THE PARTICLE PROJECT 2020

Scientific Report from DCE – Danish Centre for Environment and Energy No. 450 2021

AARHUS UNIVERSITY
DCE – DANISH CENTRE FOR ENVIRONMENT AND ENERGY

THE PARTICLE PROJECT 2020

Scientific Report from DCE – Danish Centre for Environment and Energy No. 450 2021

Thomas Ellermann
Andreas Massling
Rossana Bossi
Jacob Klenø Nøjgaard

Aarhus University, Department of Environmental Science

AARHUS UNIVERSITY
DCE – DANISH CENTRE FOR ENVIRONMENT AND ENERGY

Data sheet

Series title and no.:	Scientific Report from DCE – Danish Centre for Environment and Energy No. 450
Category:	Scientific advisory report
Title:	The Particle Project 2020
Authors:	Thomas Ellermann, Andreas Massling, Rossana Bossi & Jacob Klenø Nøjgaard
Institution:	Aarhus University, DCE / Department of Environmental Science
Publisher: URL:	Aarhus University, DCE – Danish Centre for Environment and Energy © http://dce.au.dk/en
Year of publication: Editing completed:	July 2021 July 2021
Referee: Quality assurance, DCE:	Lise Lotte Sørensen Vibeke Vestergaard Nielsen
External comments:	The comments can be found here: http://dce2.au.dk/pub/komm/SR450_komm.pdf
Financial support:	Ministry of the Environment
Please cite as:	Ellermann, T., Massling, A., Bossi, R., & Nøjgaard, J.K., T., 2021. The Particle Project 2020. Aarhus University, DCE – Danish Centre for Environment and Energy, xx pp. Scientific Report from DCE – Danish Centre for Environment and Energy No. 450. http://dce2.au.dk/pub/SR450.pdf

Abstract: The Particle Project 2020 continues the measurements of the long term trends of particle number concentrations and size distributions for submicron particles as well as the concentrations of elemental carbon in the ambient fine particle fraction ($PM_{2.5}$) at the Copenhagen Urban background measurement station (HCØ). The results from the measurements at urban background are compared to results from urban curb side, suburban and rural locations. The results show decreasing concentrations for both particle number concentrations and elemental carbon, which are mainly due to decreasing emissions on national as well as international level. The report also presents results from an analysis of the impact of the Covid-19 restrictions on the particle number concentrations and concentrations of elemental carbon.

Keywords:	Particulate air pollution, $PM_{2.5}$, particle number and size distribution, elemental carbon.
Layout: Front page photo:	Majbritt Ulrich Thomas Ellermann
ISBN: ISSN (electronic):	978-87-7156-606-2 2245-0203
Number of pages:	26
Internet version:	The report is available in electronic format (pdf) at http://dce2.au.dk/pub/SR450.pdf

Contents

Abbreviations and definitions

AARHG	Urban curbside measurement site in Aarhus.
BC	Black carbon, which is roughly equivalent to elemental carbon.
°C	Degrees Celsius.
DMPS	Differential Mobility Particle Sizer.
Dp	Particle diameter.
EC	Elemental carbon, roughly equivalent to black carbon (BC).
HCAB	Urban curbside measurement site in Copenhagen.
HCØ	Urban background measurement site at H.C. Ørsted Institute in Copenhagen.
HVID	Suburban measurement site at Fjelstedvej in Hvidovre.
LVS	Low Volume Sampler for atmospheric particles.
OC	Organic carbon, only the mass of carbon itself.
PM	Particle mass in ambient air.
$PM_{2.5}$	Particles less than 2.5 micrometers in diameter, i.e. fine particles.
PM_{10}	Particles less than 10 micrometers in diameter.
RISØ	Rural measurement site at Risø, North of Roskilde.
RWC	Residential Wood Combustion.
SMPS	Scanning Mobility Particle Sizer.
SOA	Secondary Organic Aerosols, i.e. particulate species formed during atmospheric oxidation of VOCs, including VOCs from residential wood combustion.
TEOM	Tapered Element Oscillating Microbalance.
VOC	Volatile Organic Compounds.

.

Sammenfatning

I *The Particle Project 2020* rapporteres resultater af målinger af partikelstørrelsesfordelingen fra 11 nm op til 478/550 nm, partikelantal samt elementært kulstof i Københavns bybaggrund. Disse målinger er et supplement til luftovervågningsprogrammet under NOVANA. Resultaterne for bybaggrund sammenholdes med tilsvarende resultater fra en landligt baseret station nord for Roskilde (RISØ), en station i den Københavnske forstadsby Hvidovre (HVID), samt fra en station i en trafikeret gade i København (HCAB). Herudover rapporteres udviklingstendenser for $PM_{2,5}$ og PM_{10} fra målinger med høj tidsopløsning. $PM_{2,5}$ og PM_{10} angiver koncentrationen af partikler med diameter mindre end hhv. 2,5 og 10 µm.

Langtransporterede partikler og deres gasformige forstadier bidrager til partikelantal for partikler med en diameter mindre end 1 µm. Langtransporterede partikler udgør den største andel på de landlige stationer og en mindre andel på stationerne i bybaggrund og forstad, hvor lokale kilder spiller en relativt større rolle. Mindst er andelen af langtransporterede partikler målt på gadestationerne, hvor den største andel udgør bidraget fra lokal trafik. Det relativt største bidrag fra langtransporterede partikler ser man i landlig baggrund på målestationen RISØ. Det næststørste relative bidrag fra langtransporterede partikler finder man i bybaggrund på HCØ, dernæst kommer HVID (forstadsby), mens dette bidrag udgør den mindste relative andel på HCAB (trafikeret gade).

Set over lang tid er der målt et fald i partikelantal ved alle målestationerne, men der er forskel i udviklingstendensen for de forskellige partikelstørrelsesfraktioner og målestationer. Der er også forskel på dataseriernes længde, hvor målingerne på målestationerne HCØ og HCAB blev begyndt i 2002, RISØ i 2005 og HVID først i 2015. Dataserien for forstadsmålestationen (HVID) er for kort til at kunne vurdere udviklingstendenserne. Grundet tekniske problemer med udstyret er der endvidere huller i tidsserierne fra 2016-2019, som varierer fra målestation til målestation. Disse huller gælder for den mindste partikelfraktion (11-41 nm), hvilket også medfører huller i tidsserien for det fulde måleområde (11 – 478/550nm).

11 og 41 nm: For de seneste 10 til 15 år ses en tendens til, at partikelantallet ligger på et relativt stabilt niveau ved bybaggrundsmålestationen (HCØ) og landbaggrundsmålestationen (RISØ). Ved gademålestationen (HCAB) ses et betydeligt fald i partikelantallet, som dog også flader ud inden for de seneste år. Ved gademålestationen kommer den største andel af partiklerne i denne fraktion fra udstødning og det store fald hænger sammen med den øgede brug af partikelfiltre på køretøjerne.

41 og 110 nm og 110 og 478/550 nm: Ved bybaggrundsmålestationen (HCØ), landbaggrundsmålestationen (RISØ) og gademålestationen (HCAB) måles et generelt fald i partikelantal for partikler med diameter mellem 41 og 110 nm og partikler mellem 110 og 478/550 nm i løbet af de seneste 15-20 år. Ved bybaggrundsmålestationen (HCØ) og landbaggrundsmålestationen (RISØ) har der dog været et nogenlunde stabilt niveau gennem de seneste fem år. I 2018 blev der målt en noget højere værdi sammenlignet med de omkringliggende år, hvilket formentligt skyldes at 2018 var et usædvanligt tørt år, hvor sommernedbøren var 40% lavere end for de tilsvarende perioder i 2017 og

2019. Ved gademålestationen (HCAB) måles fortsat et fald i partikelantal gennem de seneste fem år, hvilket skyldes, at det løbende fald i udledningerne som følge forbedring af vognparken slår tydeligere igennem på gademålestationen end ved baggrundsmålestationerne.

