard the number of particles in the range from 41 to 550 has decreased with 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 rural background station (r).



**Figure 7.2.** Annual averages for PM<sub>10</sub> measured at street stations (s), urban background stations (u) and at rural background stations (r). The change from gravimetric determination using the SM200 as a filter sampler to the use of the same instrument as a  $\beta$ -gauge from 2006 gives rise to a 5-10% increase due to the shift in method. Data are given at standard temperature- and pressure conditions (0°C and 1 atm.). PM given at ambient temperature and pressure conditions is on an annual average approximately 3-4% lower than PM-results given at standard conditions.



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



**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 are 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<sup>-3</sup> 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 particle in the range from 478 – 550 nm is very small.

## 7.3 Impact of salt from winter salting and sea

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

On the basis of this, the monitoring program was expanded in 2010 with daily sampling and analysis of sodium at the street stations H.C. Andersens Boulevard, Copenhagen (1103) and Aarhus (6153) and at the urban background station in Copenhagen (H.C. Ørsted Institute/1259). Table 7.6 gives the annual average concentrations for sodium and estimate for total salt (NaCl) in 2017 (calculated from the measured sodium concentration).

	Na µg/m³	NaCl µg/m <sup>3</sup>				
Street:						
Copenhagen/1103	1.2	3.2				
Aarhus/6153	1.3	3.2				
Urban Background:						
Copenhagen/1259	0.9	2.4				

Table 7.6. Annual statistics for sodium and estimate of total salt (NaCl) in 2017.

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



Figure 7.6. Diurnal concentrations in 2017 of sodium at H.C. Andersens Boulevard, Copenhagen (1103) and at the urban background in Copenhagen (H.C. Ørsted Institute/1259).

In 2017, the permitted number of exceedances in a year of the diurnal limit value of 50  $\mu$ g/m<sup>3</sup> for PM<sub>10</sub> was not violated at any stations in the measuring network and therefore it has not been necessary to correct PM<sub>10</sub> for the content of NaCl due to sea salt and salting of the roads in the winter.

## 7.4 PM<sub>2.5</sub> and PM<sub>10</sub> 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 performed and reported for the first time within the Danish Air Quality Monitoring Program. 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 5,400 and 67,600 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  $\mu$ g/m<sup>3</sup> for  $PM_{2.5}$  and  $PM_{10}$ , respectively (EC, 2008).

Modelled  $PM_{2.5}$  and  $PM_{10}$  concentrations for Copenhagen are shown in figure 7.7 and figure 7.8, respectively. The rank numbers from the ranking of  $NO_2$  is maintained and street numbers are shown in table 7.7.

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



**Figure 7.7.** Modelled PM<sub>2.5</sub> concentrations for Copenhagen in 2017. The streets are ranked according to the concentrations of NO<sub>2</sub> (Chapter 3.3). Arrows indicate street segments with a measuring station.



**Figure 7.8.** Modelled  $PM_{10}$  concentrations for Copenhagen in 2017. The streets are ranked according to the concentrations of NO<sub>2</sub> (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 7.9 and figure 7.10, respectively. The rank numbers from the ranking of  $NO_2$  is maintained and street numbers are shown in table 7.7.

Modelled  $PM_{2.5}$  and  $PM_{10}$  concentrations for Aalborg are shown in figure 7.9 and figure 7.10, respectively. The rank numbers from the ranking of  $NO_2$  is maintained and street numbers are shown in table 7.7.

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



Figure 7.9. Modelled  $PM_{2.5}$  concentrations for Aalborg in 2017.



Figure 7.10. Modelled  $PM_{10}$  concentrations for Aalborg in 2017.

**Table 7.7.** 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<sub>2</sub> levels in 2017 (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		Street name
1	H C Andersens Boulevard(2)	34	Vester Farimagsgade	67	Grøndals Parkvej
2*	H C Andersens Boulevard(1)	35	Vesterbrogade(3)	68	Godthåbsvej(2)
3	H C Andersens Boulevard(3)	36	Torvegade	69	Jagtvej(2)
4	Gyldenløvesgade	37*	Jagtvej(1)	70	Hulgårdsvej(2)
5	Øster Søgade	38	Gammel Kongevej(1)	71	Bülowsvej(2)
6	Stormgade	39	Nørre Farimagsgade	72	Øster Voldgade(2)
7	Åboulevard(1)	40	Jagtvej(3)	73	Røde Mellemvej(1)
8	Åboulevard(3)	41	Nordre Fasanvej(3)	74	Frederiksborgvej(1)
9	Ågade	42	P Knudsens Gade(2)	75	Ålholmvej(2)
10	Bernstorffsgade(1)	43	Frederikssundsvej(1)	76	Rebildvej
11	Nørre Søgade	44	Nørre Voldgade(2)	77	Englandsvej(1)
12	Amagerbrogade(2)	45	Strandvejen(1)	78	Dag Hammarskjølds Allé
13	Vesterbrogade(1)	46	Frederikssundsvej(8)	79	Blegdamsvej
14	Bernstorffsgade(2)	47	Amagerfælledvej	80	Frederikssundsvej(2)
15	Bredgade	48	Nørrebrogade	81	Tuborgvej(1)
16	Frederikssundsvej(3)	49	Søndre Fasanvej(2)	82	Folke Bernadottes Allé
17	Østerbrogade(4)	50	Godthåbsvej(3)	83	Peter Bangs Vej(1)
18	Fredensgade	51	Tagensvej(1)	84	Slotsherrensvej(2)
19	Tagensvej(2)	52	Roskildevej(1)	85	Amagerbrogade(3)
20	Enghavevej	53	Østerbrogade(1)	86	Vesterfælledvej
21	Toftegårds Allé(1)	54	Jyllingevej(1)	87	Peter Bangs Vej(2)
22	H.C. Ørsteds Vej(2)	55	Ålholmvej(1)	88	Bellahøjvej
23	Nordre Fasanvej(1)	56	Folehaven(1)	89	Slotsherrensvej(1)
24	Falkoner Alle(2)	57	Tuborgvej(2)	90	Halmetgade
25	Øster Voldgade(1)	58	Amager Boulevard	91	Artillerivej
26	Tomsgårdsvej(2)	59	Ingerslevsgade	92	Strandvænget(2)
27	Hammerichsgade	60	Istedgade	93	Strandvejen(2)
28	Amagerbrogade(1)	61	Hillerødgade(1)	94	Gammel Køge Landevej(2)
29	Gothersgade(1)	62	Kalvebod Brygge	95	Frederiksborgvej(2)
30	Tagensvej(3)	63	Gammel Køge Landevej(1)	96	Vigerslevvej(2)
31	Toldbodgade	64	Tagensvej(4)	97	Røde Mellemvej(2)
32	Lyngbyvej(2)	65	Hillerødgade(3)	98	Englandsvej(2)
33	Scandiagade	66	Frederikssundsvej(5)		

