THE PARTICLE PROJECT 2022

Scientific Report from DCE – Danish Centre for Environment and Energy No. 562 2023

AARHUS UNIVERSITY
DCE – DANISH CENTRE FOR ENVIRONMENT AND ENERGY

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Data sheet

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Abstract: The Particle Project 2022 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 street, 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 temporal variations of the particulate air pollution.

Keywords:	Particulate air pollution, $PM_{2.5}$, particle number and size distribution, elemental carbon, status 2022, temporal variation, long term trends, urban background,
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Contents

Sammenfatning

I *The Particle Project 2022* 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 og bidrager med forskning omkring kilderne til partikelforureningen i Danmark. Resultaterne for bybaggrund sammenholdes med tilsvarende resultater fra en landligt baseret station nord for Roskilde (RISØ), en station i den Københavnske forstad Hvidovre (HVID), samt fra en station i en trafikeret gade i København (HCAB). I rapporten præsenteres resultater for niveauer og udviklingstendenser for partikelantal og elementært kulstof sammen med en analyse af sæson- og døgnvariation af disse partikelkomponenter. Analysen af den tidsmæssige variation suppleres med resultaterne fra analyse af døgnvariation i $PM_{2,5}$ og PM_{10} baseret på målinger af PM med høj tidsopløsning via TEOM-metoden (Tapered Element Oscillating Microbalance).

Partikelantal

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 på HCØ (bybaggrund), dernæst kommer HVID (forstad), 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 begyndte 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 måleområdet 11 – 478/550nm.

Partikler mellem 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.

Partikler mellem 41 og 110 nm og partikler mellem 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 af forbedring af vognparken slår tydeligere igennem på gademålestationen end ved baggrundsmålestationerne.

Partikler mellem 11 – 478/550nm: Når partikelantallet for det samlede 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 – navnlig i den første del af måleserien (2002-2012), hvorefter faldet flader ud.

Elementært kulstof, EC

Årsmiddelkoncentrationer for EC blev i 2022 målt til 0,23 µg/m^3 ved bybaggrundsmålestationen HCØ, hvilket er omkring 6% lavere end målt i 2021. Ved den landlige målestation RISØ var faldet på omkring 17% og ved gademålestationen HCAB på omkring 11%. Da forstadsmålestationen HVID blev flyttet i sommeren 2021, var der kun data for omkring 6,5 måneder. Dermed er der ikke data for 2021 til sammenligning med 2022 og ikke data til beregning årsmiddelværdier. Niveauet ved forstadsmålestationen HVID var i 2022 omtrent på niveau med årsmiddelkoncentrationen for 2020.

Siden 2015 er koncentrationerne af EC faldet med omkring 40% ved bybaggrundsmålestationen 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 målt et stort fald på omkring 78% i perioden fra 2010 til 2022, hvilket hovedsageligt skyldes fald i udledningerne fra vejtrafik.

EC er faldet med omkring 60% ved den landlige målestation RISØ i perioden fra 2010 til 2022. Ved forstadsmålestationen HVID er der målt et fald på omkring 23% 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.

Tidsmæssige variationer

Denne årsrapport præsenterer resultater fra analyse af de tidsmæssige variationer i luftforureningen med partikler. Døgnvariationen i partikelmassen er undersøgt ud fra målinger af $PM_{2,5}$ og PM_{10} med TEOM-metoden, som måler partikelmassen på timemiddelniveau ved den landlige målestation RISØ (PM_{10}) og ved gademålestation HCAB ($PM_{2,5}$ og PM_{10}). Målingerne af EC udføres som døgnmiddelmålinger, og disse er anvendt til fastlæggelse af månedsvariationen af EC ved alle fire målestationer. Partikelantal måles på timemiddelniveau, og disse data er anvendt til både at bestemme måneds- og døgnvariationen.

Analysen af månedsvariationen af EC viser, at EC har de højeste koncentrationer om vinteren og de laveste koncentrationer om sommeren. Dette hænger sammen med, at EC hovedsageligt stammer fra udledninger fra vejtrafik og boligopvarmning baseret på brændefyring. Månedsvariationen er mindst ved gademålestationen HCAB og højest ved forstadsmålestationen HVID, hvorefter følger bybaggrundsmålestationen HCØ og den landlige målestation RISØ. I 2022 var månedsmiddelkoncentrationen særligt høj i marts set i forhold til tidligere år. Dette skyldtes hovedsageligt, at det stort set ikke regnede i marts og at det tørre vejr banede vej for episoder med særligt høje koncentrationer af EC. Tilsvarende sås relativt høje månedsmiddelkoncentrationer af de øvrige partikelparametre rapporteret i denne rapport.

I modsætning til dette, så er partikelantal højere om sommeren end om vinteren i både 2021 og 2022 ved bybaggrundsmålestationen HCØ og den landlige målestation RISØ. Ved gademålestationen HCAB ses også lidt højere partikelantal sommer end vinter om end forskellen ikke er helt så tydelig, som ved de andre målestationer. Årsagen til disse månedsvariationer er, at partikler med diameter mellem 11 og 478 nm stammer fra udledninger fra vejtrafik og boligopvarmning med brændefyring samt fotokemiske reaktioner i atmosfæren. De fotokemiske reaktioner forløber kun, når der er sollys, og derfor ses et relativt stort bidrag fra de fotokemiske reaktioner om sommeren. Ved gademålestationen HCAB er partikelantallet stærkt domineret af udledninger fra vejtrafikken, og bidraget fra de fotokemiske reaktioner er derfor formentlig for lille til at kunne blive observeret.

Døgnvariationen af PM_{10} ved gademålestationen HCAB viste i både 2021 og 2022 det forventede mønster præget af udledningerne fra vejtrafikken, men der blev også observeret en uventet bred top fra sent på formiddagen til midt på eftermiddagen i den årlige gennemsnitlige døgnvariation. Denne brede top blev set i både 2021 og 2022, men af to forskellige årsager. Analyserne af data fra 2021 viste, at den brede top stammede fra vintersaltning af vejen i den første del af februar, mens den brede top i 2022 skyldtes en række episoder med relativt høje koncentrationer, som følge af meget lav nedbør i marts. Ved den landlige målestation RISØ blev der ikke observeret en klar døgnvariation, hvilket hænger sammen med, at langt hovedparten af $PM_{2,5}$ stammer fra langtransporteret luftforurening.

Døgnvariationen i partikelantal i 2022 var overordnet set tilsvarende i 2021. Døgnvariationen viste mange interessante mønstre som for en stor del kan tilskrives de forskellige kilder til partikelantal. Eksempler på dette ses på døgnvariationen ved gademålestationen HCAB, hvor der tydeligt kan identificeres toppe relateret til myldretid, og toppe der kan tilskrives udledninger i forbindelse med de sociale aktiviteter lørdag aften og anvendelse af fyrværkeri. Ved bybaggrundsmålestationen HCØ, ses en tydelig top i partikel antal fra sidst på formiddag til ud på eftermiddagen om sommeren (april-september). En lignende top kan ikke ses om vinteren (januar-marts og oktober-december). Denne top kan bedst forklares ved de fotokemiske reaktioner i atmosfæren, som fører til dannelse af partikler, når solen skinner om sommeren. Ved forstadsmålestationen HVID blev der observeret relativt højt partikelantal på vinteraftner, hvilket formentligt skal tilskrives boligopvarmning med brændefyring.

Summary

The *Particle Project 2022* presents results from measurements of particle number concentrations of submicrometer particles and Elemental Carbon (EC) in fine particles with diameter smaller than 2.5 µm ($PM_{2.5}$) at the urban background station HCØ and for comparison also results from three other stations in the Copenhagen area. In order to increase knowledge on the sources of these particulate air pollutants the measurements are carried out as a supplement to the Danish air quality monitoring program under NOVANA (the National Monitoring and Assessment Programme for the Aquatic and Terrestrial Environment). Trends, seasonal - and diurnal variations are presented for four stations: urban background (HCØ), rural location (RISØ), suburban location (HVID) and urban street (HCAB).

Particle number

Regional and long-range transported aerosols 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. At the street station HCAB, 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 periods of the order of a decade. A general decreasing trend is observed over the past 15 – 20 years for the rural background station RISØ, the urban background station HCØ and the street station HCAB when considering the size range 41 - 110 nm and 110 – 478/550 nm. However, for these fractions over the past five years, the measurements show very stable numbers for the rural station RISØ and the urban background station HCØ 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, a decreasing trend is continuously observed at the street station HCAB for these two size fractions over the past five years where substantial local contributions from traffic are expected.

Data for particle numbers in the size range 11 – 41 nm 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 station RISØ and the urban background station HCØ. A decreasing trend can be observed at the street station HCAB which levels more and more out over the past years.

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

Elemental carbon, EC

The annual mean concentration of EC in 2022 at the urban background station HCØ in Copenhagen was 0.23 µg/m^3, which is about 6% lower than measured in 2021. At the rural station, RISØ the decrease was about 17% and at the urban street station HCAB about 11%. The suburban station HVID had to be moved during summer 2021 and hence there were only data for about 6.5 months and no data available for a comparison between 2021 and 2022. However, the concentration level at the suburban station HVID was about the same level in 2022 as in 2020.

Since 2015, EC has decreased about 40% 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 60% at the rural background station RISØ over the period 2010 to 2022. At the suburban station HVID there has been a reduction from 2016 to 2022 at about 23%. Since 2016, the decreasing trends have been quite similar at urban background, suburban and rural stations, indicating that the general reductions in emissions at national and international level can explain the major part of the decreasing trends.

At HCAB, there has been a pronounced reduction at about 78% in the annual average concentrations over the period 2010 to 2022, predominantly due to a reduction in local road traffic emissions, caused by introduction of cars with newer and cleaner engine technology and exhaust after treatment such as particle filters.

Temporal variations

This annual report presents results from an analysis of the temporal variations of the particulate air pollution. The diurnal variation of particulate mass is analysed based on measurements of $PM_{2.5}$ and PM_{10} using the TEOM method (Tapered Element Oscillating Microbalance) that measures particulate mass with hourly time resolution at the regional background station RISØ (PM_{10}) and the street station HCAB ($PM_{2.5}$ and PM_{10}). The measurements of EC are carried out with a time resolution of 24 h and these data are used to determine the monthly variations at all four stations. Particle number is measured with hourly variation and these data are used to study both the monthly and diurnal variation at all four stations.

The analysis of the monthly variations of EC shows that EC has the highest concentrations during winter and lowest concentrations during summer. This is because EC mainly originates from emissions from road traffic and household warming using wood burning and that emissions due to wood burning has a strong seasonal variation with highest emissions during winter and lowest during summer. The smallest variation is found at the street station HCAB and the highest variation at the suburban station HVID followed by the urban background HCØ and regional background station RISØ.

In contrast to the EC variation, the particle number is higher during summer than during winter at the urban background station HCØ and regional background station RISØ. At the street station HCAB, the particle number is also higher during summer than winter although the difference between the seasons is smaller. The reason for these monthly variations is the particle number originating from road traffic and wood burning and additionally from photochemical reactions in the atmosphere. The photochemical reactions require sunlight and therefore represent an additional source to the particle number during summer. At the street station HCAB the particle number is strongly dominated by the road traffic emissions and the contribution from the chemical formation of particles is most likely too small to be observed in the monthly variations.

The diurnal variations of PM_{10} at the street station HCAB show the expected patterns consistent with road traffic being the main source of PM_{10} although an unexpected broad peak from late morning to middle afternoon was also found. This broad peak was seen both in 2021 and 2022 but due to different reasons. In 2021 the broad peak was caused by winter salting of the street during the first part of February while the peak in 2022 was due to episodes with high concentrations due to the very low amount of precipitation during March. $PM_{2.5}$ at HCAB showed a less pronounced diurnal variation in agreement with the main part of $PM_{2.5}$ originating from long-range transport and only minor contributions coming from road traffic emissions. At the rural background station RISØ, there was no clear diurnal variation consistent with long-range transport as the main source.