11 – 478/550nm: Når partikelantallet for det fulde måleområde vurderes, ses der et mindre fald i partikelantallet for bybaggrundsmålestationen (HCØ) og landbaggrundsmålestationen (RISØ), mens der for gademålestationen (HCAB) ses et betydeligt fald i partikelantallet. – navnligt i den første del af måleserien (2002-2012), hvorefter faldet flader ud.

Årsmiddelkoncentrationer for EC blev i 2020 målt til 0,27 µg/m^3 ved bybaggrundsmålestationen (HCØ), hvilket er omkring 17% lavere end målt i 2019. Tilsvarende fald blev målt ved den landlige målestation RISØ (16%) og ved forstadsmålestationen i Hvidovre (15%), mens faldet var markant højere ved gademålestationen HCAB (29%).

Siden 2015 er koncentrationerne af EC faldet med omkring 29% ved baggrundsmålestationen HCØ. Den vigtigste årsag til faldet i koncentrationerne er de generelle reduktioner i udledningerne af EC på både nationalt og internationalt plan. Ved gademålestationen HCAB er der blevet målt et stort fald på omkring 67% (1,7 µg/m^3) i perioden fra 2010 til 2020, hvilket hovedsageligt skyldes fald i udledningerne fra vejtrafik.

EC er faldet med 50% (0,23 µg/m^3) ved den landlige målestation RISØ i perioden fra 2010 til 2020. Ved forstadsmålestationen HVID er der målt et fald på omkring 25% (0,1 µg/m^3) siden 2016. Faldet i koncentrationerne af EC har siden 2016 forløbet meget ensartet ved bybaggrundsmålestationen, landmålestationen og målestationen i forstad, hvilket indikerer at det er de generelle reduktioner af udledningerne på nationalt og internationalt niveau, som er årsag til hovedparten af faldet i koncentrationerne.

Analysen af den tidslige variation i koncentrationer af EC i 2020 viste, at Covid-19-restriktionerne kun havde lille indflydelse på ændringerne i koncentrationerne fra 2019 til 2020 på bybaggrundsmålestationen (HCØ), hvilket ligeledes var tilfældet ved den landlige målestation (RISØ) og ved forstadsmålestationen i Hvidovre (HVID). Ved disse tre målestationer skyldtes faldet fra 2019 til 2020 hovedsageligt det fortsatte fald i udledningerne nationalt og internationalt og de naturlige variationer i de meteorologiske forhold fra år til år, hvor blandt andet den meget våde februar bidrog til lavere koncentrationer.

Ved gademålestationen (HCAB) blev der fra 2019 til 2020 målt et fald på næsten 50% i koncentrationen af EC i den første periode med omfattende Covid-19-restriktioner. Dette store fald viser, at Covid-19-restriktionerne resulterede i lavere koncentrationer i overensstemmelse med at reduktionerne gav et stort fald i vejtrafikken i den tidlige del af foråret 2020. På den anden side står det dog også klart, at reduktioner i udledningerne som følge af den løbende forbedring af vognparken og de meteorologiske variationer er afgørende faktorer bag de observerede fald fra 2019 til 2020.

Analysen af de tidslige variationer i partikelantal indikerede, at effekten af Covid-19-restriktionerne ikke var særlig stor ved gademålestationen (HCAB) eller at de komplekse fysiske og kemiske processer i luften, som har afgørende betydning på partikelantallets størrelse har maskeret indflydelsen af Covid-

19-restriktionerne på partikelantallet. Af tekniske årsager var der desværre ikke tilstrækkelig data for de første måneder af 2019 til, at der kunne laves en tilsvarende analyse for målestationerne i bybaggrund (HCØ), landområder (RISØ) og forstad (HVID).

Summary

The *Particle Project 2020* reports time series from measurements of particle number concentrations of submicrometer particles and Elemental Carbon (EC) in fine particles with diameter smaller than 2.5 µm ($PM_{2.5}$) in urban background in Copenhagen. These measurements are carried out as a supplement to the Danish air quality monitoring program under NOVANA. Trends in urban background (HCØ) are compared to rural location (RISØ), suburban location (HVID) and urban curbside (HCAB).

Regional and long-range transported aerosols and precursors hereof contribute to the particle number concentration in the submicrometer size range. The highest relative contribution from long-range transported particles is found at rural background locations while the other locations are more influenced by local sources. For these reasons, long-range transported particles make up a relative smaller fraction at the suburban site (HVID) and the urban background site (HCØ) and an even smaller fraction at the urban curbside station (HCAB), where the highest particle number concentrations are measured due to the significant contributions from local traffic.

A trend of decreasing particle number concentrations is observed at all stations when considering time periods of the order of a decade. At the suburban site (HVID) only measurements covering about half a decade are available. A general decreasing trend is also observed for the rural background (RISØ), the urban background (HCØ) and the urban curbside (HCAB) when considering the measurements of particles in the range 41 - 110 nm and 110 – 478/550 nm of the last 15 – 20 years. However, the measurements show for these fractions very stable numbers for the rural location (RISØ) and the urban background station (HCØ) in the last five years with an increased value in 2018. This might be due to an unusual dry year in 2018 where summer precipitation was about 40% lower than summer precipitation in 2017 and 2019, thus leading to reduced wet deposition. In contrast the decreasing trend is still observed at the urban curbside station (HCAB) for these two fractions in the last five years where also local contributions from traffic are expected.

Data for particle numbers in the size range 11 – 41 nm do show a gap due to instrumental problems. The general tendency shows stable particle numbers for this small size fraction in the last ten to fifteen years at the rural background (RISØ) and the urban background (HCØ). A decreasing trend can be observed at the urban curbside (HCAB) which more and more levels out in the last years.

The total number of particles for the full size range (11 – 478/550nm) shows a small decrease at the rural background (RISØ) and the urban background (HCØ) while the decrease is much more pronounced at the curbside station (HCAB) especially between 2002 and the following ten years. It is still not possible to make a firm conclusion for the suburban site (HVID) due to the limited length of the data series.

The annual mean concentration of EC at the urban background station in Copenhagen was 0.27 µg/m^3 in 2020, which is about 17% lower than measured in 2019. Similar changes were observed at the rural (16%) and suburban (15%)

stations while the decrease was higher at the urban curbside station HCAB (29%).

Since 2015, EC has decreased with about 29% (0.1 µg/m^3) at the background station HCØ. The main reason for the decrease in the concentrations at HCØ is the general reductions in the emissions both at national and international level.

EC has decreased by 50% (0.23 µg/m^3) at RISØ over the time period 2010 to 2019. At the suburban site (HVID) there has been a reduction since 2016 of about 25% (0.1 µg/m^3). Since 2016 the decreasing trends have been quite similar at urban background, suburban and rural sites indicating that the general reductions in emissions on national and international level can explain the major part of the decreasing trends.

At HCAB there has been a pronounced reduction of about 67% (1.7 µg/m^3) in the annual average concentrations over the time period 2010 to 2020, predominantly due to a reduction in local road traffic emissions.

Analysis of the temporal variation of EC in 2020 showed that the Covid-19 restrictions in 2020 only had minor impact on the concentration change from 2019 to 2020 at the urban background station (HCØ). The same was concluded for the rural station (RISØ) and suburban station (HVID). At these three stations the main reasons for the change between 2019 and 2020 is the continuing decreases in the emissions nationally and internationally and the natural variations in the meteorological conditions from year to year.

At the urban curbside station (HCAB) a nearly 50% reduction in the concentrations of EC were measured for the first period with widespread Covid-19 restrictions in 2020 when compared to 2019. This large decrease shows that the Covid-19 restrictions had impact on the concentrations of EC as expected from the large reductions in traffic in connection to the widespread Covid-19 restrictions in early spring. However, it is also evident that the reductions in emissions due to the continuing improvements of the vehicle fleet and variations in meteorology are important for the annual reduction in the concentrations of EC at the urban curbside station.