## 8. Heavy metals

Heavy 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 selected elements. At the measurement stations HCAB, HCOE and in Aarhus, LVS equipped with  $PM_{10}$  inlets have replaced existing air samplers in the second half of 2017. Results for 10 heavy metals are presented in table 8.1. The table also presents results for analysis of heavy metals in TSP at the measurement station Risø. The content of these heavy metals in  $PM_{10}$  and TSP at the rural measurement station Risø are approximately equal since these metals are mainly found in the fine particle fraction.

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

## 8.1 Annual statistics

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

**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 2017. For comparison the table also includes results for these heavy metals measured in TSP at the rural background station Risø.

Unit ng/m <sup>3</sup>	V	Cr	Mn	Ni	Cu	Zn	As	Se	Cd	Pb
PM <sub>10</sub> , Street										
Copenhagen/1103	1.8	6.6	18	1.8	71	48	0.7	0.4	0.07	4.2
Aarhus/6153	1.0	2.5	6.4	1.4	26	19	0.5	0.4	0.05	2.0
PM <sub>10,</sub> Urban background:										
Copenhagen/1259	1.1	0.8	3.5	1.0	10	12	0.5	0.4	0.06	2.5
TSP, Rural background										
Risø	0.9	1.0	2.6	0.6	2.8	7.8	0.4	0.3	0.05	1.9
EU Target (Limit) Values *				20			6		5	500
Guideline value (WHO)**	1000		150						5	
Life time risk level at 1:105				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 life time risk for the carcinogenic metals are established by WHO (WHO, 2000). The lifetime risk level is defined as the concentration that through a lifelong exposure is estimated to give an excess risk of 1:105 for developing cancer.

Table 8.2. Annual statistics for Hg 2017 measured at Råö in southern Sweden by the Swedish Environmental Research Institute.

Unit: ng/m³	Total Gas Hg (ng/m³)	Total Particles Hg (ng/m³)
Råö (SE00014)	1.4	0.005

#### 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).

For Mn the long-term trend at H.C. Andersens Boulevard deviates from the other stations. This is believed to be due to high Mn concentrations in the asphalt used at H.C. Andersens Boulevard during the period from 1991 to 2008 (in 2008 the type of asphalt was changed on HCAB). The sharp decrease in concentration in 2006 is due to the change in sampling method from TSP to  $PM_{10}$ .



**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<sub>2</sub>) has reached very low levels in Denmark and it is therefore only necessary with a limited monitoring of the concentrations; both with respect to the number of stations and the quality of the measurements. Hence it is only measured at two traffic stations (Copenhagen and Aalborg) with focus on episodes with high concentrations of SO<sub>2</sub>. It is measured using gas monitors based on ultraviolet fluorescence. The concentrations of SO<sub>2</sub> 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 2017 for SO<sub>2</sub> are shown in table 9.1. None of the limit values (EU, 2008) were exceeded in 2017. In 2017, there was no information to the public due to exceedance of the alert threshold for SO<sub>2</sub> (one-hour average 500  $\mu$ g/m<sup>3</sup>).

**Table 9.1.** Annual statistics for SO<sub>2</sub> in 2017. 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.

10	,	0			0		
Unit: µg/m³	Number of results	Average year	Average winter	Median	98- percentile	Max. Hour	4th highest diurnal mean
Traffic:							
Copenhagen/1103	8197	1.4	1.6	1.1	5	20.8	5.7
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 2017 excluding hours used for calibration.

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

## 9.2 Trends

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



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

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

Particulate matter ( $PM_{10}$  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 2017 was 0.18 ng/m<sup>3</sup> and 0.29 ng/m<sup>3</sup> at the street station on HCAB and the suburban station in Hvidovre, respectively. The concentrations of the measured PAH are generally lower in 2017 than in 2016 at the HCAB station. On the other hand, the average concentrations of some PAH are slightly higher in 2017 in respect to 2016 at the Hvidovre station. Overall, it can be concluded that the target value for benzo[a]pyrene of 1 ng/m<sup>3</sup> was not exceeded in 2017.

Table 10.1 shows the average annual concentrations of the other five PAH's listed in the EU Directive. There are no target values for these compounds.

	НСАВ	Hvidovre
	ng/m³	ng/m³
Benzo[a]pyrene	0.18	0.29
Benzo[a]antracene	0.15	0.19
Benzo[b]fluoranthene	0.25	0.44
Benzo[j+k]fluoranthenes	0.24	0.54
Indeno[1,2,3-cd]pyrene	0.27	0.31
Dibenzo[a,h]anthracene	0.02	0.03

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

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 2013-2017 while the summer concentrations are at the same level in 2013 and 2014 and lower at Hvidovre than at HCAB in 2015 and 2016. Summer average concentrations are the same at the two stations in 2017. This is because the sources of benzon[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 2017.



**Figure 10.2.** Monthly average concentrations of the sum of all analyzed PAH at H.C. Andersens Boulevard and Hvidovre in 2017.

Table 10.2.	Winter, summer and	annual average concentrations	of benzo[a]pyrene for 2013-20	16
	,			

	Hvidovre			НСАВ						
	2013	2014	2015	2016	2017	2013	2014	2015	2016	2017
Winter	0,53	0,73	0,46	0,42	0,49	0,38	0,50	0,44	0,33	0,26
Summer	0,12	0,10	0,06	0,04	0,09	0,11	0,10	0,12	0,08	0,09
Annual	0,34	0,38	0,25	0,23	0,29	0,24	0,29	0,29	0,20	0,18

## 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 four years of data from the suburban 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. A slight increase in the concentration has been observed at Hvidovre in 2017, mainly due to high concentrations observed in January, February and November. The changes between 2016 and 2016 may be due to the natural variations in meteorology from year to year. Longer time series are needed in order to show whether the long-term tendency for a reduction in the concentrations is persistent.