The diurnal variations of particle number were to a large extend similar in 2021 and 2022 and showed many interesting patterns that to a large part could be assigned to the sources. Examples on these are the clear rush hour peaks at the street station HCAB and the impact from social activities during Saturday evenings and use of fireworks. At the urban background station HCØ a large peak during midday and afternoon during the summer half year was observed and that peak could not be found during the winter half year. This peak can best be explained by the atmospheric photochemical reactions that leads to particle formation during sunlight periods in the summer half year. This peak was also seen at the rural background station RISØ but not at the street station HCAB. At the suburban station HVID relatively high concentrations were found during winter evenings most likely due to household warming using wood burning.

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 (as an example see Nøjgaard et al., 2020 or Ellermann et al., 2021). 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 submicrometer 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 with studies of health impact of airborne particle pollution in Denmark.

This report presents the results from the particle project in 2022 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 (HCØ). 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 long-term measurements of particle number at all four Danish locations have also been extremely valuable as validation dataset during the introduction of particle number as a new pollutant in the DCE modelling system (DEHM-UBM-AirGIS; Frohn at al. 2021 and Ketzel at al. 2021)

The main purpose of the project is the measurements in urban background (HCØ). In order to set these results into context, they will throughout the report be compared with the results from the street station at H.C. Andersens Boulevard (HCAB) in Copenhagen, the suburban station in Hvidovre (HVID) and the rural background station at Risø (RISØ) north of Roskilde.

The report also presents results from an analysis of temporal variation in $PM_{2.5}$ and PM_{10} based on the results from the measurements of $PM_{2.5}$ and PM_{10} at hourly resolution using the TEOM method (Tapered Element Oscillating Microbalance). These measurements are part of the Danish air quality monitoring program, and they are useful in connection with the evaluation of the reasons behind the observed temporal variations in particle number concentrations and EC.

The analysis of $PM_{2.5}$ and PM_{10} is presented first in the report since this analysis serves as a background for the understanding of the temporal variations in particle number and EC. Chapter 3 presents results on EC and chapter 4 on particle number.

2. Temporal variation of $PM_{2.5}$ and PM_{10} based on TEOM measurements

In the Danish national air quality monitoring program, the main measurements of $PM_{2.5}$ and PM_{10} are carried out using low volume sampling and gravimetric determination of the particle mass with 24-hour time resolution. These measurements follow the reference method specified in the EU air quality directive (EU, 2008).

$PM_{2.5}$ and PM_{10} are also measured using the TEOM method (Tapered Element Oscillating Microbalance) at the street station HCAB ($PM_{2.5}$ and PM_{10}) and the rural background station RISØ (PM_{10}). No measurements are carried out with the TEOM method at the urban background station HCØ.

The reason for using this additional particulate matter measurement method is that the TEOM method enables measurements of hourly averages of $PM_{2.5}$ and PM_{10} and hence these measurements can be used for the near real time information of the public concerning the actual status of the air quality of $PM_{2.5}$ and PM_{10}. Moreover, the results from the TEOM method can be used for analysis of the diurnal temporal variations in $PM_{2.5}$ and PM_{10}.

The TEOM method has a higher uncertainty than the gravimetric method and one of the main reasons for this is that part of the more volatile particle components can evaporate during the measurements and this has to be kept in mind when the results from the TEOM method are used. Consequently, the long-term trends of $PM_{2.5}$ and PM_{10} are only determined using the reference method and not the TEOM method. The long-term trends are reported in the annual reporting from the monitoring program (see Ellermann et al., 2023 for the most recently updated reporting).

In the following, diurnal variations in $PM_{2.5}$ and PM_{10} will be presented based on the data from the TEOM method.

2.1 Diurnal variation

The average diurnal variation in the concentrations of particulate matter is shown in Figure 2.1 - 2.3 for 2021 and 2022. The average diurnal variations are calculated by averaging all the hourly concentrations measured from 00:00 - 1:00 for the entire period of interest and the same for all 24 diurnal hours. If the period is long enough (typically 2-3 years), it is possible to average out the variations in meteorology and the final variation will illustrate eventual diurnal variations in emissions or chemical formations of an air pollutant. If the period is too short some "random" variation due to the meteorological variations will have a higher degree of influence on the calculated average diurnal variation.

The diurnal variations have been divided into working days (includes a few holidays due to Easter, Christmas etc.), Saturdays and Sundays in order to capture the difference in the diurnal variations of emissions on the different days of the week. There are only 52 Saturdays or Sundays during a year compared with 260 working days. Hence there will be higher remains from the meteorological variations left in the curves for Saturdays and Sundays compared with working days i.e. that the greater spread between the curves for Saturdays and Sundays compared to the curves for working days is mostly due to the fewer number of measurements.

Figure 2.1 shows the average diurnal variation for PM_{10} at the rural background station RISØ. The majority of PM_{10} at RISØ originates from long-range transport and it is therefore expected that there will be no clear diurnal variations and no variation between different days of the week. The observed variations differs between 2021 and 2022 both with respect to season and days of the week (Figure 2.1) although the small broad peak around the morning rush hour might be an indication of a small contribution from local traffic. d. The variations seen in Figure 2.1 for 2021 and 2022 are therefore most likely due to the natural variations in the meteorological conditions with a possible minor impact from local traffic. Figure 2.1 also shows data for the two-year average for 2021-2022 and these curves are flatter than for the individual years because of the longer averaging period will average out more of the natural variation in the meteorological conditions corresponding to the one-year averages.

Figure 2.2 shows the average diurnal variations for $PM_{2.5}$ at the street station at HCAB. About 75% of $PM_{2.5}$ at HCAB originates from long-range transport while road traffic is responsible for the main part of the remaining 25% of $PM_{2.5}$ (Ellermann et al., 2022a). Hence, it is expected that the average diurnal variation is the sum of a large quite constant part from the long-range transported $PM_{2.5}$ and a smaller contribution reflecting the impact of the diurnal variation of road traffic on $PM_{2.5}$.

The same pattern is seen for 2021, 2022 and the two-year average for 2021-2022 for working days and all three curves are very similar. All curves show the expected pattern with a morning peak 5:00 – 9:00 corresponding to the morning rush hour. The afternoon rush hour is less clearly seen because of higher meteorological dispersion of the emissions during the afternoon and most likely also because the afternoon rush hour period is spread over a longer time period than the morning rush hour.

There is relatively high concentration of $PM_{2.5}$ during the evening. This might be because of increased road traffic due to leisure activities (i.e. Friday) and for the winter period it might also be due to a contribution from use of wood burning as household warming, but this has to be investigated further.

Saturdays and Sundays show different patterns than working days and with some more variations between the years. One of the main differences is lack of the working day rush hours in the average diurnal variations. In addition, there are higher values of $PM_{2.5}$ from late morning until the late evening and early night reflecting a more even road traffic during weekends with a later start in the morning compared with working days. Moreover, there is significant difference between 2021 and 2022 for especially two hours during Saturdays i.e. at midnight from 00:00-01:00 and during late evening between 22:00-23:00. The large difference between 2021 and 2022 from 00:00-01:00 is because the new-year began on a Saturday in 2022, so the large peak in 2022 is due to firework. In 2021 new-year began on a Friday.

It is also fireworks, that are the most likely explanation for the second instance since Tivoli has large firework Saturday evenings from 22:00-23:00. Difference in meteorology between 2021 and 2022 is the most likely reason why the fireworks are more clearly seen in 2021 and not in 2022.

Figure 2.3 shows the average diurnal pattern of PM_{10} at the street station HCAB. As expected the pattern for PM_{10} looks like the pattern for $PM_{2.5}$ but with a stronger influence from road traffic since road traffic makes up a larger part of PM_{10} than $PM_{2.5}$. However, as stated in the report for 2021 (Ellermann et al., 2022b), the measured average diurnal variation for 2021 did not look quite as expected due to a large broad peak during midday and early afternoon. The large broad peak has been assigned to winter salting of the road during a period with frosty conditions and no precipitation in February 2021. A similar broad peak is seen for 2022 (Figure 2.3), but detailed analysis of data for 2022 showed that the broad peak is due to unusual high concentrations during two episodes in March 2022. It nearly did not rain in March 2022 (Figure 2.5) and this lead to episodes with very high concentrations of PM_{10} in Copenhagen. In addition, there was also a large episode around 17th, March 2022 with dust transported all the way from Sahara to Denmark (TV2, 2022). The impact of the episodes with high concentrations of PM_{10} during March 2022 are illustrated in Figure 2.4 where the annual average diurnal variation on working days is shown with and without data for March 2022. It is clear from this comparison that these episodes in March has a high contribution to the broad peak in 2022. The impact of fireworks are also seen for PM_{10} on Saturdays although the impact of fireworks are somewhat lower for PM_{10} compared with $PM_{2.5}$.

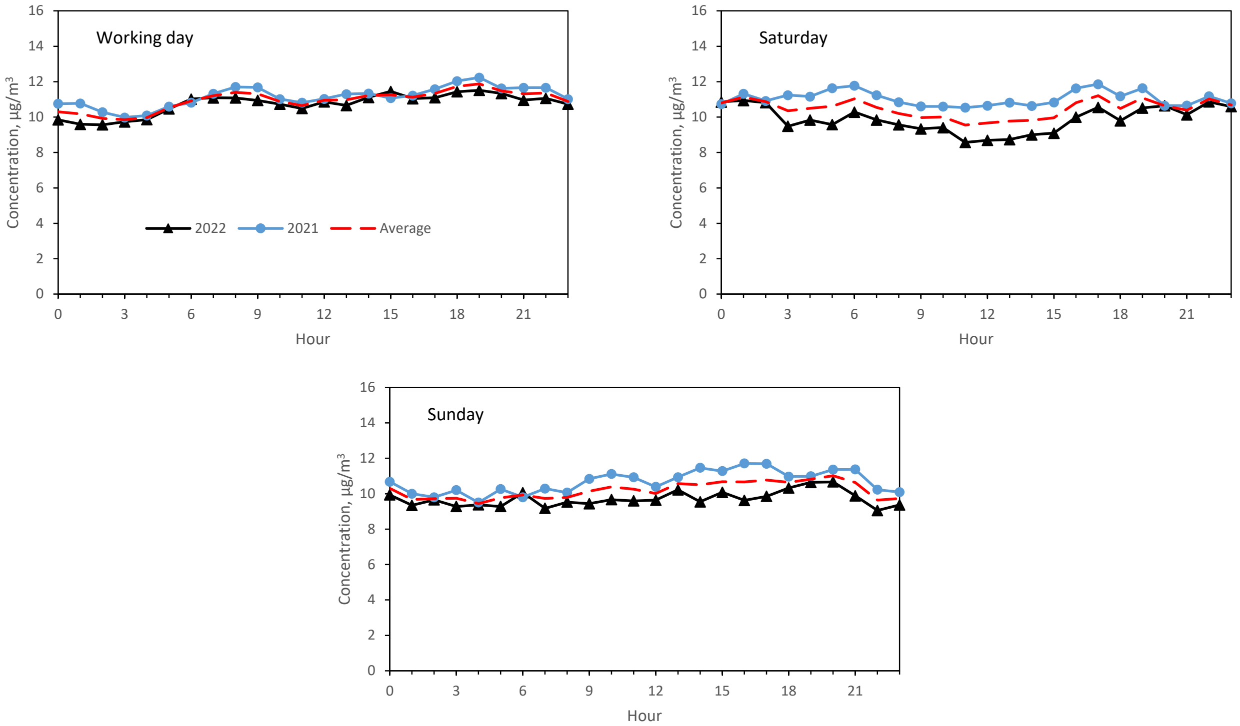

Figure 2.1. Average annual diurnal variations of PM_{10} in 2021 (blue), 2022 (black) and two-year average for 2021 and 2022 (red) at the rural background station RISØ divided into working days, Saturdays and Sundays.