The temporal analysis of the particle number concentrations at the urban curbside station (HCAB) indicated that the impact of Covid-19 was not very large for the particle number concentration or that the complex physical and chemical processes determining the particle number concentrations counteracted the impact from the Covid-19 restrictions. Unfortunately, the data coverage for the first months in 2019 is not sufficient to carry out a similar temporal analysis for the urban background, rural and suburban stations.

1 Introduction

DCE – Danish Centre for Environment and Energy has since 2001 carried out particle research in Denmark through a series of projects funded by the Danish Environmental Protection Agency (to an example see Nøjgaard et al., 2020). These projects have enabled DCE to obtain fundamental new knowledge on the particulate air pollution in Denmark including knowledge on the contributions to particulate air pollution from wood burning and traffic. Besides this, the projects have been the basis for establishment of unique time series on the air pollution with elemental carbon and sub-micrometer particles focusing on particle number and size distribution. In popular terms these air pollution components are often termed soot and ultrafine particles. These air pollution components are associated with health effects and the knowledge established through the particle projects has been used in connection to studies of the health impact of airborne particle pollution in Denmark.

This report presents the results from the particle project in 2020 where the main aim has been to continue the time series based on measurements of elemental carbon, particle size distribution and particle number concentration at the urban background station at H.C. Ørsted Institute in Copenhagen. These time series are important because they are fundamental to research concerned with the impact of air pollution on health. The measurements in urban background are especially important since the level of air pollution in urban background has been regarded as the best available measured proxy for the exposure of citizens to air pollution. Moreover, these time series can be used to evaluate the impact of emission regulations on the levels of these air pollution components in urban areas.

The main focus of the project are the measurements in urban background. However, in order to set these results into context, they will throughout the report be compared to the results from the urban curbside station at H.C. Andersens Boulevard in Copenhagen, the suburban station in Hvidovre and the rural station at Risø north of Roskilde.

The report presents also results from analysis of the impact of the Covid-19 restrictions on the levels of elemental carbon and particle number concentrations in Denmark.

2 Measurements of particle number concentration and particle size distribution

2.1 Particle number

Custom built DMPS instruments (Differential Mobility Particle Sizer) have been used from 2001/2002 and onwards during several Particle Projects to measure particle number size distributions in the submicrometer size regime. Particle number size distributions of diameters 6 - 700 nm were measured at the rural station RISØ (previously Lille Valby), urban background HCØ and urban curbside HCAB. From 2017 and onwards, the instruments at HCAB and RISØ were replaced with commercial instruments delivered by TSI (Model 3938) (Table 2.1). These are SMPS instruments (Scanning Mobility Particle Sizer) and measure in the size range 11 - 478 nm. At HCØ, one of the original DMPS instruments was still in use in 2017 and 2018, but was exchanged in the beginning of 2019 with a new SMPS system (Model 3938). From ultimo 2015 and onwards, an additional SMPS instrument delivered by TSI (Model 3938) has been operated at the suburban station HVID in Hvidovre. The new instruments were connected to new inlet systems, which unfortunately introduced losses, which has turned out to affect the general uncertainty of particle number concentrations (Nøjgaard et al., 2018). In addition, some problems occurred with some of the DMAs (Differential Mobility Analyzer), which are a part of the new SMPS systems. During 2019, these inlets and DMAs have been exchanged, implying that data from smaller size regimes can be used from February 2020 and onwards with good data coverage (Table 2.2).

As discussed in *The particle Project 2017-2018 (Nøjgaard et al., 2018)*, the slightly different measurement ranges between the new and the old instruments have implications for data comparison. The size range 11 - 550 nm will be discussed for the old DMPS instruments, whereas the size range 11 - 478 nm will be discussed for the new SMPS instruments. Only in this way a comparison of historical and new data is possible, and moreover, particles in the range 478 - 550 have very little impact on the particle number. However, the problems with particles in the range 11 - 41 nm, which were observed based on inlets and DMAs and discussed in the former Particle Project, were not solved until primo 2020. Hence, data reported in the years 2017 to 2019 (RISØ, HCAB), 2019 (HCØ) and 2015 to 2019 (HVID) is based on size ranges from 41 to 478 nm and particle numbers within this size range.

Table 2.1. Operation of instruments (old and new) at the four measurement stations during the years 2002 to 2020.

	Rural background (RISØ)	**Suburban background** (HVID)	**Urban background** (HCØ)	**Urban curbside** (HCAB)
Old instrument 6 nm – 550 nm	2005-2016	-	2002-2018	2002-2016
New instrument 11 nm – 478 nm	2017-2020	2015-2020	2019-2020	2017-2020

Table 2.2. Data coverage for SMPS measurements at the rural location (RISØ), suburban location (HVID), urban background location (HCØ) and urban curbside (HCAB).

	Rural (RISØ)	Suburban (HVID)	Urban background (HCØ)	Urban curbside (HCAB)
2020	75%	77%	90%	91%

Particle number size distributions are obtained at a time resolution of 30 minutes. In Figure 2.1, annual mean particle number size distributions at RISØ, HVID, HCØ and HCAB are shown for the year 2020 in the size range from 11 – 478 nm (new instruments) and for the year 2017 in the size range from 41 – 478 nm (new instruments: RISØ, HVID, HCAB) and from 11 – 550 nm (old instrument: HCØ). The data for 2020 covers only 11 month average for the year including the months February to December (January is left out as the problems in the size range between 11 to 41 nm were solved from February and ongoing). In Figure 2.1 the derivative dN/d(log Dp) is plotted against the logarithm of the particle diameter Dp, which has the obvious advantage that the area under the curve corresponds to the particle number. In that way, it is clearly visualized that e.g. the number of particles at HCAB is much higher compared to the other three stations. This difference is predominantly pronounced for the smallest fraction of the particles below about 100 nm which are also called ultrafine particles and where emissions from traffic at the curbside station strongly impact the particle number size distribution and thus also the particle number in the submicrometer size range.

At the rural location (RISØ), particle numbers above 41 nm show only a slight decrease in 2020 compared to 2017. Similarly, this is observed for the suburban background station HVID for particle numbers above 41 nm and the urban background station HCØ with view on the full size range which is available for HCØ for both years, 2020 and 2017. A strong decrease in particle number above 41 nm is observed at the curbside station HCAB which is heavily influenced by emissions from local traffic even at this size range. The largest particle number, however, at highly trafficked curbside measurement stations, is found below 100 nm in diameter and especially below 30 – 50 nm.

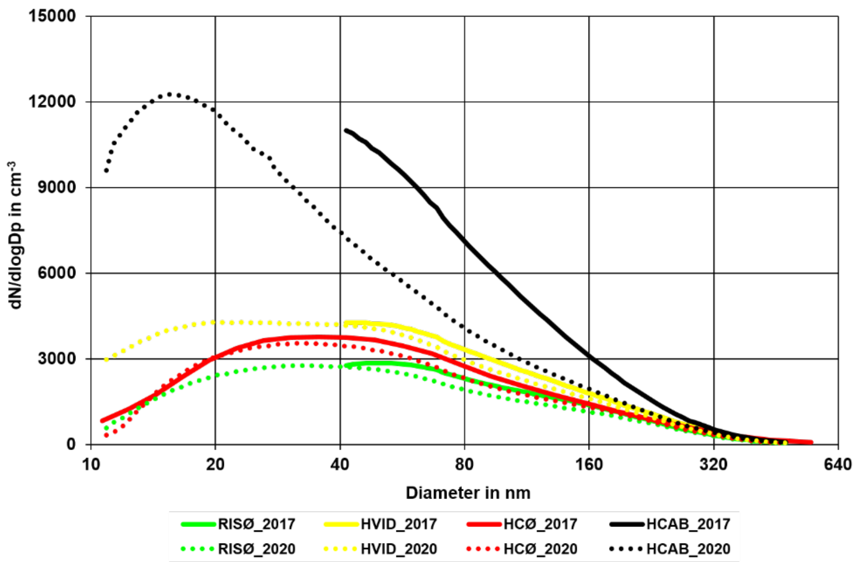


Figure 2.1. Annual means of particle number size distributions at the rural location (RISØ), suburban location (HVID), urban background location (HCØ) and urban curbside (HCAB) during 2020 compared to 2017. Note the logarithmic x-axis.