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

## 11. Organic carbon and elemental carbon

Ambient concentrations of particulate Organic Carbon (OC) and Elemental Carbon (EC) are measured at the kerbside station H.C. Andersens Boulevard/1103 in Copenhagen and at the semi-rural station Risø/2090 north of Roskilde. Additionally, EC is measured on H.C. Ørsted Instituttet/1259 (Urban background, Copenhagen) and at Fjeldstedvej/2650 (suburban site, Hvidovre), which is considered to be a hotspot for residential wood burning. PM<sub>2.5</sub> is sampled on two filters in tandem, i.e. quartz-behind-quartz, to correct for positive artifacts from adsorption of volatile and semivolatile organic compounds, which are not particulate material. The filters are analyzed for OC and EC by a thermal/optical method according to the European EUSAAR2 temperature protocol (Cavalli et al., 2010).

#### 11.1 Annual statistics and trends

OC and EC have been measured in PM<sub>2.5</sub> since 2010. During this relatively short period, the annual averages of semi-rural OC has oscillated between 1.1 and 1.8  $\mu$ g/m<sup>3</sup>. Since biogenic sources are expected to account for the majority of the OC in PM<sub>2.5</sub> a constant trend 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 2017 average EC in rural background (0.27  $\mu$ g/m<sup>3</sup>) has decreased by 39% of its 2010 concentration. The kerbside station (1.2  $\mu$ g/m<sup>3</sup>), which is largely impacted by local traffic, has experienced a 51% decrease in EC in the same period. In 2017, Copenhagen urban background (0.33  $\mu$ g/m<sup>3</sup>) and the suburban site in Hvidovre (0.39  $\mu$ g/m<sup>3</sup>) experienced EC concentrations 21 and 44% higher than the semi-rural site. The ratio of EC to total carbon (TC) differs significantly between rural background (0.19) and the kerbside station in Copenhagen (0.37). 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 2017.	The values are based on	I daily averages of Copenha	gen kerbside and semi-rural
background 30 km west of Copenhagen.			

Concentration µg/m <sup>3</sup>	Data capture	OC, average
Copenhagen/1103	91%	2.0
Risø/2090	93%	1.1

**Table 11.2.** Annual statistics for EC in 2017. 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 <sup>3</sup>	Data capture	EC, average
Copenhagen/1103	91%	1.2
Copenhagen/1259	92%	0.33
Risø/2090	93%	0.27
Hvidovre/2650	89%	0.39



**Figure 11.1.** OC, EC and the ratio of EC to total carbon (EC/TC) at kerbside (HCAB), semi-rural background (RISØ), urban background (HCØ) and at a suburban site (HVID).

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

In addition to the measurements of elemental and organic compounds, also measurements of the main inorganic compounds in  $PM_{2.5}$  (ammonium (NH<sub>4</sub><sup>+</sup>), sodium (Na<sup>+</sup>), potassium (K<sup>+</sup>), calcium (Ca<sup>2+</sup>), magnesium (Mg<sup>2+</sup>), chloride (Cl<sup>-</sup>), nitrate (NO<sub>3</sub><sup>-</sup>), sulfate (SO<sub>4</sub><sup>2-</sup>)) have been conducted at the rural measurements station Risø. PM<sub>2.5</sub> is responsible for the majority of the health impacts from air pollution and determination of the chemical constituents are important in connection to identify the composition of PM<sub>2.5</sub>. These measurements are carried out on the basis of the air quality directive from 2008 (EC, 2008). The method is chemical analysis of the daily PM<sub>2.5</sub> particle filters sampled using the LVSs.

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 analyzed constituents. The unknown mass is water attached to the particles, dust (e.g. SiO<sub>2</sub>), heavy metals and other trace constituents.



Figure 12.1. Daily variations of the concentrations of  $PM_{2.5}$  (Na<sup>+</sup>,  $SO_4^{2-}$ ,  $NH_4^+$  and  $Ca^{2+}$ ) at Risø in 2017.

**Table 12.1.** Annual average contributions and relative distribution of the chemical composition of  $PM_{2.5}$  at Risø in 2017. Organic matter (OM) has been estimated from the measured concentrations of OC by multiplication of OC with a factor of 2.1 for the aged OM at Risø, (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.

	μg/m³	Ditribution %
PM <sub>2.5</sub>	9,1	100
Chemical constituents		
Na	0,2	2,3
CI	0,3	3,1
Mg	0,03	0,3
NH <sub>4</sub>	0,9	9,6
NO <sub>3</sub>	2,0	22,0
SO <sub>4</sub>	1,0	11,4
К	0,1	0,9
Са	0,02	0,2
EC	0,3	3,0
OM	2,3	25,4
Unknown mass	2,0	21,8

## 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 outdoor air pollution exposure (WHO, 2014). This high impact of air pollution on human health is the background for inclusion of model calculations of the health risk and associated external economic cost of air pollution in Denmark in the Air Quality Monitoring Program under NOVANA.

The model calculations are carried out with the model system EVA. EVA 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 EVA-system, see chapter 2.3.

The health effects are associated with  $PM_{2.5}$ ,  $NO_2$ , CO,  $SO_2$  and  $O_3$ . Of these,  $PM_{2.5}$  and  $O_3$  are the most extensively used in studies of economic 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  $\mu$ g/m<sup>3</sup> increase in PM<sub>2.5</sub>, which is used in EVA.

## 13.1 Status and trend for health effects

In table 13.1, the number of cases for the different health outcomes due to the total air pollution concentrations calculated using the EVA model system as a mean over the three years 2015-2017 is given. The impact from short-term exposure of  $SO_2$  and  $O_3$  is given as "acute deaths", while the impacts from long-term exposure of  $PM_{2.5}$  is given as Years Of Life Lost (YOLL), which is then applied to calculate the number of "chronic" premature deaths 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 2015-2017 is calculated to around 3,200 cases in Denmark. Health impacts due to exposure of  $NO_2$  are presently not included in the EVA system. However, in the air quality assessment for Europe for 2016 the European Environmental Agency included premature deaths for  $NO_2$  (European Environmental Agency, 2016). In these calculations, about 2% of all predicted premature deaths for Denmark are due to  $NO_2$ .