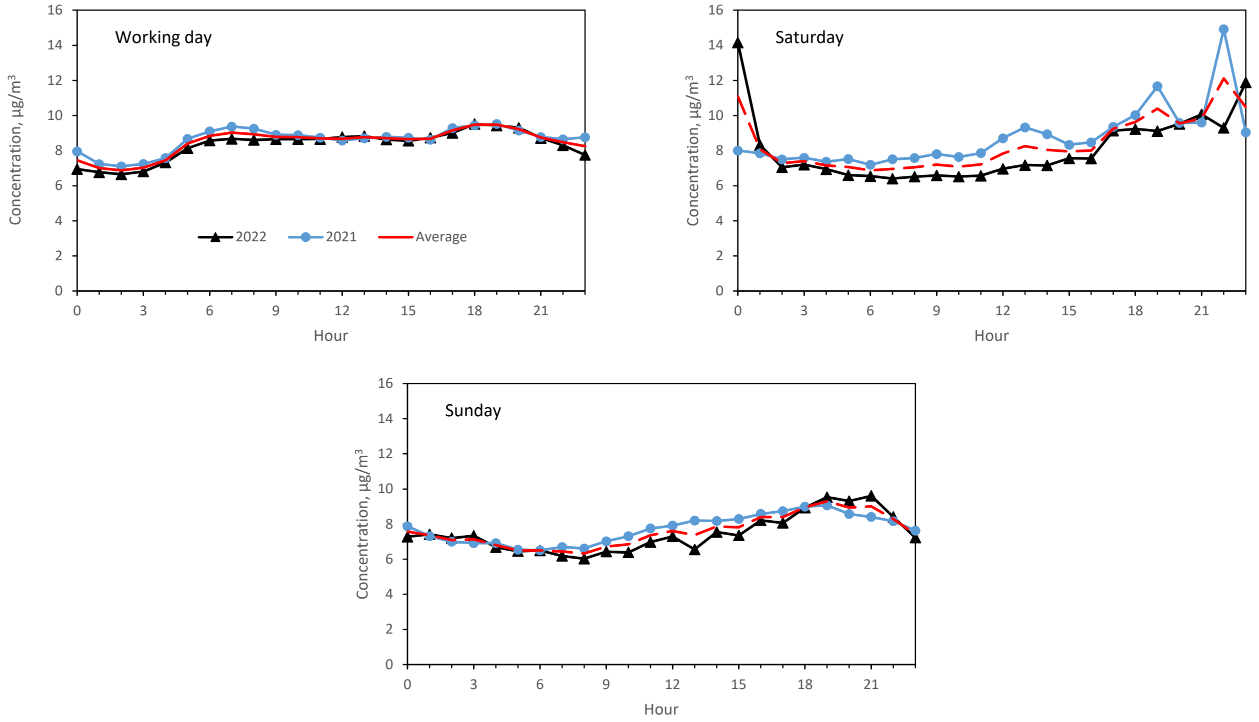

Figure 2.2. Average annual diurnal variations of $PM_{2.5}$ in 2021 (blue)), 2022 (black) and two-year average for 2021 and 2022 (red) at the street station HCAB divided into working days, and Saturdays and Sundays.

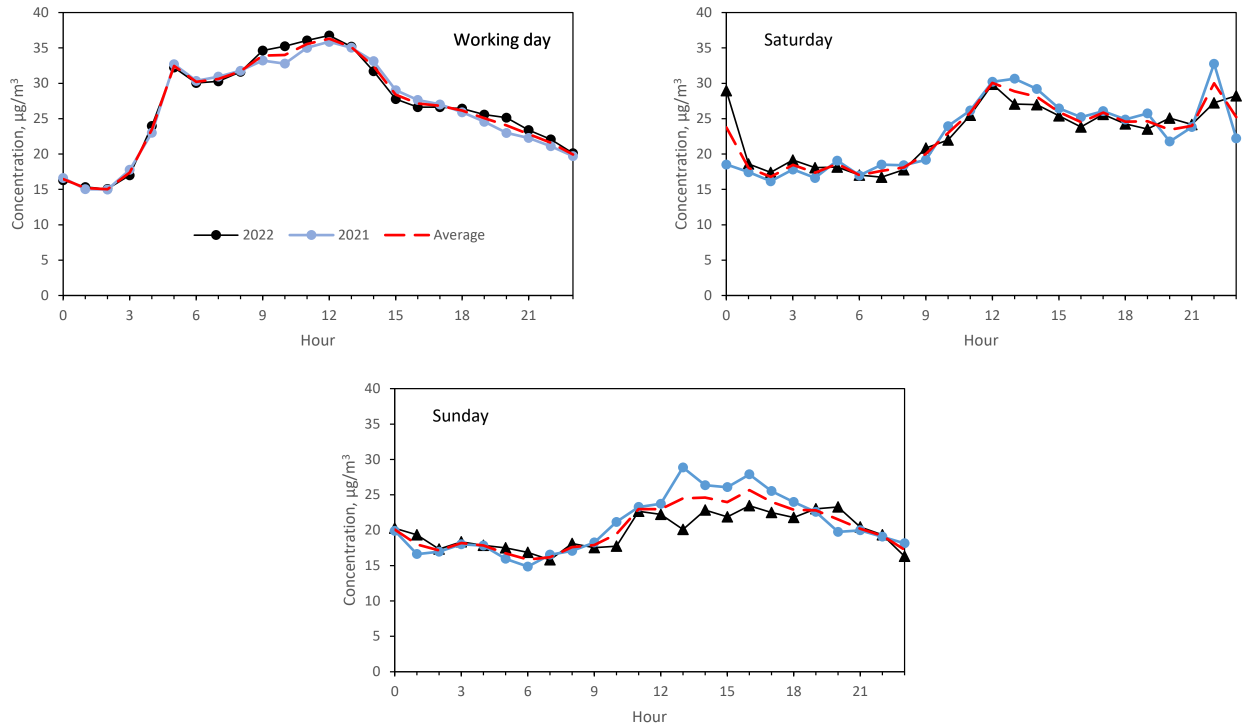

Figure 2.3. Average annual diurnal variations of PM_{10} in 2021 (blue)), 2022 (black) and two-year average for 2021 and 2022 (red) at the street station HCAB divided into working days, and Saturdays and Sundays.

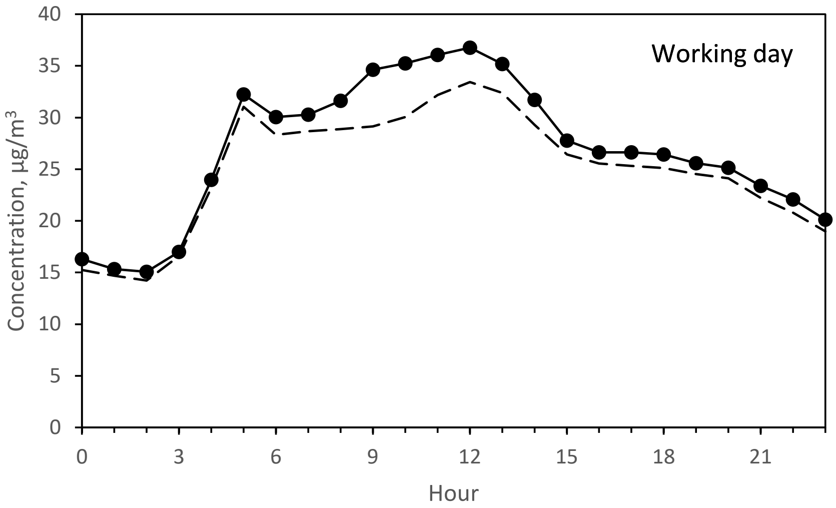

Figure 2.4. Average annual diurnal variations of PM_{10} in 2022 at the street station HCAB for working days. The dashed curve shows the diurnal variations when the episodes in March 2022 has been omitted.

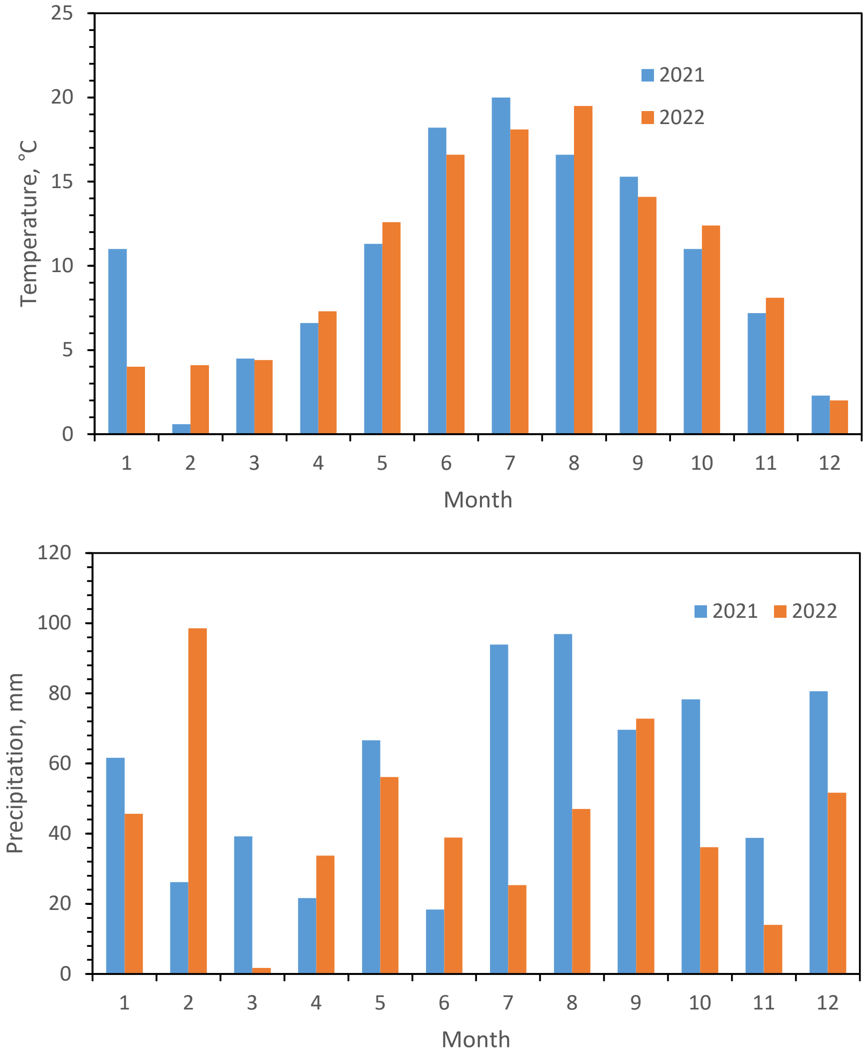

Figure 2.5. Monthy average temperature (above) and total precipitation (below) in Copenhagen Municipality in 2021 and 2022. Data are from DMI (2023).

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) based on a thermal optical technique where the carbon atoms are 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 the street station HCAB, urban background station HCØ, rural background station RISØ and suburban station HVID. 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 in the case of HCØ station. At the three other stations LVS are dedicated to OC/EC sampling and thus the filters for EC determination are not weighted. Punches of the filters (1.48 cm^2) are analyzed for EC using 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. In 2021 the suburban station HVID had to be moved to another location in Hvidovre (roughly 600 m north) and in connection with this, the station was renovated and the new location had to be prepared. This caused a data gap at HVID of about 5.5 months during 2021.

3.1 Long-term trends

Table 3.1 presents the annual averages measured at the four stations in 2022. In 2022, annual mean EC values were measured at the 4 stations in the following ranking: rural 0.18 $\mu g/m^3$ < urban background 0.23 $\mu g/m^3$ < suburban 0.34 $\mu g/m^3$ < urban street 0.52 $\mu g/m^3$ (Table 3.1).

This pattern is consistent with the fact that the highest concentrations are measured at the street locations 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 increase the concentrations at urban background station HCØ with about 28% compared with the rural station RISØ. The street station has the highest concentrations and here the concentrations are 192% higher or nearly three times higher than at RISØ.

Table 3.1. 2022 annual statistics for EC at Danish measurement sites. Annual mean value for EC and annual mean value for EC at the urban background station HCØ, suburban station HVID and street station HCAB relative to the rural background station RISØ (=100%).

	Data coverage %	EC µg/m³	EC relative to rural site (RISØ = 100%)
Street			
HCAB	99%	0.52	292%
Suburban			
HVID	98%	0.34	189%
Urban background			
HCØ	99%	0.23	128%
Rural background			
RISØ	99%	0.18	100%

Figure 3.1 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). Between 2015 and 2022, EC has decreased about 40% (difference of 0.15 µg/m³) at the urban 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 78% (difference 1.9 µg/m³) in the annual average concentrations over the period 2010 to 2022, predominantly due to a reduction in road traffic emissions. This is a result of cleaner 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 about 60% (difference 0.27 µg/m³) at rural background station RISØ over the period 2010 to 2022. At the suburban station HVID there has been a reduction from 2016 to 2022 of about 23% (difference 0.10 µg/m³). Since 2016, the reductions have been quite similar at the urban background, suburban and rural background stations indicating that it is the general reductions on national and international level that explain the main part of the reductions.