It has to be stated that in general particle numbers measured below 20 nm using typical SMPS instruments that cover large size ranges of the submicrometer size range are highly uncertain as calibration protocols in this range are challenging and require extremely sophisticated laboratory set ups. Nevertheless, for higher size ranges SMPS instruments as used in our study give quite good results regarding general uncertainties, which can be estimated smaller than about 10 – 15 % in number. It should also be noted, that year to year differences in size distributions can also be caused by meteorological variations and for this reason trends on particle numbers are discussed in the following section based on selected size regimes and as decadal trends. Data have to be investigated over many years to draw conclusions and analyse real trends in time series.

Particle numbers generally decrease with distance to major sources of aerosol particles. This is a result of polluted air being diluted with cleaner air that in turn leads to a general decrease in particle numbers. These mixing processes go along with aging processes as condensation, evaporation, coagulation and cloud processing, leading to changes in the general particle population. A result of this particle aging is an increase in the mean particle diameter with distance to the major sources (Nøjgaard et al., 2015). It is expected, that local sources in urban areas mainly contribute to particle number concentrations, and especially to the sizes around and smaller than 100 nm in diameter. Particle number size distributions close to major sources are characterized by smaller mean diameters in general, and this is seen from Figure 2.1, when comparing the particle number size distribution at the urban curbside (HCAB) to the other stations.

The annual averages at HVID in 2020 exceeded those at HCØ and RISØ for the full submicrometer size range, which reflects larger emissions from wood burning in suburban areas during wintertime, and the location of a number of highly busy roads and highways close to the suburban station.

At RISØ, HVID, and HCØ, the profile of the particle number size distributions looks rather similar in 2017 for particles larger than 41 nm and in 2020 for the full size range. Only at HVID higher number concentrations below 41 nm are observed. This can be due to a combination of larger uncertainties and variation in measured sizes below 20 nm in between systems plus the location of HVID and its special surroundings as discussed above. As expected, largest numbers of small particles are found at the urban curbside (HCAB) in 2020 compared to all three other sites. This large number observed at HCAB is mainly a result of ultrafine particles originating from vehicle exhaust emissions in busy roads and was also found in previous reports (Nøjgaard et al., 2017).

Substantial decreases are observed at HCAB between 2020 and 2017 also for the number of particles larger than 41nm in diameter. Changes in traffic density and especially technological development reducing pollutant emissions can be one of the major reasons behind this finding. In addition the Covid-19 restrictions could explain part of this decrease (see Section 2.4).

2.2 Particle number fractions

For a more detailed analysis, particle number concentrations were determined in specific size regimes, in this case particles with diameters of Dp = 11 – 41 nm (small size regime), Dp = 41 – 110 nm (medium size regime), and Dp = 110

– 550 nm (large size regime) for the old instruments operated from 2002 to 2016 (HCAB), 2005 to 2016 (RISØ), and 2002 to 2018 (HCØ), and likewise particles with diameters Dp = 11 – 41 nm (small size regime), Dp = 41 – 110 nm (medium size regime), and Dp = 110 – 478 nm (large size regime) for the new instruments operated from 2017 to 2020 (RISØ, HCAB), 2019 to 2020 (HCØ), and 2015 to 2020 (HVID). Figure 2.2 shows annual means for Dp = 11 – 41 nm, Dp = 41 – 110 nm, and Dp = 110 – 478/550 nm for the years 2002 to 2020. Because of previously stated problems with inlets and DMAs, data for Dp = 11 – 41 nm are missing for the years 2017 – 2019 (RISØ and HCAB), 2019 (HCØ), and 2015 -2019 (HVID) for the new instruments.

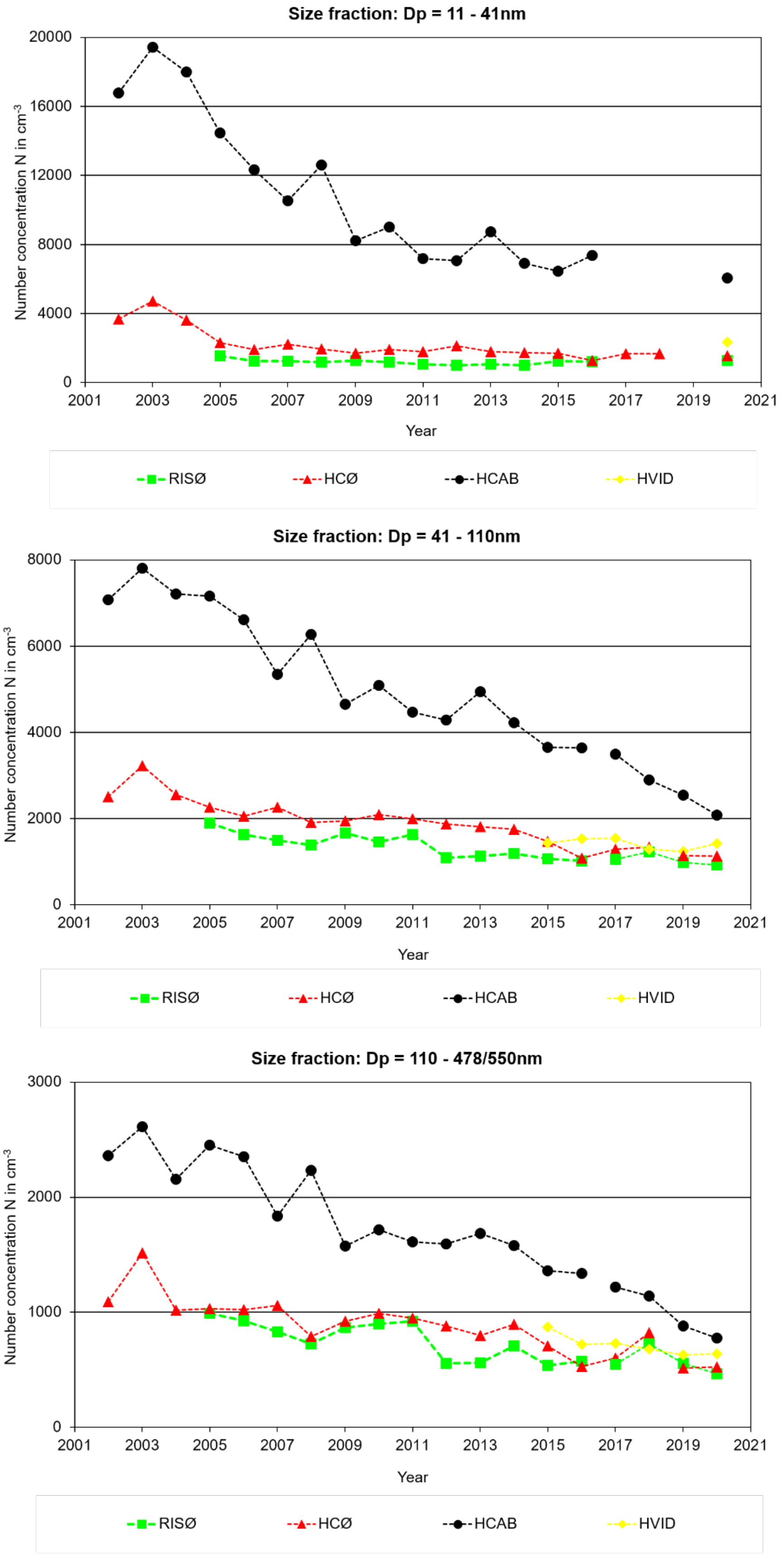


Figure 2.2. Annual mean particle number concentrations in specific size regimes Dp = 11 - 41 nm (upper Figure), Dp = 41 – 110 nm (middle Figure), and Dp = 110 - 478/550nm (lower Figure) combined for old and new instruments at the rural location (RISØ), suburban location (HVID), urban background (HCØ) and urban curbside (HCAB) during 2002 to 2020. Note that values for the size range 11-41 nm were not available at RISØ and HCAB from 2017-2019, HCØ in 2019 and HVID before 2020.