The main driver for the health impacts is  $PM_{2.5}$ , which in these calculations consist of the total primary emissions of  $PM_{2.5}$ , including mineral dust, fresh and aged black carbon (BC), OC, 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 93% of all premature deaths, O<sub>3</sub> for about 6% and SO<sub>2</sub> for less than 1% (as a mean over the three years 2014-2016).

The risk of premature death resulting from exposure to  $PM_{2.5}$ ,  $O_3$  and  $SO_2$  is rather homogeneously distributed over Denmark. 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 due to the large contribution

to PM<sub>2.5</sub> originating from long-range transport of air pollution mainly from the northern parts of the European continent, giving however a smaller gradient from south to north.

Model calculations with the EVA system have been carried out in order to calculate the development of the health impacts for the period 1990-2017. In figure 13.1, the total number of premature deaths due to  $PM_{2.5}$ ,  $O_3$  and  $SO_2$  in Denmark as annual averages due to the total air pollution, is presented. The total number of premature deaths has decreased from around 5,800 cases/year to around 3,200 cases/year – a reduction of 44% over this period. The variation 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 2017, the number of premature deaths in Europe has decreased to ~525,000, and hence there has also been a significant reduction in health impacts of air pollution on a European level. The decrease in the health impact in Denmark and in Europe as well is due to decrease in the emissions.

The development from 1990 to 2017 of the number of premature deaths in Denmark due to exposure of  $O_3$  and  $SO_2$  is displayed in figure 13.2. The number of premature deaths due to  $SO_2$  has decreased from around 290 cases/year to around 6 cases/year, a decrease of 98%. The long term trend in premature deaths due to  $O_3$  shows a slight increase until 2016 followed by a large decrease from 2016 to 2017 (see below). The increase until 2016 is due to the generally increasing background levels of  $O_3$ , partly due to decreased NO<sub>x</sub> emissions in Denmark and Europe over this period. Lower emissions of NO<sub>x</sub> reduce the degradation of  $O_3$  locally and hence lead to higher  $O_3$  concentrations.

The model calculations show an extraordinary large decrease in the number of premature deaths due to  $O_3$  and  $SO_2$  from 2016 to 2017. For  $O_3$  the decrease is due to the rainy and cold summer in 2017 compared to 2016 that decreased the peak concentrations in  $O_3$  during the summer 2017. This was observed in the measurement results of peak concentrations as well (Figure 4.1 below). The health impacts of ozone are related to the peaks with high  $O_3$  concentrations.

For SO<sub>2</sub> the low number in 2017 is due to the large decrease in the ship emissions as a consequence of the regulation of the sulphur content in ship fuel from 2015 and onwards. Since the most updated available emissions inventories for the international emissions are from 2015. The model calculations are therefore to years behind the actual changes with respect to the emissions. The change in the ship emissions that took place from 2015 and onwards are therefore seen for the first time in this report for 2017. The results from 2015 and 2016 will therefore be recalculated for the next reporting in 2019.

An emission reduction scenario with the DEHM model has been conducted in order to estimate the contribution from emissions in foreign countries to Denmark (in this case all emissions in the Northern Hemisphere (both natural and anthropogenic) and the contribution from anthropogenic emissions in Denmark to the number of premature deaths, calculated by the EVA model system, see table 13.2. The contribution from foreign countries to Denmark is estimated to 2,470 (76% of the total number of cases in Denmark), while the contribution from Danish emissions contributes with 770 premature deaths in Denmark (24%). The contribution from Danish emissions to the number of premature deaths in Europe (excl. Denmark) is estimated to about 2,180 cases/year. The "import" of air pollution related health impacts is therefore about 13% greater than the "export". It is also seen that Danish emissions cause nearly a factor of three more premature deaths in foreign countries as they do in Denmark, due to long-range transport.

**Table 13.1** The number of cases for the different health outcomes in the EVA model system due to the total air pollution concentrations as a mean over the three years 2015-2017 for the whole of Denmark.

Health outcome	Number of cases
Chronic Bronchitis	3.060
Restricted Activity Days	3.130.000
Respiratory Hospital Admissions	159
Cerebrovascular Hospital Admissions	382
Congestive Heart Failure	356
Lung Cancer	469
Bronchodilator Use Children	78.500
Bronchodilator Use Adults	599.000
Cough Children	271.000
Cough Adults	617.000
Lower Respiratory Symptoms Children	105.000
Lower Respiratory Symptoms Adults	222.000
Acute premature deaths (SO2)	8
Acute premature deaths (O3)	109
Chronic YOLL (PM2,5)	33.100
Total no. of premature deaths	3.240
Infant mortality (PM2,5)	3



Figure 13.1. Total number of premature deaths due to  $PM_{2.5}$ ,  $O_3$  and  $SO_2$  in Denmark as annual averages, due to the total air pollution, calculated with the EVA model system.


Figure 13.2. Total number of premature deaths due to  $O_3$  and  $SO_2$  in Denmark as annual avarage, calculated with the EVA 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 by the EVA model system for the year 2017.

Contributions 2017	Number of premature deaths	% of total
Total air pollution in Denmark	3240	100
Foreign contribution to Denmark	2.470	76
Denmark's contribution to Denmark	771	24
Denmark's contribution to Europe incl. Denmark	2951	100
Denmark's contribution to Europe excl. Denmark	2180	74

#### 13.2 Status and trend for external costs of health effects

An external cost occurs when producing or consuming a good or service imposes a cost upon a third party, as e.g. activities leading to increased air pollution concentrations, which results in impacts on health, nature or climate. In the EVA 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 3.5 billion euros (~26 billion DKK) as an average over the three years 2015-2017 using the economic valuation of the individual health outcomes in Brandt et al. (2016) in 2013 prices. The similar number for 2017 is 3.3 billion euros (~25 billion DKK). The trend of 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 cost as an average over the years 1988-1990 is 6.5 billion euros (~49 billion DKK) and has therefore decreased by 40% since then.