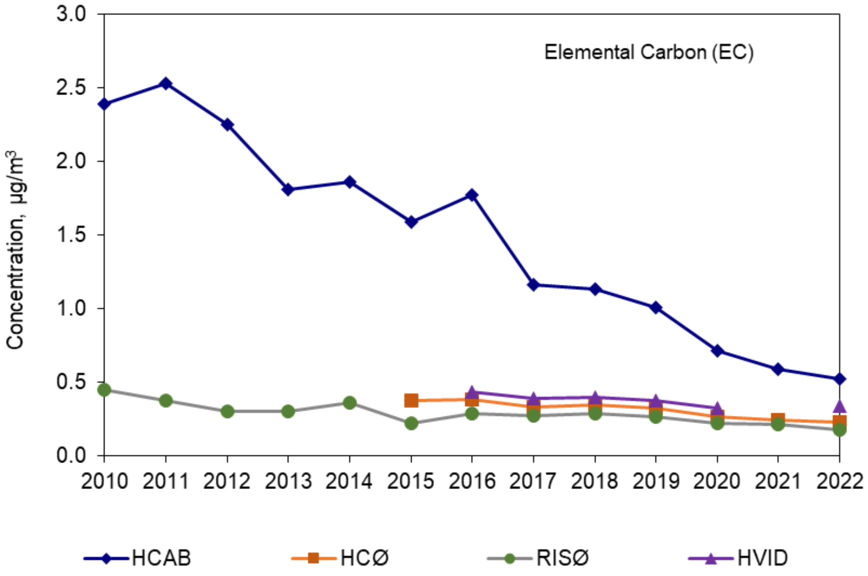

Figure 3.1. Long-term trends in annual average concentrations of EC at urban background station HCØ, suburban station HVID, rural background station RISØ and street station HCAB. There is no annual average for 2021 from the suburban station HVID since it was relocated and this caused a gap in the data.

3.2 Monthly variations in EC

Figure 3.2 shows the measured time series of diurnal averages from all four stations since 2018. 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 with the other three stations. More striking is the clear decreasing trend in EC, which matches the trend in particle number (see next chapter Figure 4.2).

Figure 3.3 shows monthly averages for the four stations in 2022 and Figure 3.4 the monthly variation averaged over the period from 2018 to 2022. 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 both for 2022 and the five years average. Based on five years of measurements at HCØ, mean winter concentrations average 0.34 µg/m³ during December-January-February, while summer concentrations show mean values of 0.22 µg/m³ during June-July-August. Autumn concentrations are slightly higher than spring concentrations, i.e. 0.30 vs. 0.28 µg/m³. Peak concentrations of 1 to 2 µg/m³ are occasionally observed at HCØ (Figure 3.2).

All four stations show higher average concentrations during March compared with February and April. These high monthly concentrations can be explained by the meteorological conditions with nearly no precipitation in March in entire Denmark (DMI, 2023 and Figure 2.5). Low precipitation leads to low removal of particles by wet deposition and hence high concentrations in the air.

High concentrations were also observed for $PM_{2,5}$ and PM_{10} (Chapter 2) and particle number concentration (Chapter 4).

The monthly average concentration in December at the suburban station HVID is unexpectedly high. Especially three days during December had unusually high concentrations most likely due to special meteorological conditions with very low wind conditions and minus degrees (Figure 3.2 and 3.3). These meteorological conditions can lead to enhanced concentrations due to use of wood burning for household heating and low dispersion of the locally emitted EC. In order to investigate the impact from this three day episode, we have calculated the monthly average for December omitting data from 15.12.2022 to 17.12.2022 (Figure 3.5), and it can be seen the monthly average is much lower in December when this episode is excluded. Moreover, the seasonal variation for the second half year in 2022 is in agreement with the pattern observed for the four years average. This indicate that the high monthly average in December 2022 at the suburban station HVID is due to a single episode with unusually high concentrations and not due to a general increase in December due to for example an increase in local emissions from wood burning.

At the street 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 0.79 µg/m^3 vs. summer 0.77 µg/m^3). The relatively low concentrations in July and December are most likely due to the summer and Christmas vacations.

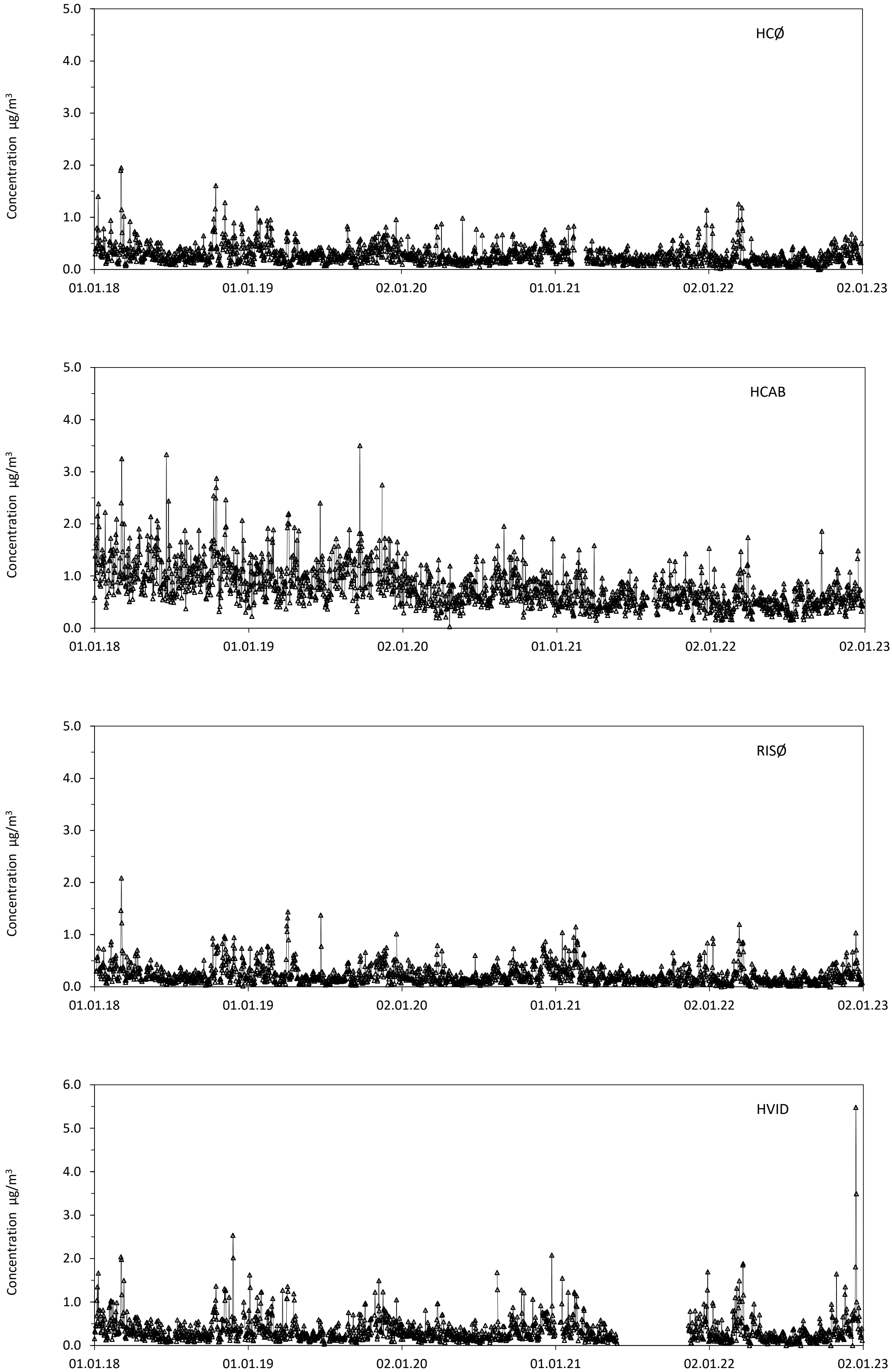

Figure 3.2. Time series for the diurnal average concentrations of EC at urban background station HCØ, street station HCAB, rural background station RISØ and suburban station HVID. The lack of data from the suburban station HVID are due to relocation and renovation of the measurement station.

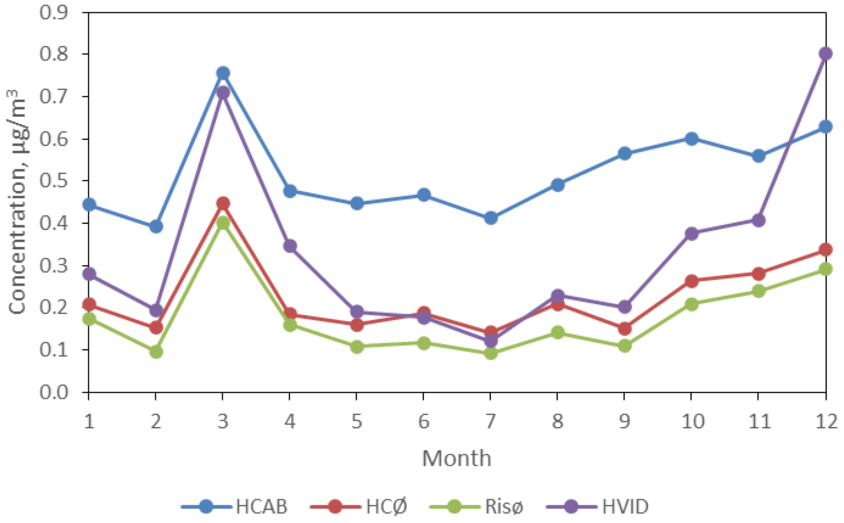

Figure 3.3. Monthly averaged concentrations of EC in 2022 at the urban background station HCØ, suburban station HVID, rural background station RISØ and urban street station HCAB.

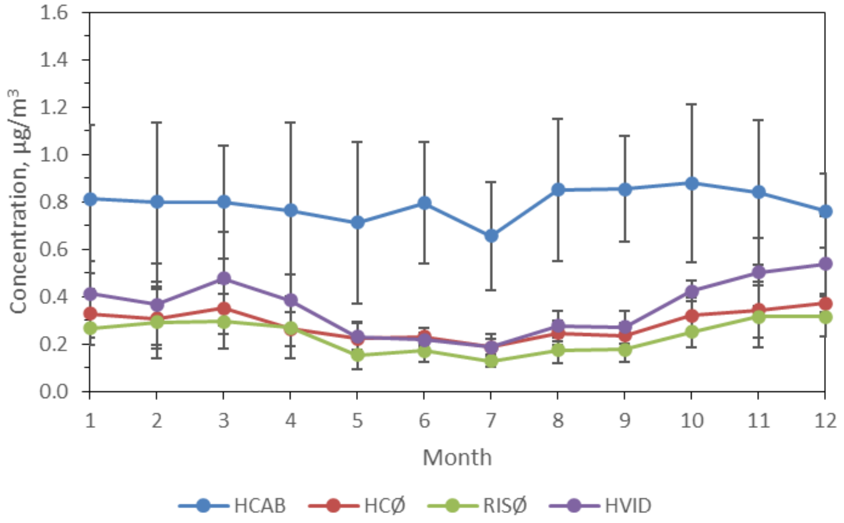

Figure 3.4. Monthly averaged concentrations of EC based on the period 01.01.2018 – 31.12.2022 at the urban background station HCØ, suburban station HVID, rural background station RISØ and urban street station HCAB. The error bars show the standard deviation of the monthly averages for the five years period. The suburban station HVID covers the same period, but without 2021due to data gap in connection with the relocation of the measurement station in the middle of 2021.