For HCAB, the largest particle number is typically found in the smallest size regime from 11 to 41 nm (Wåhlin, 2009). The small particles mostly originate from traffic emissions and thus the highest concentrations are found close to their sources. Especially in the first ten years of measurements, the particle number concentrations in this size regime have been significantly decreasing to about 50% which is a result of technological development of vehicle engines and changes in fuel composition (Figure 2.2).

No clear trend can be observed at the rural station at Risø and the urban background station at HCØ. This may be explained by the fact that the main sources of particles (11 to 41 nm) at these background locations to a large extent are natural emissions and atmospheric processes and only to a minor extent comes from diluted aerosols originating from anthropogenic sources.

The tendency of decreasing particle numbers is evident at HCAB also for the medium size regime (41 – 110 nm) over all the years. A general decrease in particle number is in accordance with the European trend of avoiding particulate emissions especially from road traffic, for which new environmental regulations and cleaner technologies have continuously been introduced over the years. We observe a slight decreasing trend also at the urban background and the rural measurement site, HCØ and RISØ, respectively, but the particle numbers have been stabilizing in the last five years. Similarly, this finding is observed at the suburban background (HVID), where concentrations seem to be more stable but only measurements are available for the last five years. At HVID, trends have to be investigated in more detail when longer time series are available. It is possible, that at the suburban station concentrations in the middle size regime are influenced by wood combustion emissions in the area. Which could explain higher concentrations compared to the urban background and the rural stations.

For the large size regime (110 – 478/550 nm), a strong decreasing trend is observed at HCAB with respect to the entire measurement period from 2002 to 2020. Similarly, such trend is observed at HCØ, HVID, and RISØ. Nevertheless, some variations are observed for some years which may be linked to this particular meteorological year. In general, decreasing trends are expected as the background aerosol is affected by general emission decreases in Europe and worldwide as a result of emission regulation. This implies that especially at the curbside similar trends as for the smaller size regimes are observed. On the other hand, also natural processes result in relatively stable values over shorter time periods (few years). Such processes include e.g. the emission of VOCs from vegetation that in turn contribute to the formation of new particles through chemical conversion in the atmosphere. Such sources are rather more stable over shorter time periods, but may also change as a result of changing climate over larger time periods as e.g. decades. These freshly formed particles will after some atmospheric transformation as condensation, coagulation and cloud processing enter into the large size regime where their lifetime in the atmosphere are longer

2.3 Total particle number

In Figure 2.3 the total particle number is presented for the rural station (RISØ), the suburban background (HVID), the urban background (HCØ) and the curbside station (HCAB) in the corresponding size range for the old instruments (Dp = 11- 550 nm) and for the new instruments (Dp = 11- 478 nm).

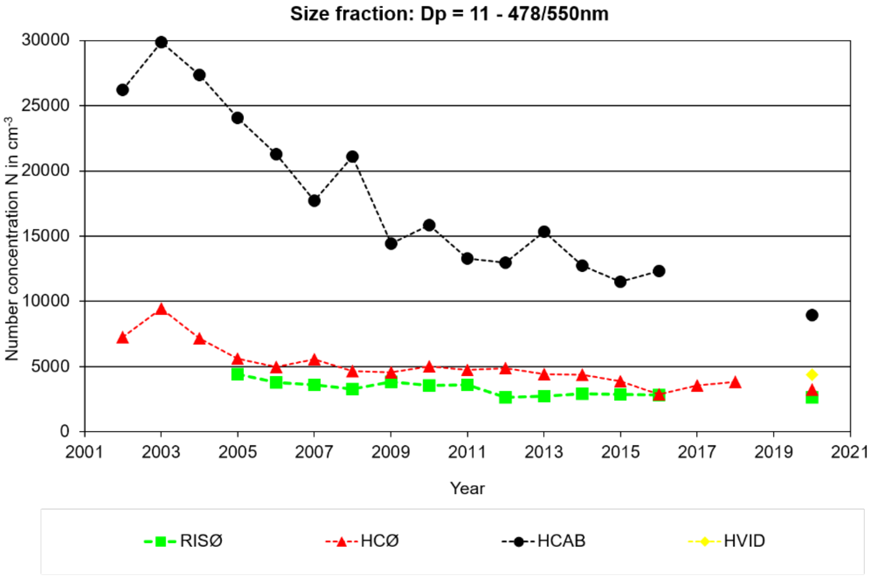


Figure 2.3. Annual mean particle number concentrations for the total size range Dp = 11 – 550nm (old instruments) and Dp = 11 – 478 nm (new instruments) combined at the rural location (RISØ), suburban location (HVID), urban background (HCØ) and urban curbside (HCAB) during 2002 to 2020. Note that values were not available at RISØ and HCAB from 2017-2019, HCØ in 2019 and HVID before 2020.

The general patterns in terms of annual trends reflect the picture that is shown in detail in Figure 2.2 (upper, middle, lower panel). An overall decline in submicrometer particle number is observed at all three stations RISØ, HCØ, and HCAB. The magnitude of this decline is decreasing with the distance to sources of aerosol particles. The rural background station (RISØ) is predominantly affected by long-range transported aerosols, which show a general decline in Europe as emission regulations have been implemented and effective for various sectors, especially the transport sector. At the urban background this decrease in detected background aerosol concentrations is also reflected while at the same time this station receives more influence from the city of Copenhagen, where emission reductions have a larger impact on the trend of total particle number. Finally, the largest decrease is observed at the urban curbside station (HCAB), which is largely influenced by direct traffic emissions from passing light and heavy duty vehicles. For this reason, any emission reductions with respect to the changing car fleet, implementation of technological developments as e.g. exhaust after treatment or changes in fuel composition do affect the total particle number directly at this station and especially in the smallest size fraction where emitted particle numbers are typically highest at curbside locations.

2.4 Impact of Covid-19 restrictions

Covid-19 restrictions were implemented from 13.03.2020 in Denmark and in early spring for large parts of the remaining Europe resulting in major reductions in the activities of the society. These restrictions lead to significant reductions in the transport emissions in Denmark and Europe especially in spring 2020.

The largest decrease from 2019 to 2020 in the annual particle number concentration (41 – 478 nm) was measured at the curbside station HCAB (17%), and it is possible that the Covid-19 restrictions partially explain this decrease. Figure 2.4 shows the temporal variations of particle number concentration in

2019 and 2020 as two-month-average except for the first four months where averages has been calculated for 01.01.2020-12.03.202 (Period 1) and 13.03.2020-30.04.202 (Period 2), respectively. Period 1 represents pre-Covid-19 conditions in 2020 while Period 2 represents the period with the strongest Covid-19 restrictions in 2020 and hence the second period should be the period with the largest impact on the concentrations of particle number concentrations.

The average particle number concentrations are lower in 2020 compared to 2019 for all periods though the difference is small for Period 4 and Period 5. These generally lower particle number concentrations are most likely due to the continuing downward regulations of the emissions (DCE, 2021). Moreover, it was expected that the clearest signal of the Covid-19 restrictions should be seen in Period 2. However, Period 2 does not stand out with an especially large decrease between 2019 and 2020. This indicates that the impact of Covid-19 was not very large or that all the complex physical and chemical processes determining the particle number concentrations and being largely dependent on varying meteorological conditions during the time of the year counteracted the impact from the Covid-19 restrictions.

The difference between the annual particle number concentration in 2019 and 2020 for the three other stations vary from 10% reduction at the rural site RISØ, unchanged at urban background site HCØ and 10% increase at the suburban site HVID. These changes could also be impacted by the Covid-19 restrictions. To an example, information from citizens have indicated that increased work from home has led to an increase in the use of wood burning in residential areas. However, more substantial evidence is needed to support these observations. Unfortunately, there is not sufficient data coverage for the first months in 2019 to carry out a detailed temporal analysis for these three stations.