The contribution from emissions in foreign countries to Denmark and the contribution from emissions in Denmark to the total health related external costs, calculated by the EVA model system, is given in table 13.3. The contribution from foreign countries to Denmark is estimated to 2.7 billion euros or 20 billion DKK (81% of the total health related external costs in Denmark), while the contribution from Danish emissions contributes with 0.61 billion euros (4.6 billion DKK) in Denmark (19%). The contribution from Danish emissions to the total health related external costs in Europe excluding Denmark is 2.7 billion euros (20 billion DKK).

Table 13.3. Contribution from emissions in fore	ign countries to Denmark and the	e contribution from emissions in D	Denmark to the
total health related external costs, calculated by	the EVA model system for the y	year 2017.	

Contributions 2017	Billion Euro	Billion DKK	% of total
Total air pollution in Denmark	3.3	25	100
Foreign contribution to Denmark	2.7	20	81
Denmark's contribution to Denmark	0.6	4.6	19
Denmark's contribution to Europe incl. Denmark	3.3	25	100
Denmark's contribution to Europe excl. Denmark	2.7	20	81

The Ministry of Finance announced new external costs for a statistical life in August 2017. The updated external cost is 32 million DKK. The external cost of a statistical life in the EVA-system is 15.5 million DKK (2013-prices). The external costs in table 13.3 would be about twice as high if the assumptions of the Ministry of Finance were assumed as premature deaths constitute the majority of the external costs.

### 13.3 Underestimation of health impacts and external costs

The air quality data used in the EVA system (Chapter 2.2.2) is based on a coupling of the two chemistry transport models (DEHM and UBM). PM2.5 is responsible for the majority of the health impact from air pollution in Denmark and as shown in Table 2.3 DEHM/UBM predicts PM2.5 concentrations at urban background stations within -5% to 11% of measured concentrations, and overestimates by 7% for the rural background station after a calibration of the results with a factor of 1.26. However, for this reporting it has not been possible to calibrate the entire EVA-system and it is therefore most likely that the results on health impact and external costs reported her are underestimated with up to about 25%. The underestimation is most likely related to underestimation of the non-exhaust particles (road wear, tyre wear, brake wear and re-suspension) and/or underestimation of certain particle components as secondary organic aerosols (SOA) or the water content. Besides this there are considerable other uncertainties related to the calculations of health impact and external costs among others because newer results indicate that there is an independent health impact due to nitrogen oxides and this impact has not been included in the model yet.

## 14. References

Andersen, Mikael Skou, Lise M. Frohn, Steen S. Jensen, Jytte S. Nielsen, Peter B. Sørensen, Ole Hertel, Jørgen Brandt, and Jesper Christensen, 2004. Sundhedseffekter af luftforurening – beregningspriser. Faglig rapport fra DMU, nr. 507, pp. 85. <u>http://www2.dmu.dk/1\_Viden/2\_Publikationer-/3\_Fagrapporter/rapporter/FR507.pdf</u>

Andersen M. S. and J. Brandt, 2014. NOTAT Miljøøkonomiske beregningspriser for emissioner. Institut for Miljøvidenskab, Aarhus Universitet. Contract for the Danish Ministry for Environment. <u>http://dce.au.dk/fileadmin/dce.au.dk/Udgivelser/Notater\_2014/Miljoeoekonomiske\_beregningspriser\_for\_emissioner.pdf</u>

Anenberg, S. C., A. Belova, J. Brandt, N. Fann, S. Greco, S. Guttikunda, M.-E. Heroux, F. Hurley, M. Krzyzanowski, S. Medina, B. Miller, K. Pandey, J. Roos, R. Van Dingenen, 2015. Survey of ambient air pollution health risk assessment tools. *Risk Analysis*. DOI: 10.1111/risa.12540.

Bach, H., M. S. Andersen, J. B. Illerup, F. Møller, K. Birr-Pedersen, J. Brandt, T. Ellermann, L. M. Frohn, K. M. Hansen, F. Palmgren, J. Seested and M. Winther, 2006. Vurdering af de samfundsøkonomiske konsekvenser af Kommissionens temastrategi om luftforurening, Faglig rapport fra DMU, Nr 586, 2006.

Brandt, J., Silver, J.D., Christensen, J.H., Andersen, M.S., Bønløkke, J.H., Sigsgaard, T., Geels, C., Gross, A., Hansen, A.B., Hansen, K.M., Hedegaard, G.B., Kaas, E. &Frohn, L.M., 2011: Assessment of Health-Cost Externalities of Air Pollution at the National Level using the EVA Model System, CEEH Scientific Report No 3, Centre for Energy, Environment and Health Report series, March 2011, pp. 98. <u>http://www.ceeh.dk/CEEH\_Reports/Report\_3/-CEEH\_Scientific\_Report3.pdf</u>

Berkowicz, R., 2000a: OSPM - A parameterized street pollution model, Environmental Monitoring and Assessment 2000, 65, 323-331. doi: 10.1023/-A:1006448321977.

Berkowicz, R., 2000b: A simple model for urban background pollution, Environmental Monitoring and Assessment 2000, 65, 259-267. doi: 10.1023/-A:1006466025186.

Brandt, J., Christensen, J.H., Frohn, L.M., Berkowicz, R. & Palmgren, F. 2000: The DMU-ATMI THOR Air Pollution Forecast System: System Description, National Environmental Research Institute, Roskilde Denmark 60 pp. -NERI Technical Report No. 321.

Brandt, J., Christensen, J.H., Frohn, L.M. & Berkowicz, R., 2003: Air pollution forecasting from regional to urban street scale – implementation and validation for two cities in Denmark. Physics and Chemistry of the Earth, Vol. 28, pp. 335-344.

Brandt, J., Christensen, J.H. Frohn, L.M. Palmgren, F. Berkowicz R. & Zlatev, Z., 2001: Operational air pollution forecasts from European to local scale. Atmospheric Environment, Vol. 35, Sup. No. 1, pp. S91-S98.

Brandt, J., Silver, J.D., Frohn, L.M., Geels, C., Gross, A., Hansen, A.B., Hansen, K.M., Hedegaard, G.B., Skjøth, C.A., Villadsen, H., Zare, A. & Christensen, J.H., 2012: An integrated model study for Europe and North America using the Danish Eulerian Hemispheric Model with focus on intercontinental transport, Atmospheric Environment, Volume 53, June 2012, pp. 156-176, doi:10.1016/j.atmosenv.2012.01.011.