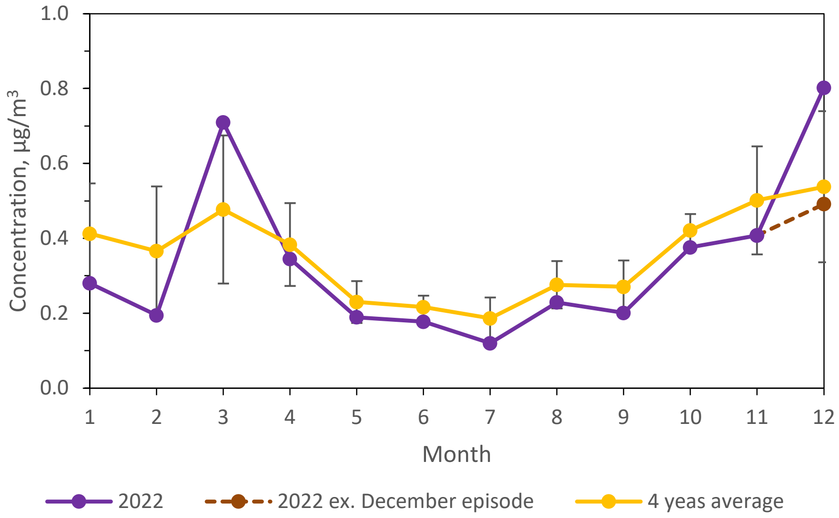

Figure 3.5. Monthly average concentrations of EC from the suburban monitoring station HVID in 2022 excluding the December episode (15:12:2022 to 17:12:2022) with unusual high concentrations of EC and four years average based on 2018 to 2022 excluding 2021.The error bars show the standard deviation of the monthly averages for the four years period.

4. Measurements of particle number concentrations and particle size distribution

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. In our case particles with diameters in the range from 11 nm and up to 478-700 nm (see further details below) were measured. Submicrometer particles originate mainly from combustion processes where emissions from road traffic and household warming using wood burning are the most important sources. Submicrometer particles can also originate from biogenic sources and they can be formed via the chemical and physical processes in the atmosphere based on biogenic and anthropogenic precursors.

Particle number concentrations - similar to other air pollution concentrations - 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 particle dynamics processes as condensation, evaporation, coagulation and cloud processing, leading to changes in the general particle population (also called "aging" of aerosols). A result of this particle aging process 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 in the size range around and smaller than 100 nm in diameter.

Particle number size distributions of diameters 6 - 700 nm were measured at the rural station RISØ (previously Lille Valby), urban background HCØ and street station HCAB. From 2017 and onwards, the instruments at HCAB and RISØ were replaced with commercial instruments delivered by TSI (Model 3938) (Table 4.1). 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). At the suburban station (HVID) an additional new SMPS system of the same type was used since the end of 2015. 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. In 2022, the data coverage was very good for all stations.

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 size range 478 - 550 nm have very little impact on the total particle number. However, the problems with particles in the range 11 - 41 nm, which were observed based on issues with the inlets and DMAs as discussed in the former Particle

Project, were not solved until the beginning of 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 4.1. Operation of instruments (old and new) at the four measurement stations and valid size ranges during the years 2002 to 2022.

	Rural background (RISØ)	**Suburban** (HVID)	**Urban background** (HCØ)	**Street** (HCAB)
Old instrument 11 nm – 550 nm	2005-2016	-	2002-2018	2002-2016
New instrument 41 nm – 478 nm	2017-2019	2015-2019	2019	2017-2019
New instrument 11 nm – 478 nm	2020-2022	2020-2022	2020-2022	2020-2022

Table 4.2. Data coverage in 2022 for SMPS measurements at rural background station RISØ, suburban station HVID, urban background station HCØ and street station HCAB.

	Rural background (RISØ)	**Suburban** (HVID)	**Urban background** (HCØ)	**Street** (HCAB)
Data coverage, %	94	80	98	90

4.1 Particle number size distribution

Figure 4.1 shows the particle number size distribution in the size range from 11 – 478 nm for the four stations in 2022 and for comparison also the results from 2021 are shown. The figure shows the annual average particle number size distributions that are calculated from the measurements with a time resolution of 30 minutes.

It has to be stated, that in general particle numbers measured below 20 nm using typical SMPS instruments that cover large parts 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 size ranges (above 20 nm), SMPS instruments as used in our study give good results with uncertainties, which can be estimated smaller than about 10 – 15 %.

In Figure 4.1 the derivative dN/d(log Dp) is plotted against the logarithm of the particle diameter Dp, which has the large advantage that the area under the curve (numeric integral) 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 with the other three stations (Figure 4.1). This difference is predominantly pronounced for the smallest size fraction of the particles below about 100 nm which are also called ultrafine particles and where emissions from traffic at the street station strongly impact the particle number size distribution and thus also the particle number.

At the street station HCAB, the particle number size distribution looks similar in 2021 and 2022 for particles with diameters above about 25 nm (Figure 4.1). For particles in the size range from 11 to about 25 nm the values are higher in 2022 compared with 2021. Particles in this size range predominantly originate from road traffic. The difference between 2021 and 2022 is most likely mainly due to the natural variations in the meteorological conditions from year to year.

At the urban background station HCØ and the rural background station RISØ the particle number size distributions look similar when comparing the years 2021 and 2022. Nevertheless, we see a consistent increase in ultrafine particles in the nucleation mode (~ 10 - 30 nm) as well as in the Aitken mode (~ 30 - 80 nm) size range. These differences between 2021 and 2022 are most likely related to the natural variations in the meteorological conditions (Chapter 4.4).

At the suburban station HVID the particle number size distribution for 2022 differs compared with 2021 in much the same way as for HCØ and RISØ. However, since the data coverage is very low the particle number size distribution is very uncertain and it is therefore not possible to make a proper evaluation of the differences between 2021 and 2022.

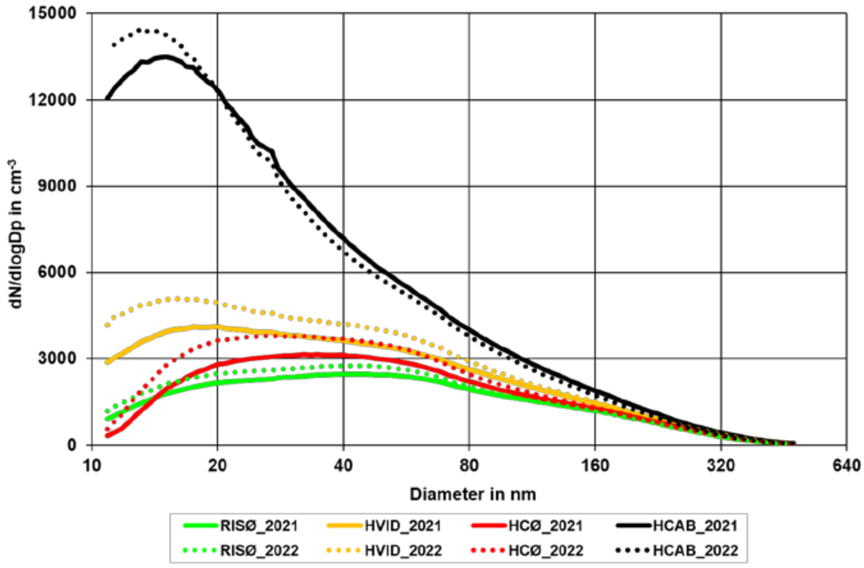

Figure 4.1. Annual average of particle number size distributions at the rural background station RISØ, suburban station HVID, urban background station HCØ and street station HCAB during 2022 compared to 2021. Note the logarithmic x-axis. HVID in 2021 is uncertain due to low data coverage.

4.2 Long-term trends for particle number fractions

For a more detailed analysis of the long-term trends, particle number concentrations were determined in specific size regimes. In this report, we have divided particles in three size fractions:

- Small size fraction: Diameters between 11 – 41 nm (both DMPS and SMPS),
- Medium size fraction: Diameters between 41 – 110 nm (both DMPS and SMPS),
- Large size fraction: Diameters between 110 – 550 nm for the old instruments (DMPS) and diameters between 110 – 478 nm for the new instruments (SMPS). Table 4.1 gives the details on the type of instruments used at the different stations.

Figure 4.2 shows the long-term trends based on annual averages for the three size fractions. Because of previously stated problems with inlets and DMAs, data for the small size fraction (Dp = 11 – 41 nm) are missing for the years 2017 – 2019 (RISØ and HCAB), 2019 (HCØ), and 2015 -2019 (HVID) for the new instruments. The change between the old and new instruments is visible in the graphs by an interruption of the connection line between the symbols. In addition, data are missing for 2021 at the suburban station HVID due to the low data coverage in connection with the relocation of the station.

For HCAB, the highest particle number is typically found in the smallest size range 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 range have been significantly decreasing to about 50%, which is a result of technological development of vehicle engines and changes in fuel composition (Figure 4.2).

No clear trend can be observed for the small size fraction at the rural background station RISØ, the suburban station HVID and the urban background station HCØ. This may be explained by the fact that one of the main sources of the small particles (11 to 41 nm) at these background locations are natural emissions of precursors and formation of particles through atmospheric processes and only to a minor extent do these particles come from anthropogenic sources. Nevertheless, this number fraction does show a slight increase from 2021 to 2022 as discussed above.

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 reducing 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 site up to 2016, and at the rural measurement site up to 2012, HCØ and RISØ, respectively, but the particle numbers have been stabilizing over the past five years at HCØ and the past ten years at RISØ. Similarly, this finding is observed at the suburban station HVID, where concentrations seem to be stable during the observational period which is much shorter compared to other stations. At HVID, trends can only be investigated in more detail when longer time series are available.

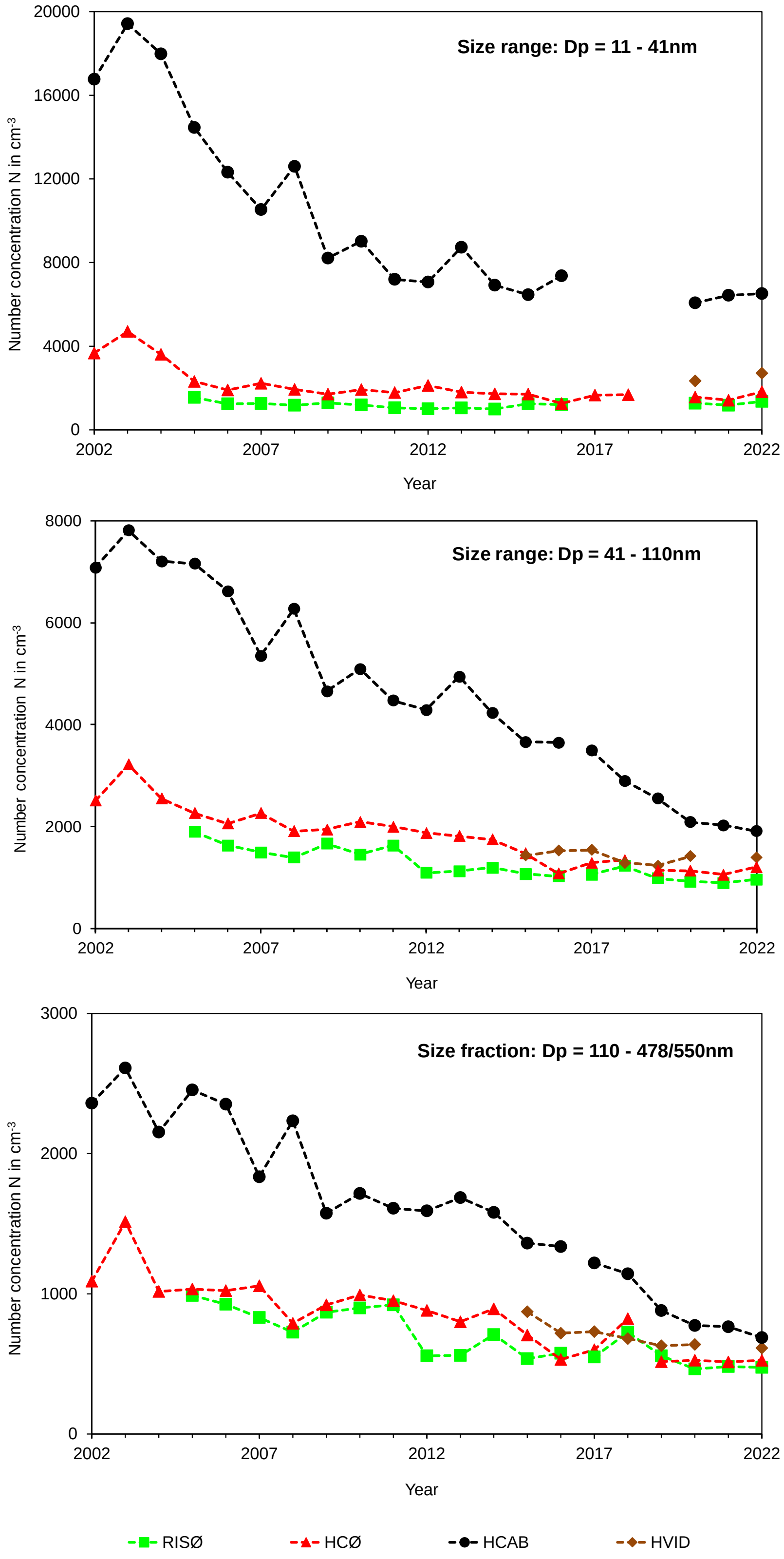

Figure 4.2. Annual mean particle number concentrations in specific size ranges Dp = 11 - 41 nm (upper Figure), Dp = 41 – 110 nm (middle Figure), and Dp = 110 - 478/550nm (lower Figure) combined for old and new instruments at the rural background station RISØ, suburban station HVID, urban background station HCØ and street station HCAB during 2002 to 2022. 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 is only available for 2020 and 2022.