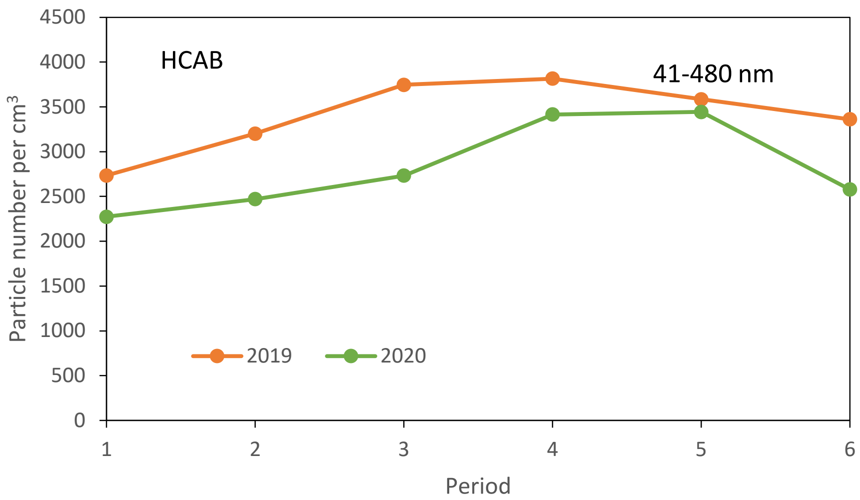


Figure 2.3. Temporal variation of the particle number concentrations at the urban curbside station (HCAB) for the years 2019 and 2020. The results represent averages for 01.01.2020-12.03.202 (Period 1), 13.03.2020-30.04.202 (Period 2), May-June (Period 3), July-August (Period 4), September-October (Period 5) and November-December (Period 6).

3 Elemental carbon (EC) mass concentration

Soot, the blackish or brownish substance formed during incomplete combustion (Andrea and Gelencsér, 2006) is typically measured by exploiting its light absorbing properties as Black Carbon (BC) or its chemically inertness as elemental carbon (EC). In the Danish Air Quality Monitoring Programme, EC is measured using the European standard thermal optical protocol EUSAAR2 (Cavalli et al., 2010). EC is based on a Thermal Optical technique as carbon, which is only combusted in the presence of oxygen at temperatures higher than 500°C (Birch and Cary, 1996; Cavalli et al., 2010).

Low Volume Samplers (LVS) equipped with $PM_{2.5}$ inlets are located at urban curbside (HCAB) and urban background (HCØ) in addition to rural (RISØ) and suburban (HVID) locations. Atmospheric aerosols are collected on quartz fiber filters using 24-hour time resolution. The filters are subsequently weighted to measure $PM_{2.5}$ mass concentration, and punches of the filters are analyzed for EC by a Thermal/Optical carbon analyzer (Sunset Laboratory, Oregon USA) according to the EUSAAR 2 protocol (Cavalli et al., 2010).

EC has been monitored routinely at RISØ and HCAB from 2009/2010 and onwards. Monitoring of EC was extended to urban background in Copenhagen by September 2014, and to a suburban location in Hvidovre from October 2015.

Table 3.1 presents the annual averages measured at the four stations in 2020. In 2020, annual mean values were: rural EC 0.22 $\mu g/m^3$ < urban background EC 0.27 $\mu g/m^3$ < suburban EC: 0.32 $\mu g/m^3$ < urban curbside EC 0.72 $\mu g/m^3$ (Table 1.5). This pattern is consistent with the fact that the highest concentrations are measured at the curbside due to the strong emissions of EC from road traffic and that the lowest concentrations are measured in rural areas with only few local sources. The emissions in the city increases the concentrations at urban background with about 21% compared to the rural location. The concentrations at the suburban site are about 46% higher than in the rural areas. The higher concentrations at the suburban site are mainly due to the local impact from residential combustion.

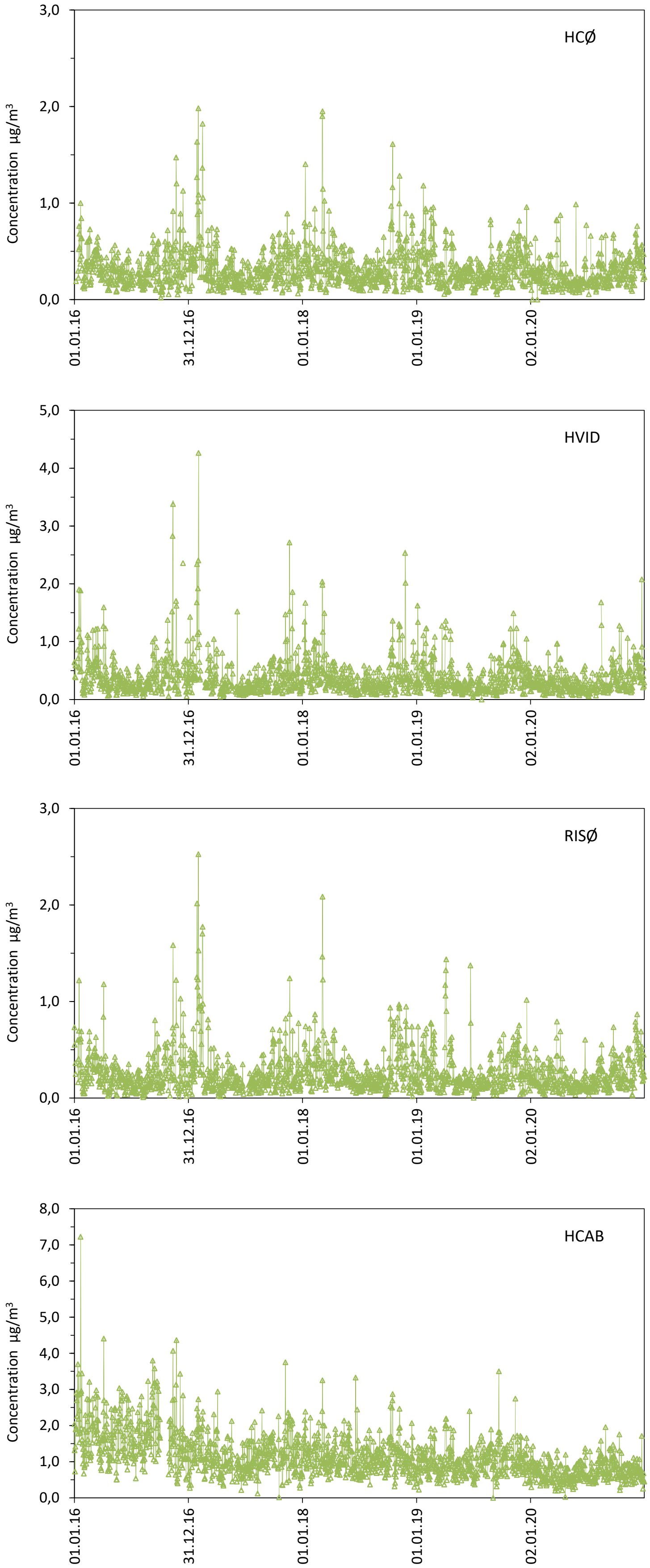
Table 3.1. 2020 annual statistics for EC at Danish measurement sites. Annual mean value for EC and annual mean value for EC at urban background (HCAB), suburban (HVID) and urban curbside (HCØ) relative to rural site (Risø).