Brandt, J., J. D. Silver, J. H. Christensen, M. S. Andersen, J. Bønløkke, T. Sigsgaard, C. Geels, A. Gross, A. B. Hansen, K. M. Hansen, G. B. Hedegaard, E. Kaas and L. M. Frohn, 2013a. Contribution from the ten major emission sectors in Europe to the Health-Cost Externalities of Air Pollution using the EVA Model System – an integrated modelling approach. *Atmospheric Chemistry and Physics*, Vol. 13, pp. 7725-7746, 2013. <u>www.atmos-chem-phys.net/13/-7725/2013/</u>, doi:10.5194/acp-13-7725-2013.

Brandt, J., J. D. Silver, J. H. Christensen, M. S. Andersen, J. Bønløkke, T. Sigsgaard, C. Geels, A. Gross, A. B. Hansen, K. M. Hansen, G. B. Hedegaard, E. Kaas and L. M. Frohn, 2013b. Assessment of Past, Present and Future Health-Cost Externalities of Air Pollution in Europe and the contribution from international ship traffic using the EVA Model System. *Atmospheric Chemistry and Physics.* Vol. 13, pp. 7747-7764, 2013. <u>www.atmos-chem-phys.net/13/-7747/2013/</u>. doi:10.5194/acp-13-7747-2013.

Brandt, J., M. S. Andersen, J. Bønløkke, J. H. Christensen, K. M. Hansen, O. Hertel, U. Im, S. S. Jensen, M. Ketzel, O.-K. Nielsen, M. S. Plejdrup, T. Sigsgaard and C. Geels, 2015. High-resolution modelling of health impacts and related external cost from air pollution using the integrated model system EVA. Proceedings from ITM 2015, 34th International Technical Meeting on Air Pollution Modelling and its Application. 4-8 May, 2015, Montpellier, France. pp. 125-128.

Brandt, J., Jensen, S.S., Andersen, M.S., Plejdrup, M.S., Nielsen, O.K. 2016. Helbredseffekter og helbredsomkostninger fra emissionssektorer i Danmark. Aarhus Universitet, DCE – Nationalt Center for Miljø og Energi, 47 s. - Videnskabelig rapport fra DCE - Nationalt Center for Miljø og Energi nr. 182 http://dce2.au.dk/pub/SR182.pdf

Bønløkke, J. H., T. Sigsgaard, J. Brandt, L. M. Frohn, E. M. Flachs, H. Brønnum-Hansen, M.-L. Siggaard-Andersen, 2011. CEEH Scientific Report No. 7a - Description of the CEEH health effect model. Centre for Energy, Environment and Health Report Series, pp. 76, 2011. ISSN 1904-7495.

Christensen, J.H., 1997: The Danish Eulerian Hemispheric Model – a three-dimensional air pollution model used for the Arctic, Atm. Env., 31, 4169–4191.

DCE (2015): http://envs.au.dk/videnudveksling/luft/emissioner/air-pollutants/

EC, 1999: Directive 1999/30/EC of 22 April 1999 relating to limit values for sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead in ambient air. J. Europ. Commun. L163/41.

EC, 2005: Directive 2004/107/EC of the European Parliament and of the Council of 15 December 2004 relating to arsenic, cadmium, mercury, nickel and polycyclic aromatic hydrocarbons in ambient air. Official Journal of the European Union L23/3.

EC, 2008: Directive 2008/50/EC of the European Parliament and of the Council of 15 December 2004 on ambient air quality and cleaner air for Europe: Official Journal of the European Union L152/1.

Ellermann, T., Ketzel, M., Winther, M., & Nordstrøm, C., 2012: Foreløbige resultater for NO<sub>2</sub> i 2011 og vurdering af årsag til de høje koncentrationer på H.C. Andersens Boulevard. Thomas Ellermann, Notat til Miljøstyrelsen 31. januar 2012, 7 s. Available at: <u>http://dce.au.dk/fileadmin/dmu.au.dk/Notat\_NO2\_2011.pdf</u>

Ellermann, T., Wåhlin, P., Nordstrøm, C., & Ketzel, M. 2010: Vejbelægningens indflydelse på partikelforureningen (PM<sub>10</sub>) på stærkt trafikerede gadestrækninger i Danmark. Paper presented at Trafikdage 2010, University of Aalborg; Aalborg, Denmark. August 2010. Paper. Available at: <u>http://www.trafikdage.dk/papers\_2010/380\_ThomasEllermann.pdf</u>

Ellermann, T., Nøjgaard, J.K. & Bossi, R. 2011: Supplerende målinger til luftovervågning under NOVANA – benzen og PAH. Aarhus Universitet, DCE – Nationalt Center for Miljø og Energi. 42 s. – Teknisk rapport fra DCE – Nationalt Center for Miljø og Energi nr. 3

Ellermann, T., Nøjgaard, J.K., Nordstrøm, C., Brandt, J., Christensen, J., Ketzel, M., Jansen, S., Massling, A. & Jensen, S.S. 2013: The Danish Air Quality Monitoring Programme. Annual Summary for 2012. Aarhus University, DCE – Danish Centre for Environment and Energy. 59 pp. Scientific Report from DCE – Danish Centre for Environment and Energy. No. 67.

Ellermann, T., Brandt, J., Jensen, S.S., Hertel, O., Løfstrøm, P., Ketzel, M., Olesen, H.R. & Winther, M. 2014: Undersøgelse af de forøgede koncentrationer af NO<sub>2</sub> på H.C. Andersens Boulevard. Aarhus Universitet, DCE – Nationalt Center for Miljø og Energi, 100 s. - Videnskabelig rapport fra DCE - Nationalt Center for Miljø og Energi nr. 111.