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 2022. Similarly, such a trend is observed at HCØ, HVID, and RISØ except for the past four to five years. Nevertheless, some variations are observed for some years which may be linked to the natural meteorological variations. In general, decreasing trends are expected as the background aerosol is affected by general emission decreases in Europe and worldwide because of emission regulations. This implies that especially at the street station similar trends as for the smaller size regimes are observed. On the other hand, also natural processes result in relatively stable values over shorter 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 periods, but may also change because of changing climate over larger periods as e.g. decades. In addition, the strength of these sources can vary from year to year because of variations in meteorological conditions. These freshly formed particles will after some atmospheric transformation as condensation, coagulation and cloud processing enter into the large size regime where their lifetimes in the atmosphere is longer. It is possible, that at the suburban station concentrations in the smaller, middle and large size regime are strongly influenced by wood combustion emissions in the area. This could explain higher concentrations compared with the urban background and the rural stations in all three size regimes.

4.3 Long term trend for total particle number

In Figure 4.3 the total particle number is presented for the rural background station RISØ, the suburban station HVID, the urban background station HCØ and the street station HCAB in the corresponding size ranges for the old instruments (Dp = 11- 550 nm) and for the new instruments (Dp = 11- 478 nm).

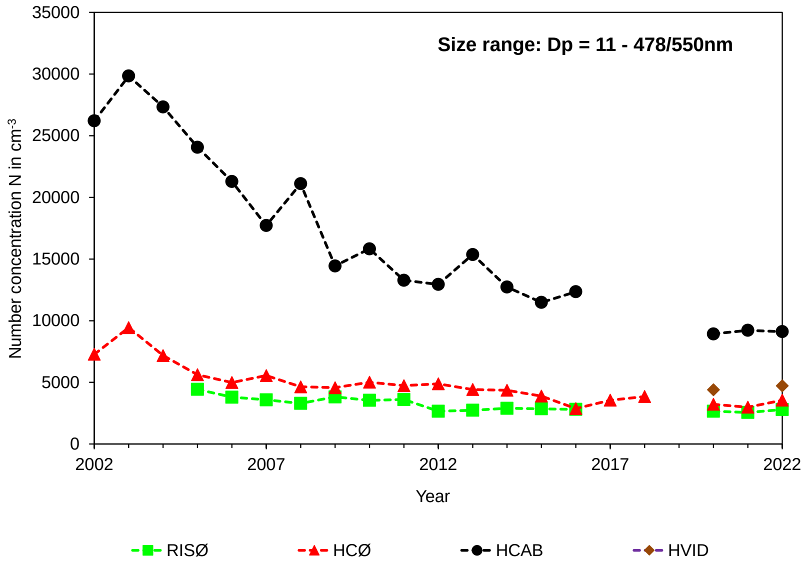

Figure 4.3. Annual mean particle number concentrations for the total size range Dp = 11 – 550 nm (old instruments) and Dp = 11 – 478 nm (new instruments) combined at the rural station RISØ, suburban station HVID, urban background station HCØ and street station HCAB during 2002 to 2022. Note that values were not available at RISØ and HCAB from 2017-2019, HCØ in 2019 and at HVID only data from 2020 and 2022 are available.

The general patterns in terms of annual trends reflect the picture that is shown in detail in Figure 4.2 (upper, middle, lower panel). An overall decline in total submicrometer particle number is observed at all three stations RISØ, HCØ, and HCAB except for the past few years. The particle number has since 2002 declined about 70% and 60% at HCAB and HCØ, respectively. At RISØ the decrease is about 34% since the beginning of these measurements in 2005. There is a tendency to a smaller decrease with the distance to anthropogenic sources of submicrometer 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 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. This is similar at the suburban background where the data series is too short to make such judgements. Finally, the largest decrease is observed at the urban street 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 street locations.

4.4 Monthly variations in particle number concentrations

Figure 4.4 shows the monthly average concentrations for the particle number concentrations in 2021 and 2022 at the urban background station HCØ, rural background station RISØ, street station HCAB and suburban station HVID. The suburban station had to be relocated in 2021 and hence there is only data from seven months in 2021. The graphs show the monthly averages for the full particle range measured with the instruments (11 - 478 nm) and for the small size fraction (diameters between 11 – 41 nm), medium size fraction (diameters between 41 – 110 nm) and large size fraction (diameters between 110 – 478 nm).

At the urban background station HCØ the highest particle number concentration is observed during the summer half year with a maximum peak of about 4000 particles per cm^3 in June in 2021 and about 4800 particles per cm^3 in August in 2022 for particles in the size range from 11 - 478 nm. The average particle number concentration is 22% and 33% lower in the winter half year than the summer half year for 2021 and 2022, respectively. This seasonal variation is also seen for the small and medium size fractions while the large size fraction shows approximately equal particle number concentrations for the winter and summer half of the years.

The emissions of the small size fraction originate mainly from road traffic and wood combustion in Copenhagen. Based on these sources it would have been expected that the particle number concentrations would have been higher during winter than summer or at equal level. This is because wood combustion for household warming is strongly related to the winter half year and road traffic has only a slight seasonal variation with traffic being about 12% lower in summer in 2021 and 7% lower in 2022 (Vejdirektoratet, 2023). The stronger seasonal difference of road traffic in 2021 compared with 2022 is most likely due to the Covid-19 restrictions during the first part of 2021 (Figure 4.6).

In addition to this, meteorology leads on average to higher concentrations during winter than summer due to lower turbulent mixing of the air during the colder winter half of the year compared with the warmer summer half of the year.

The higher particle number concentration during summer is most likely due to a significant contribution from particles formed in the atmosphere by photochemical processes initiated by sunlight. Since these processes requires sunlight they occur mainly during the summer half year where there is much more sunlight available than during the winter half year (Figure 4.5).

The monthly variations observed at the urban background station HCØ is to a large extend due to the natural variations in meteorology where precipitation (Figure 4.5) is one of the main factors because wet deposition "washes" particles out of the air. The higher monthly average particle number concentration in February 2021 compared with February 2022 is most likely explained by the difference in precipitation between the two years (Figure 4.5). As mentioned above, solar light has also an important impact on the particle number concentrations due to the photochemical production of particles during summer. The high number of sunlight hours in August 2022 compared with August 2021 can be the explanation for the higher monthly average particle number concentration seen in August 2022 compared with August 2021. However, there is no simple relationship between particle number concentrations, precipitation and solar light since wind speed and direction, humidity and temperature also have significant impact on the processes that determine the amount of airborne particles transported and formed at the site.

The monthly variations observed at the rural background station RISØ follow in general the same pattern as at the urban background station HCØ for both 2021 and 2022. Moreover, the levels are only about 15% and 30% lower at RISØ for the particle size range 11 - 478 nm in 2021 and 2022, respectively. The largest difference is seen for the small size fraction and the least difference for the large size fraction. The similarities between HCØ and RISØ show that to a large extent it is the same sources that are responsible for the observed levels of particle number concentrations and that both stations experience similar meteorological conditions when compared on monthly basis.

The particle number concentration is about three times higher at the street station HCAB than at the urban background station HCØ and the monthly variations show a very different pattern with less difference between the levels during the winter (January-March and October-December) and summer half year (April – September). The particle number concentrations were on average 2% and 17% higher in summer compared with winter in 2021 and 2022, respectively. Moreover, it is especially the small size fraction (11-41 nm) that is considerably higher at HCAB (about 450% higher) while the large size fraction (110-478 nm) is only about 50% higher. These observations are in line with the large impact that emissions from road traffic have on the particle levels at busy streets. Road traffic leads to high emissions of particles in the small size fraction (Wåhlin et al., 2009) and the seasonal variation in traffic intensity is small (Figure 4.6, Vejdirektoratet, 2023). Hence, there is no large seasonal variation in the local traffic emissions and consequently there is no clear seasonal variation in the particle number concentrations due to variations in the most important emissions. Moreover, it is the meteorological variations that are the main reason for the month to month variations in the particle number concentrations.

The Covid-19 restrictions were not very strict in 2021 and based on the road traffic index the annual traffic in 2021 was only 2% lower in 2021 compared with 2022 when the traffic index was at the same level as 2019 before the Covid-19 pandemic (Vejdirektoratet, 2023). On an annual basis, the impact of the Covid-19 restrictions is very limited. However, the lower traffic during the first months of 2021 compared with 2022 may have some minor impact on the monthly average particle number concentrations since the contribution to the particulate air pollution from traffic will be slightly lower during the first part of 2021 compared with 2022 (Figure 4.6). However, as discussed above, the high precipitation in February 2022 is also an important factor and the natural variations in meteorology are most likely the main reason for the difference in the monthly average particle number concentrations between 2021 and 2022.

The monthly variations of the particle number concentrations in 2022 at the suburban station HVID follow in general the same patterns as for the urban background station HCØ and the rural background station RISØ. Some of the marked differences are that particle number concentrations are higher than at the two other stations and that it is the small size fraction (11-41 nm) that shows the largest difference compared with HCØ and RISØ. Interestingly, the suburban station HVID shows the same patterns with significantly higher concentrations during the summer half year compared with the winter half year. This is interesting since the suburban station is placed in a residential area where wood combustion is used for household heating. It seems therefore that the atmospheric formation of particles during the summer half year has a stronger influence on the particle number concentrations than wood combustion at this location during winter. Since there were only data for 6.5 months from the suburban station HVID in 2021 it is not possible to make a full comparison between 2021 and 2022. However, it might be noted that for November and December data indicates that the concentrations are at the same level in 2021 and 2022. At the other three stations, the difference between November and December in 2021 and 2022 is also small. This suggests, that the high energy prices in the second half of 2022 did not lead to large increases in particle number concentrations due to increase in wood burning for house hold warming. This aspect will have to be followed up when data for 2023 are available.