	Data coverage %	EC $\mu g/m^3$	EC relative to rural site
Urban Street			
HCAB	99%	0.72	223%
Suburban site			
HVID	95%	0.32	46%
Urban background			
HCØ	97%	0.27	21%
Rural site			
RISØ	96%	0.22	

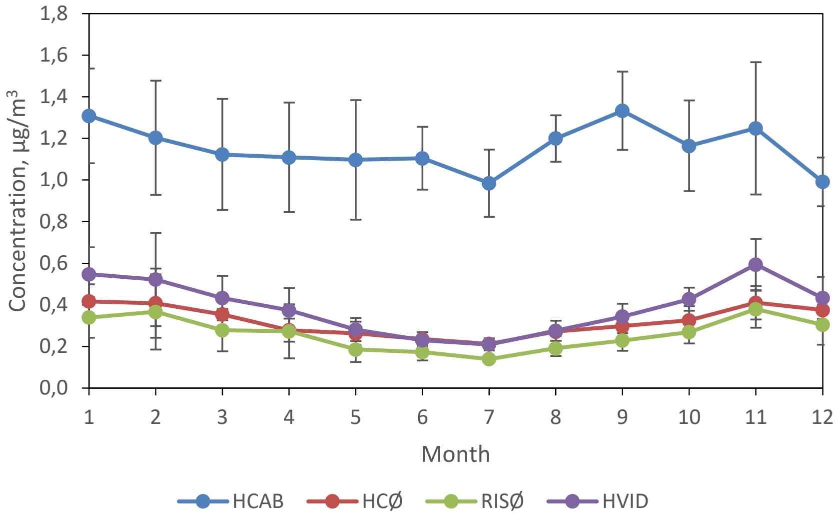
Figure 3.1 shows the measured time series of diurnal averages from all four stations since 2016. A clear seasonal variation can be seen in urban background and at the rural and suburban locations with higher concentrations during winter compared to summer. This variation is mainly caused by biomass combustion, including residential wood combustion, as concluded in *The Particle Project 2017-2018* (Nøjgaard et al., 2018). In contrast, the seasonal variation at the curb site station HCAB is less pronounced. Here the emissions of EC from the road traffic are the most dominant source and hence the seasonal variation is much smaller compared to the other three stations. More striking is the clear decreasing trend in EC, which matches the trend in particle number concentration in Figure 2.2

Figure 3.2 shows monthly averages for the four stations averaged over the period from 2016 to 2020. Again, the seasonal variation with higher winter concentrations and lower summer concentrations is evident for the three stations in urban background and at the rural and suburban locations. Based on five years of measurements at HCØ, mean winter concentrations average 0.43 $\mu g/m^3$ during December-January-February, while summer concentrations show mean values of 0.24 $\mu g/m^3$ during June-July-August. Autumn concentrations are slightly higher than spring concentrations, i.e. 0.36 vs. 0.31 $\mu g/m^3$. Peak concentrations of 1 to 2 $\mu g/m^3$ are occasionally observed at HCØ (Figure 3.1).

At the curbside station HCAB only a small seasonal variation is observed due to the low seasonal variation in the emissions from the road traffic. The difference between winter and summer is less than 10% for the five year averages (winter 1.3 $\mu g/m^3$ vs. summer 1.2 $\mu g/m^3$). The relatively low concentrations in July and December are most likely due to the summer and Christmas vacations.



Figur 3.1. Time series for the diurnal average concentrations of EC at urban background (HCØ), suburban (HVID) and rural location (RISØ) and urban curbside (HCAB).



Figur 3.2. Monthly averaged concentrations of EC based on the period 01.01.2016 – 31.12.2020 at urban background (HCØ), suburban (HVID) and rural location (RISØ) and urban curbside (HCAB).The error bars shows the standard variation of the monthly averages for the five year period.

Figure 3.3 shows the long term trends for the annual averages of EC at the four stations. Urban background measurements of EC were not available until 2015 when initiated in *The Particle Project 2014-2016* (Nøjgaard et al., 2017). Since 2015, EC has decreased about 29% (0.1 µg/m^3) at the background station HCØ. The main reason for the decrease in the concentrations at HCØ is the general reductions in the emissions at national as well as at international level.

At HCAB there has been a pronounced reduction of about 67% (1.7 µg/m^3) in the annual average concentrations over the time period 2010 to 2020, predominantly due to a reduction in road traffic emissions. This is a result of improved combustion technology in general, and the fact that the share of particle filters within the fleet of diesel vehicles has increased with more stringent emission standards for newer vehicles. The general reduction in emissions in Europe and urban areas has also contributed to the reductions at HCAB although these reductions play a minor role at HCAB.

EC has decreased by 41% (0.19 µg/m^3) at RISØ over the time period 2010 to 2020. At the suburban site HVID there has been a reduction since 2016 of about 25% (0.1 µg/m^3). Since 2016 the reductions have been quite similar at the urban background, suburban and rural sites indicating that it is the general reductions on national and international level that explain the majority of the reductions.

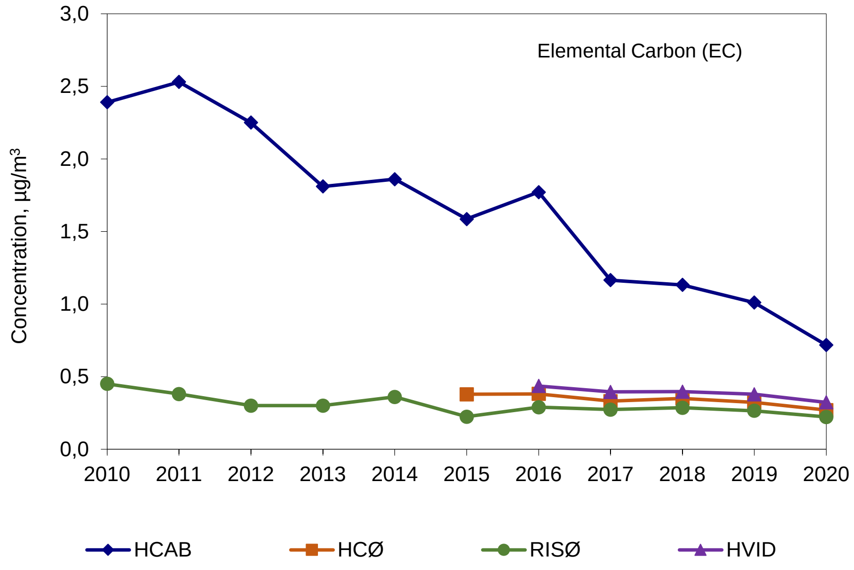


Figure 3.3. Long term trends in annual average concentrations of EC at urban background (HCØ), suburban (HVID) and rural location (RISØ) and urban curbside (HCAB).

3.1 Impact of Covid-19 restrictions

As seen from Figure 3.3 there is a rather large decrease in the concentrations from 2019 to 2020. This is most clearly seen at the curbside station HCAB where the annual concentration of EC decreased with 29%. At the three other stations there is a decrease of 15-17% from 2019 to 2020.

One of the reasons for these decreases can be the Covid-19 restrictions that were introduced on 13.03.2020 in Denmark and in early spring for large parts of the remaining Europe. These restrictions resulted in significant reductions in many of activities in the society. These reductions where especially large in spring 2020 and an analysis of the temporal variation of the concentrations of EC has therefore been carried out in order to investigate the impact of Covid-19.

Figure 3.4 shows the temporal variations of the EC concentration as average of the concentrations over two-monthly periods except for the first four months where averages have been calculated for 01.01.2020-12.03.2020 (Period 1) and 13.03.2020-30.04.2020 (Period 2), respectively. Period 1 represents pre-Covid-19 conditions while the second period represents the period with the strongest Covid-19 restrictions and hence the second period should be the period with the largest impact on the concentrations of EC. Figure 3.4 includes data from the years from 2016 to 2020 in order to be able to compare the temporal variation in 2020 with the typical temporal variations from the previous years.