Ellermann, T., Nøjgaard, J.K., Nordstrøm, C., Brandt, J., Christensen, J., Ketzel, M. Jansen, S., Massling, A. & Jensen, S.S. 2015: The Danish Air Quality Monitoring Programme. Annual Summary for 2013. Aarhus University, DCE – Danish Centre for Environment and Energy, 72 pp. Scientific Report from DCE – Danish Centre for Environment and Energy No. 134

EN 12341-2014: Ambient air. Standard gravimetric measurement method for the determination of the  $PM_{10}$  or  $PM_{2,5}$  mass concentration of suspended particulate matter.

EN 12341-1998: Air quality - Determination of the PM 10 fraction of suspended particulate matter - Reference method and field test procedure to demonstrate reference equivalence of measurement methods.

EN 14907-2005: Ambient air quality. Standard gravimetric measurement method for the determination of the  $PM_{2,5}$  mass fraction of suspended particulate matter.

Geels, C., C. Andersson, O. Hänninen, A. S. Lansø, P. Schwarze and J. Brandt, 2015. Future Premature Mortality due to Air Pollution in Europe – Sensitivity to Changes in Climate, Anthropogenic Emissions, Population and Building stock, *Int. J. Environ. Res. Public Health, Int.* 2015, 12, 2837-2869; <u>http://www.mdpi.com/1660-4601/12/3/2837</u>. doi:10.3390/ijerph120302837.

Grell, G.A., Dudhia, J. & Stauffer, D.R., 1995: A description of the fifth-generation Penn State/NCAR Mesoscale Model (MM5). Mesoscale and Microscale Meteorology Division, National Centre for Atmospheric Research, Boulder, Colorado, USA. NCAR Technical Note, NCAR/TN-398+STR, pp. 114.

Harrison, D. 2006: UK Equivalence Programme for Monitoring of Particulate Matter. Report BV/AQ/AD202209/DH/2396 Bureau Veritas London, England (for: Defra <u>http://www.airquality.co.uk/archive/reports/cat05/-</u>0606130952\_UKPMEquivalence.pdf)

Hoek, G., Krishnan, RM., Beelen, R., Peters, A., Ostro, B., Brunekreef, B., et al., 2013. Long-term air pollution exposure and cardio- respiratory mortality: a review. Environ Health 12:43; doi:10.1186/1476-069X-12-43.

Hvidtfeldt UA, Ketzel M, Sørensen M, Hertel O, Khan J, Brandt J, Raaschou-Nielsen O. 2018. Evaluation of the Danish AirGIS air pollution modeling system against measured concentrations of PM2.5, PM10, and black carbon. Environmental Epidemiology, DOI: 10.1097/ee9.000000000000014

Jensen, S.S., Berkowicz, R., Hansen, H. Sten. & Hertel, O. 2001: A Danish decision-support GIS tool for management of urban air quality and human exposures. Transportation Research Part D: Transport and Environment, Volume 6, Issue 4, 2001, pp. 229-241.

Jensen, S.S., Ketzel, M., Nøjgaard, J.K. & Becker, T. 2011: Hvad er effekten af miljøzoner for luftkvaliteten? - Vurdering for København, Frederiksberg, Aarhus, Odense, og Aalborg. Slutrapport. Danmarks Miljøundersøgelser, Aarhus Universitet 110 s. – Faglig rapport nr. 830. Available at: http://www.dmu.dk/Pub/FR830.pdf

Ketzel, M., Jensen, S.S, Brandt, J., Ellermann, T., Olesen, H.R., Berkowicz, R. & Hertel, O. 2013: Evaluation of the Street Pollution Model OSPM for Measurements at 12 Streets Stations Using a Newly Developed and Freely Available Evaluation Tool. J Civil Environ Eng, S1:004. doi:10.4172/2165-784X.S1-004

Khan, J.; Konstantinos Kakosimos; Ole Raaschou-Nielsen; Jørgen Brandt; Steen S Jensen; Thomas Ellermann; Matthias Ketzel, 2018: Development and Performance Evaluation of New AirGIS - A GIS Based Air Pollution and Human Exposure Modelling System. Submitted to Atmospheric Environment.

Miljø- og fødevareministeriet, 2016: Bekendtgørelse om vurdering og styring af luftkvaliteten. Bekendtgørelse nr. 1233 af 30.09.2016 (In Danish). København, Danmark.

Ottosen, T-B., Kakosimos, K. E., Johansson, C., Hertel, O., Brandt, J., Skov, H., Berkowicz, R., Ellermann, T., Jensen, S. S. & Ketzel, M. 2015: Analysis of the impact of inhomogeneous emissions in a semi-parameterized street canyon model. I : Geoscientific Model Development Discussions. 8, 3231–3245, 2015. doi:10.5194/gmd-8-3231-2015.

Plejdrup, M.S. & Gyldenkærne, S., 2011: Spatial distribution of emissions to air – the SPREAD model. National Environmental Research Institute, Aarhus University, Denmark. 72 pp. – NERI Technical Report no. FR823. Available at: http://www.dmu.dk/Pub/FR823.pdf

Turpin, B.J. & Lim, H.-J., 2001: Species Contributions to PM2.5 Mass Concentrations: Revisiting Common Assumptions for Estimating Organic Mass, Aerosol Science and Technology, 35: 1, 602 – 610, First published on: 30 November 2010 (iFirst). DOI: 0.1080/02786820119445URL. Available at: http://dx.doi.org/10.1080/02786820119445

WHO, 2000: Air Quality Guidelines for Europe, Second Edition, WHO Regional Publications, European Series, No. 91, Copenhagen 2000. Availabel at: <u>http://www.euro.who.int/air</u>

WHO, 2014: <u>http://www.who.int/mediacentre/news/releases/2014/air-pollution/en/</u>

# Appendix 1

#### Relpacement of the station at H.C. Andersens Boulevard

On 3 October 2016 the station at H. C. Andersen 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 0.1 and 0.2). 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.



Inner traffic lane on H.C. Andersens Boulevard. Direction towards Rådhuspladsen

Figure 0.1. Sketch of the old and new location of the measurement station at H.C. Andersens Boulevard.



Figure 0.2. 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<sub>2</sub> in the urban atmosphere is produced by oxidation of NO by O<sub>3</sub>. The reaction will take place immediately, if sufficient O<sub>3</sub> is present. O<sub>3</sub> is often the limiting component for a complete oxidation in the street canyons, but practically all NO is oxidised at the urban background and rural stations. Within a few hours the NO<sub>2</sub> is further oxidised to nitrate and/or nitric acid, which may cause acid precipitation and eutrophication. NO<sub>2</sub> is a toxic gas, which may cause respiratory problems. There are limit values for the allowed concentration of NO<sub>2</sub> in the atmosphere.