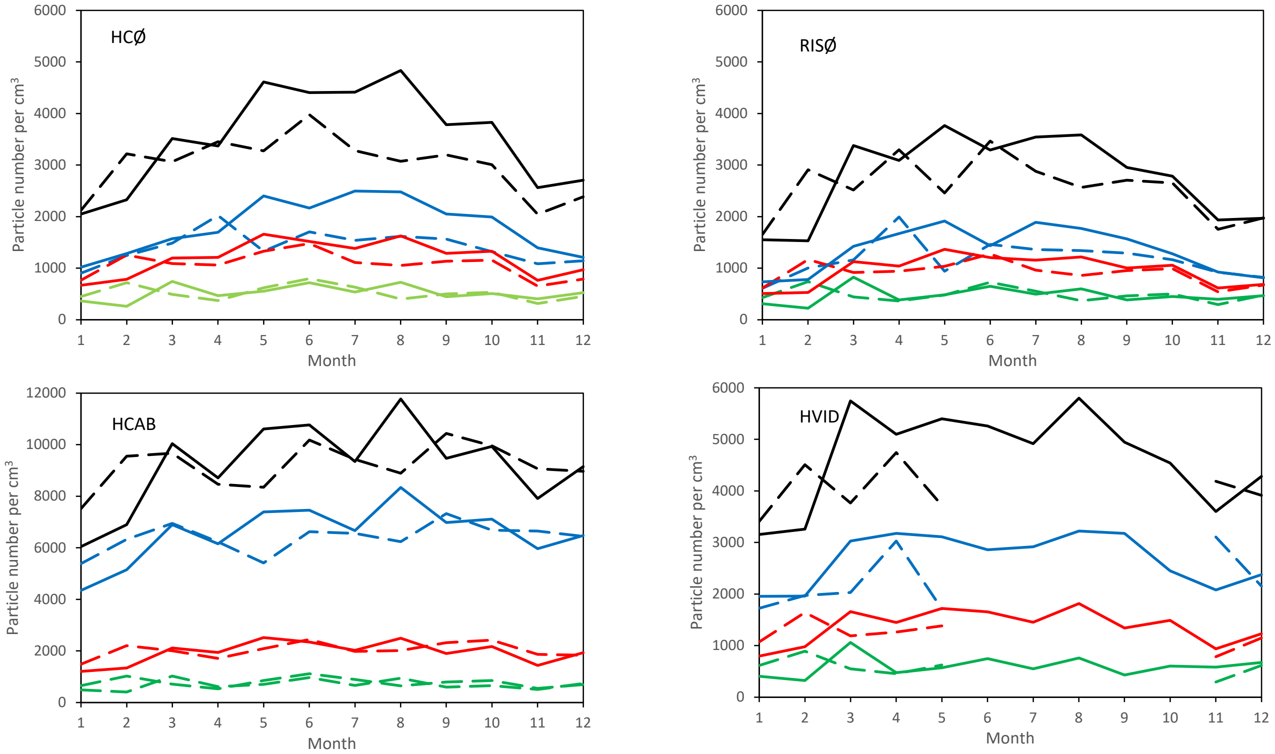

Figure 4.4. Monthly variation in the particle number concentration in 2021 (dashed lines) and 2022 (solid lines). The graphs show the full particle size range measured with the instruments (black, 11 - 478 nm), the small size fraction (blue, 11 – 41 nm), medium size fraction (red, 41 – 110 nm) and large size fraction (green, 110 – 478 nm).

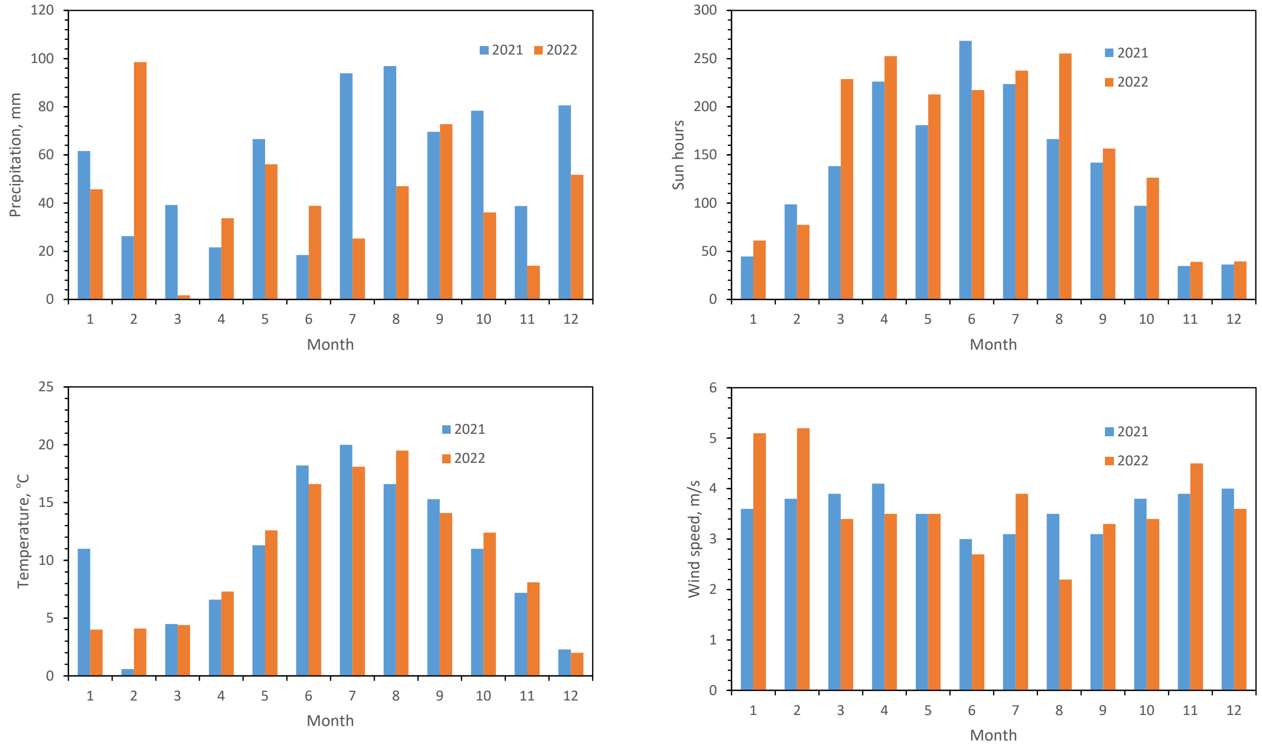

Figure 4.5. Monthly precipitation, hours with sunshine, average temperature and wind speed in Copenhagen Municipality during 2021 and 2022 (DMI, 2023).

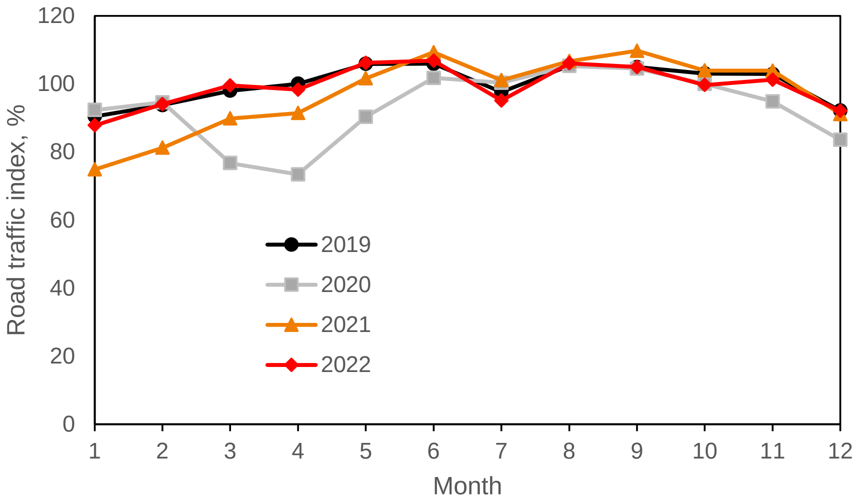

Figure 4.6. Monthly road traffic index in Denmark for 2019 - 2022 (Vejdirektoratet, 2023). The average annual road traffic index for 2019 is set to 100%. 2020 and 2021 is clearly impacted by the Covid-19 restrictions while 2019 was the last normal year before the Covid-19 restrictions. 2022 is very close to the normal conditions before Covid-19.

4.5 Diurnal variations in particle number concentrations

Figure 4.7 to 4.10 show the average diurnal variations of particle number concentrations for the four stations in 2021 and 2022. As for $PM_{2.5}$ and PM_{10} the data have been divided into working days, and Saturdays and Sundays and in addition to this they have been split into winter (January-March and October-December) and summer half year (April-September). The aim of this study is to analyze the differences between the warm and cold seasons in order to see for example the impact from wood combustion from household warming and the photochemical formation of particles. For the suburban station HVID there are no data for the summer half year from 2021 due to relocation of the station and consequently a lack of data for a large part of the summer half year. The most important findings from the analysis of the diurnal patterns are as follows:

The overall patterns for the average diurnal variation are quite similar in 2021 and 2022. The most marked difference is that the noon to afternoon peak during the summer half year is higher in 2022 compared with 2021 at the urban background station HCØ and the rural background station RISØ.

All stations show lowest particle number concentrations during the late night.

On the working day mornings, there is a clear peak that can be attributed to rush hour traffic. The morning rush hour peak (5:00 - 9:00) is seen both in winter and summer and it is shifted to one hour earlier during summer because of summertime. This is because the instruments always are operated on Danish wintertime. This peak is most pronounced at the street station HCAB and for the small size fraction. The morning rush hour peaks are not seen on Sundays and Saturdays.

The noon and afternoon patterns differ significantly between stations, season and day of the week. At the urban background station HCØ, the particle number concentrations show a morning peak corresponding to the morning rush hour traffic. During winter, the particle number concentrations decrease to relatively stable values over the day until the particle number concentration begins to increase slowly during the late afternoon and reaches an early evening maximum around 19:00. During summer, the particle number concentrations are higher than during winter and the particle number concentrations begin to increase from noon and reach a local maximum during the early afternoon. The same pattern is also seen, at the rural background station RISØ and here the early afternoon peak is even more clearly seen. Moreover, roughly the same level of particle number concentrations are observed on Saturdays and Sundays as on working days at RISØ. This early noon afternoon summer peak can best be explained by photochemically formed particles. The higher noon-afternoon peak in 2022 compared with 2021 shows that the increase in annual averages at the urban background station HCØ and rural background station RISØ (Chapter 4.2) are mainly due to an increase in the photochemically formed particles and hence for a large part due to the natural variations in the meteorological conditions.

At the street station HCAB the influence from road traffic is strong. On working days, the particle number concentrations reach a stable plateau around noon and begin slowly to decrease during the end of the afternoon. This is seen both in summer and in winter. There is no distinct afternoon rush hour peak because the afternoon rush hour is spread out over longer periods and in addition there is a higher turbulent dilution of the surface air during the late afternoon compared with the morning. Photochemically formed particles will also contribute to the particle population at HCAB but the diurnal variation is dominated by the emissions from road traffic.

On Saturdays and Sundays at the street station HCAB, there is an increase in particle number concentrations over the day reflecting that traffic activities especially in the city begin later in weekends. On Saturdays, the highest concentrations are reached around 18:00 - 19:00 during winter and around 21:00-22:00 during summer reflecting the road traffic in connection to the higher leisure activities in summer compared with winter. Activities related to Tivoli may also add to this observation because a late evening pronounced peak in particle number is observed at 22:00. This peak can best be explained by the fireworks in Tivoli, performed every Saturday from June to October. Sundays do not show as high particle number concentrations during the evening as observed on Saturdays.

At the urban background station HCØ and the regional background station RISØ, there are also high particle number concentrations during the working and Saturday evenings. At HCØ the highest particle number concentrations are observed during the Saturday summer evenings. These high summer evening particle number concentrations are most likely due to road traffic in relation to the leisure activities during evenings in combination with the remainings of the photochemically formed particles.

During winter, quite high concentrations are observed during the late afternoon and evening at most of the stations. Especially at the suburban station HVID, the highest particle number concentrations are seen during the evening and on Saturdays and Sundays. These high particle number concentrations at winter evenings are most likely due to use of wood burning for household warming at this suburban site.

Figure 4.7. The diurnal variation of the particle number concentrations in 2021 (dashed line) and 2022 (solid line) at the urban background station HCØ. The diurnal variation is devided in winter (blue) and summer (red) half years and in working days (left column), Saturdays (middle column) and Sundays (right column). The upper row shows results for the full range (11-478 nm), the second row the small size fraction (11-41 nm), the third row the medium size fraction (41-110 nm) and the fourth row the large size fraction (110-478 nm).