At the urban background station HCØ the largest difference between 2020 and the previous four years is for Period 1 that is before the onset of the restrictions in Denmark. For the remaining five periods, 2020 followed the typical temporal variation. The relatively large decrease seen for Period 1 is most likely explained by unusual large amounts of precipitation in February (DMI, 2021) that led to a decrease in the concentration of EC by wet-deposition. The temporal variation shows therefore that the Covid-19 restrictions had only minor impact on the concentrations of EC in 2020 and that the main reasons for the decrease from 2019 to 2020 are the general reductions in the emissions and the natural variations in the meteorological conditions from year to year. Similar conclusions can be drawn for the rural and suburban stations.

For the curbside station HCAB, there are in general lower concentrations throughout the entire year 2020 that most likely have to be explained by the general reductions in the emissions from traffic (DCE, 2021), the variations in the meteorological conditions, and the specific reductions from traffic during the Covid-19 lockdown. However, there is also a nearly 50 % lower concentration of EC in Period 2 in 2020 compared to 2019 and the previous years. The temporal pattern for 2020 shows therefore that the Covid-19 restrictions have decreased the concentrations of EC in 2020 at the curbside station, which is also expected as a consequence of the large decrease in traffic during especially springtime 2020. However, it is difficult to separate the influence from Covid-19 restrictions, the general decrease in the emissions and variations in meteorology and it is therefore not possible to quantify exactly the impact of the Covid-19 restrictions based on the current knowledge and data availability.

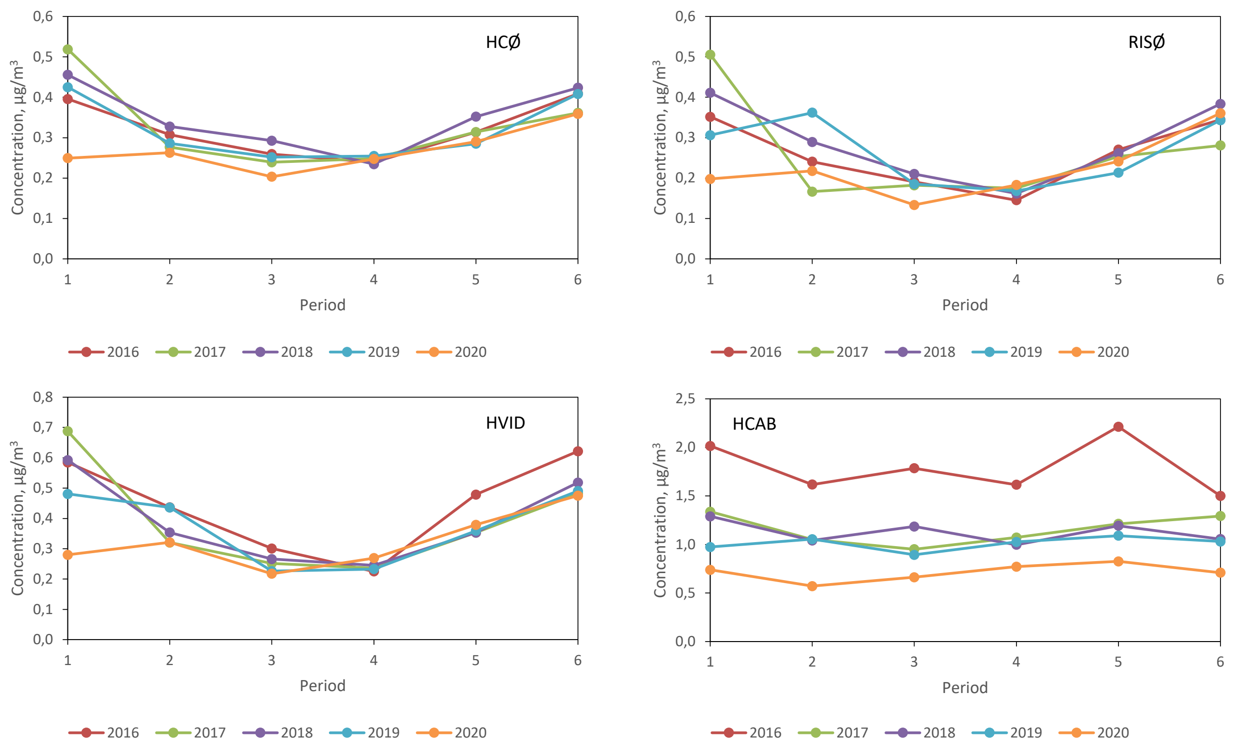


Figure 3.4. Temporal variation of the concentrations of EC at urban background (HCØ), suburban (HVID) and rural location (RISØ) and urban curbside (HCAB) for the years from 2016 to 2020. The results represent averages for 01.01.2020-12.03.2020 (Period 1), 13.03.2020-30.04.2020 (Period 2), May-June (Period 3), July-August (Period 4), September-October (Period 5) and November-December (Period 6).

References

Andrea, M.O., Gelencser, A., 2006. Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols. Atmospheric Chemistry and Physics 6, 3131 – 3148.

Birch, M.E., Cary, R.A., 1996. Elemental carbon-based method for monitoring occupational exposures to particulate diesel exhaust. Aerosol Science and Technology 25, 221 – 241.

Cavalli, F., Viana, M., Genberg, J., Putaud, J.-P., 2010. Toward a standardised thermal-optical protocol for measuring atmospheric organic and elemental carbon: the EUSAAR protocol. Atmospheric Measurement Techniques 3, 79-89.

DCE, 2021: Air emissions (au.dk)

DMI, 2020. https://www.dmi.dk/fileadmin/user_upload/Afrapportering/Seasonsammendrag/Sammendrag_2019_sommer.pdf

DMI, 2021. Sammendrag_2020.pdf (dmi.dk)

Ellermann, T., Nygaard, J., Nøjgaard, J.K., Nordstrøm, C., Brandt, J., Christensen, J., Ketzel, M., Massling, A., Bossi, R. & Jensen, S.S., 2017. The Danish Air Quality Monitoring Programme. Annual Summary for 2016. Aarhus University, DCE – Danish Centre for Environment and Energy, 78 pp. Scientific Report from DCE – Danish Centre for Environment and Energy No. 234

Nøjgaard, J. K., Massling, A., Christensen, J. H., Nordstrøm, C., & Ellermann, T. (2015) The Particle Project 2011-2013, Aarhus University, DCE – Danish Centre for Environment and Energy, Scientific Report from DCE, No. 156, 55p.

Nøjgaard, J. K., Massling, A., Ellermann, T., 2017. The Particle Project 2014-2016. Aarhus University, DCE – Danish Centre for Environment and Energy, 40 pp. Scientific Report from DCE – Danish Centre for Environment and Energy No. 233. http://dce2.au.dk/pub/SR233.pdf

Nøjgaard, J. K., Massling, A., Ellermann, T., 2018. The Particle Project 2017-2018. Aarhus University, DCE – Danish Centre for Environment and Energy, 30 pp. Scientific Report from DCE – Danish Centre for Environment and Energy No. 285. The Particle Project 2017-2018 (au.dk)

Nøjgaard, J. K., Massling, A., Ellermann, T., 2020. The Particle Project 2019. Aarhus University, DCE – Danish Centre for Environment and Energy, 22 pp. Scientific Report from DCE – Danish Centre for Environment and Energy No. 385. http://dce2.au.dk/pub/SR385.pdf

Wåhlin, P. (2009) Measured reduction of curbside ultrafine particle number concentrations in Copenhagen, Atmos Environ, 43, 3645 – 3647.

THE PARTICLE PROJECT 2020

The Particle Project 2019 continues the record of air quality parameters, which are a supplement to the The Danish Air Quality Monitoring Programme. Particle size distribution and number measurements in urban background are compared to urban street, suburb and rural locations. Monitoring of high-time-resolution $PM_{2.5}$ and PM_{10} are shown for rural and selected urban stations. Elemental Carbon (EC) in urban background is compared to rural location, suburb and curbside station in. Major sources are traffic and wood combustion that influence urban background and suburbs, for which reason these locations have higher EC concentrations than the rural site, but much less than the Copenhagen curbside station.

ISBN: 978-87-7156-606-2
ISSN: 2245-0203