 $O_3$  is formed by photochemical reactions (i.e. by the influence of sunlight) between NO<sub>x</sub> and VOCs. The VOCs can be of natural and anthropogenic origin. The major part of the O<sub>3</sub> measured in Denmark originates from sources outside the country. Usually the highest concentrations are found at rural and urban background sites. O<sub>3</sub> is removed by NO at street level. O<sub>3</sub> is a toxic gas, which may cause respiratory problems and damage on crops and forests. There are so-called target values for the concentration of O<sub>3</sub> in the atmosphere.

The main source of CO in urban air is petrol-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<sub>10</sub> is collected by high volume sampling (HVS) at a flow rate of 0.5 m<sup>3</sup> min<sup>-1</sup> over a period of 24 hours for an average total volume of 700 m<sup>3</sup>. The filters are kept frozen until analysis. Weekly based PAH concentrations are obtained by analysis of pooled fractions of daily collected samples. For each day 4 x 1.5 cm<sup>2</sup> are taken from the filter and the fractions from the whole week are pooled and extracted. The pooled filters are extracted with dichloromethane and cleaned up on silica. Before extraction, the filters are spiked with deuterium-labeled PAH. Analysis of the extracts is

carried out by gas chromatography-mass spectrometry (GC-MS). Concentrations of individual PAHs in samples are corrected for recovery of a deuterium-labelled PAH standard with the closest molecular weight. A total of 18 PAHs are 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  $\beta$ -absorption in the filter with sampled dust. This method is considered to be equivalent to the reference method (EN 12341:1999 and EN14907:2005).
- The LVS method: The particles are collected on filters for 24-hour intervals by a low volume sampler (LVS). The mass on the filters is subsequently determined in the laboratory by gravimetric measurements of the dust. This method is the current reference method for the determination of the PM<sub>10</sub> or PM<sub>2.5</sub> mass concentration of suspended particulate matter in ambient air (EN 12341: 2014, into which the previous standards for PM<sub>10</sub>, EN 12341: 1998, and for PM<sub>2.5</sub>, EN 14907:2005, have been merged).
- The TEOM method: The particles are continuously collected on a "tapered oscillating microbalance" (TEOM) and heated to 50°C. During heating volatile compounds may evaporate. The loss will be most pronounced for "secondary aerosols" containing ammonium nitrate. PM results are given with a time resolution as ½-hourly averages.

There are a number of different heavy metals (HM) in the atmosphere. They are emitted from e.g. coal and oil-fired power plants, waste incinerators and industries. 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 calibration of all final model results was necessary since a comparison with measurements showed a significant underestimation. For this comparison we used only data for 2017. 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 was done by applying a factor to all final modelling results, 1.26 for  $PM_{2.5}$  and 1.46 for  $PM_{10}$ . The same calibration factors were estimated and applied by Hvidfeldt et al. (2018) and Khan et al. (2018). 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 2017 for all available stations: street, urban background and rural.

Abbriviation	Identification number of measurement station
	Street:
HCAB	Copenhagen/HCAB/1103
JGTV	Copenhagen/Jagtvej/1257
AARHstr	Aarhus/6153
ODGR	Odense/9156
	Urban Background:
HCOE	Copenhagen/1259
AARbg2	Aarhus/6160
ODbg	Odense/9159
AALbg2	Aalborg/8159
HVID	Hvidovre/2650
	Rural:
RISOE	Risø/2090
FOEL	Føllesbjerg/9055
ANHT	Anholt/6001
ULBG	Ulborg/7005

The abbreviation for the different measuring stations and their corresponding name and identification number is shown in the table below.

In Figure 0.3 the correlation between modelled and observed annual levels of  $NO_2$  is shown for all stations for 2017. There are 13 observations and the average observed concentration is 15.1 µg/m<sup>3</sup> and the modelled 15.0 µg/m<sup>3</sup>. The Pearson and Spearman correlations (R p/s) are very high (0.99 and 0.98) and the Normalized Mean Bias (NMB%) is very low (-0.67%).



Figure 0.3. Correlation between modelled and observed annual levels of  $NO_2$  for all stations for 2017.

In Figure 0.4 the correlation between modelled and observed annual levels of  $PM_{2.5}$  is shown for all stations for 2017. There are 8 observations and the average observed concentration is 10.7  $\mu$ g/m<sup>3</sup> and the modelled 10.9  $\mu$ g/m<sup>3</sup>. The Pearson and Spearman correlations (R p/s) are very high (0.96 and 0.98) and the Normalized Mean Bias (NMB%) is low (2.2%).



Figure 0.4. Correlation between modelled and observed annual levels of  $PM_{2.5}$  for all stations for 2017.

In Figure 0.5 the correlation between modelled and observed annual levels of  $PM_{10}$  is shown for all stations for 2017. There are 6 observations and the average observed concentration is 19.8 µg/m<sup>3</sup> and the modelled 20.4 µg/m<sup>3</sup>. The Pearson and Spearman correlations (R p/s) are very high (0.95 and 0.94) and the Normalized Mean Bias (NMB%) is low (3.3%).



Figure 0.5. Correlation between modelled and observed annual levels of  $PM_{10}$  for all stations for 2017.

# THE DANISH AIR QUALITY MONITORING PROGRAMME

Annual Summary for 2017

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 2017 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 2017, no exceedeances of the NO<sub>2</sub> EU limit value for the annual average were observed whereas the limit value was exceeded in 2016 at one street station (H.C. Andersens Boulevard) in Copenhagen, while NO<sub>2</sub> levels in Odense, Aarhus and Aalborg were below the limit value. Model calculations also indicate no exceedances of the NO<sub>2</sub> limit value at a selection of streets in Copenhagen and Aalborg. Annual averages of  $PM_{10}$  and  $PM_{2.5}$  were below limit values at all stations and the average exposure indicator (PM<sub>2.5</sub> in urban background) has decreased with about 30 % since 2010. The concentrations for most pollutants have been decreasing during the last decades.