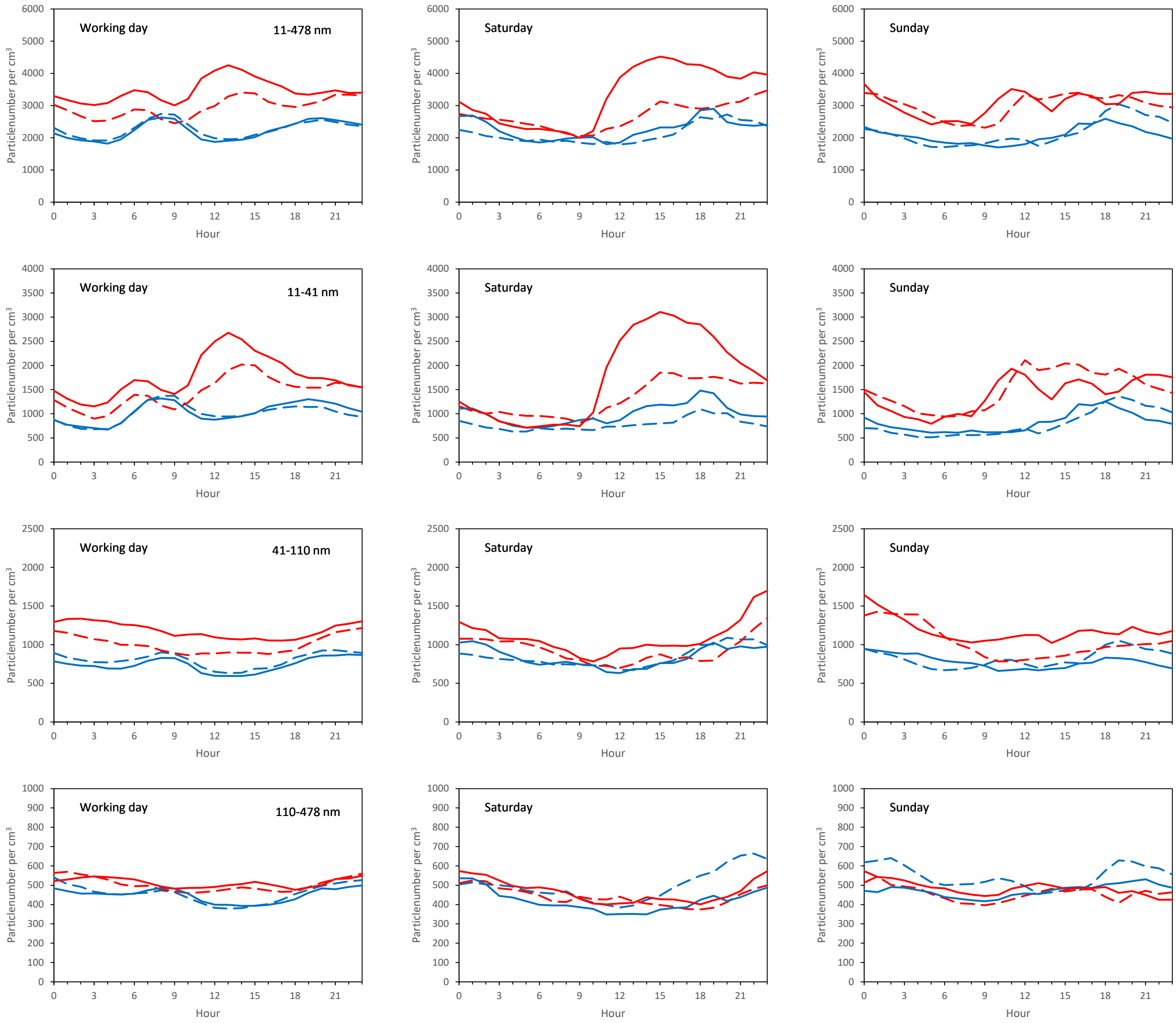

Figure 4.8. The diurnal variation of the particle number concentrations in 2021 (dashed line) and 2022 (solid line) at the rural background station RISØ. The diurnal variation is devided in winter (blue) and summer (red) half years and in working days (left column), Saturdays (middle column) and Sundays (right column). The upper row shows results for the full range (11-478 nm), the second row the small size fraction (11-41 nm), the third row the medium size fraction (41-110 nm) and the fourth row the large size fraction (110-478 nm).

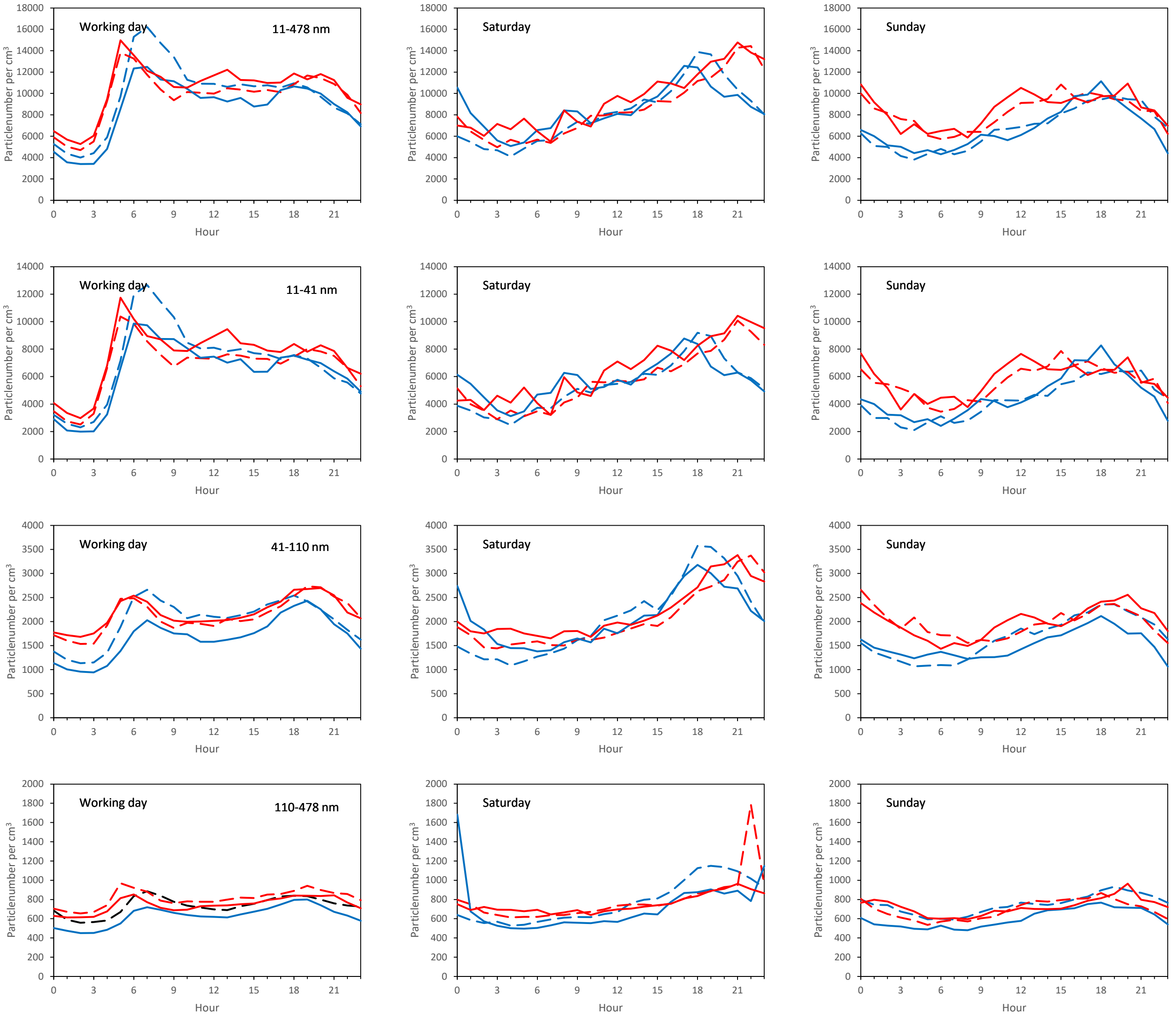

Figure 4.9. The diurnal variation of the particle number concentrations in 2021 (dashed line) and 2022 (solid line) at the street station HCAB. The diurnal variation is devided in winter (blue) and summer (red) half years and in working days (left column), Saturdays (middle column) and Sundays (right column). The upper row shows results for the full range (11-478 nm), the second row the small size fraction (11-41 nm), the third row the medium size fraction (41-110 nm) and the fourth row the large size fraction (110-478 nm),

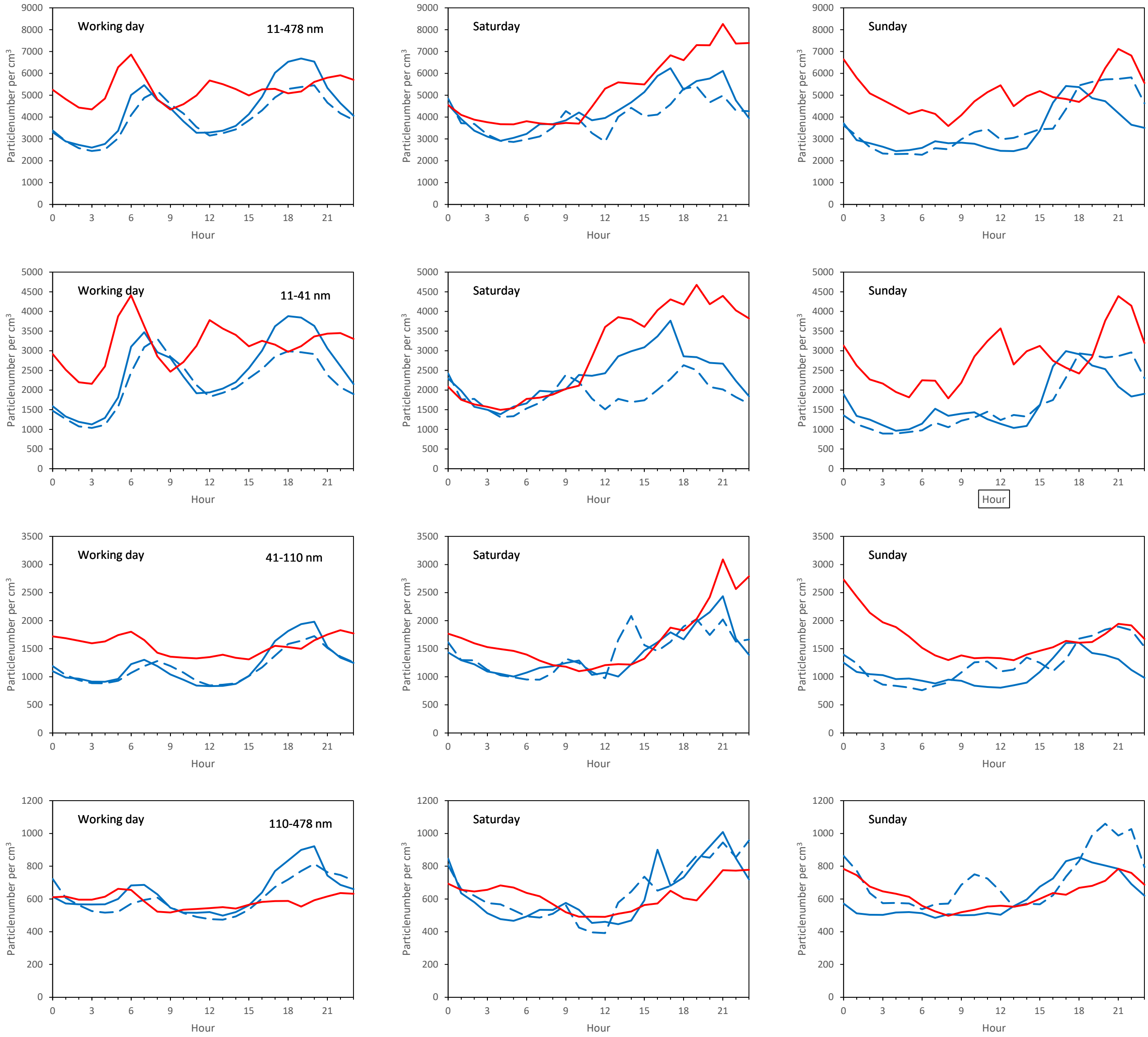

Figure 4.10. The diurnal variation of the particle number concentrations in 2021 (dashed line) and 2022 (solid line) at the suburban station HVID. The diurnal variation is devided in winter (blue) and summer (red) and in working days (left column), Saturdays (middle column) and Sundays (right column). The upper row shows results for the full range (11-478 nm), the second row the small size fraction (11-41 nm), the third row the medium size fraction (41-110 nm) and the fourth row the large size fraction (110-478 nm). Due to relocation of the station in 2021 there are no data for the summr half year in 2021.

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THE PARTICLE PROJECT 2022

The Particle Project 2022 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 street, 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 temporal variations of the particulate air pollution.

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