



# THE PARTICLE PROJECT 2019

Scientific Report from DCE - Danish Centre for Environment and Energy

No. 385

2021



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DCE - DANISH CENTRE FOR ENVIRONMENT AND ENERGY



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# Data sheet

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Abstract:	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 <sub>2.5</sub> and PM <sub>10</sub> 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.
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# Contents

<b>Abbreviations and definitions</b>	<b>5</b>
<b>Preface</b>	<b>6</b>
<b>Sammenfatning</b>	<b>7</b>
<b>Summary</b>	<b>9</b>
<b>1 Measurements</b>	<b>10</b>
1.1 Particle number	10
1.2 Particle mass concentration using TEOM	14
1.3 Elemental carbon (EC) mass concentration	18
<b>References</b>	<b>21</b>



## 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 <sub>2.5</sub>	Particles less than 2.5 micrometers in diameter, i.e. fine particles.
PM <sub>10</sub>	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.

## Preface

The Particle Project 2019 provides monitoring of submicron particles (number and size distribution) and Elemental Carbon (EC) in fine particles at urban background, which is a supplement to the National Air Quality Program under NOVANA. Likewise TEOM PM<sub>10</sub> and PM<sub>2.5</sub>, i.e. particles with diameter less than or equal to 10  $\mu\text{m}$  and 2.5  $\mu\text{m}$  in high time resolution, are also included to illustrate the trends in atmospheric particle mass concentration.

## Sammenfatning

I *The Particle Project 2019* rapporteres resultater af målinger af partikelstørrelsesfordelingen i intervallet op til 1  $\mu\text{m}$ , 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 landlig station nord for Roskilde (RISØ), en station i den Københavnske forstadsby Hvidovre (HVID), samt fra en station i trafikeret gade i København (HCAB). Herudover rapporteres udviklingstendenser for  $\text{PM}_{2,5}$  og  $\text{PM}_{10}$  fra målinger med høj tidsopløsning.  $\text{PM}_{2,5}$  og  $\text{PM}_{10}$  angiver koncentrationen af partikler med diameter mindre end hhv. 2,5 og 10  $\mu\text{m}$ .

Langtransporterede partikler og deres gasformige forstadier bidrager til partikelantal for partikler med en diameter mindre end 1  $\mu\text{m}$ . 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). Fra 2018 til 2019 aftog partikelantallet på alle stationer, men kun HCAB og HVID udviste et gennemgående fald fra 2017 til 2019, hvilket omfatter både en andel af de ultrafine partikler (her defineret som partikler fra 41 – 110 nm) og andelen fra 110 – 478/550 nm (den øvre grænse afhænger af instrumenttypen). Et mindre konsistent billede ses for RISØ og HCØ, hvor partikelantallet i 2018 oversteg det målte i både 2017 og 2019. Det skal dog bemærkes, at 2018 meteorologisk set var usædvanlig tør. Man havde ca. 40% mindre nedbør i løbet af sommeren 2018 end gennemsnittet for de tilsvarende sommerperioder i 2017 og 2019 (DMI, 2020). Denne forskel i nedbørsmængder har stor betydning for afsætningen af atmosfæriske partikler og dermed for luftens indhold af partikler.

Når man ser på partikelmasse i form af  $\text{PM}_{2,5}$  og  $\text{PM}_{10}$  målt med høj tidsopløsning, så viser resultaterne aftagende årsmiddelværdier på HCAB mellem 2017 og 2018, og ligeledes mellem 2018 og 2019. Aftagende årsmidler er ligeledes målt på den landlige lokalitet RISØ og gadestationen i Danmarks næststørste by Aarhus. Den geografiske fordeling for  $\text{PM}_{10}$  udviste et stort span, hvor årsgennemsnittet på HCAB ( $27 \mu\text{g}/\text{m}^3$ ) var dobbelt så højt som på RISØ ( $13 \mu\text{g}/\text{m}^3$ ). Den høje tidsopløsning i data er udnyttet i forbindelse med analyse af langtransport og ugentlig variation i trafikens bidrag til partikelmasse på HCAB.

Årsmiddel af EC-koncentrationen på HCØ udgjorde  $0,32 \mu\text{g}/\text{m}^3$  i 2019. Den gennemsnitlige koncentration af EC i 2019 var lavest på den landlige station RISØ. EC var 22% højere på HCØ og 43% højere på HVID, dvs. koncentrationen af EC var højere på forstadsstationen end på den Københavnske bybaggrundsstation. På den Københavnske gadestation HCAB var EC koncentrationen  $1,0 \mu\text{g}/\text{m}^3$  eller omkring en faktor 4 højere end på RISØ. EC årsmiddelkoncentrationerne ændrede sig 3-6% på stationerne RISØ, HCØ og HVID

mellem 2017 og 2019, mens der blev målt et 13% fald på HCAB mellem 2017 og 2019.

## Summary

The *Particle Project 2019* reports series of particle size distributions and Elemental Carbon (EC) in fine particles with diameter smaller than  $2.5\ \mu\text{m}$  ( $\text{PM}_{2.5}$ ) in urban background. 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). Furthermore, trends of measurements with high temporal resolution of  $\text{PM}_{2.5}$  and atmospheric particles with diameter smaller than  $10\ \mu\text{m}$  ( $\text{PM}_{10}$ ) in rural and urban locations are compared.

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 and smaller relative contributions at other locations that are more influenced by local sources. For these reasons, long-range transported particles make up a smaller relative fraction at the suburban site (HVID) and urban background site (HCØ) and an even smaller fraction at the urban curbside station (HCAB), where highest particle number concentrations are measured due the significant contributions from local traffic. A trend of decreasing concentrations is observed at all stations when considering time periods of the order of a decade. From 2018 to 2019, particle number concentrations decreased at all stations. Decreasing trends from 2017-2018-2019 were found for HCAB and HVID, whereas a less consistent trend characterized RISØ and HCØ, where concentrations in 2018 exceeded those in 2017 and 2019. These changes were observed for both a fraction of the ultrafine particles (41 - 110 nm in this study) and for particles in the range 110 - 478/550 nm. It should be noted that 2018 was an unusually dry year. The summer precipitation was thus about 40% lower than the similar summer precipitation in 2017 and 2019.

With respect to high temporal resolution measurements of particulate mass, both  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  decreased at the urban curbside (HCAB) between 2017 and 2018 and further between 2018 and 2019. Likewise, the  $\text{PM}_{10}$  concentrations were observed to decrease between 2017 and 2018 and further between 2018 and 2019 at curbside in the second largest Danish city Aarhus as well as at rural site RISØ. The  $\text{PM}_{10}$  particle mass at HCAB ( $27\ \mu\text{g}/\text{m}^3$ ) is in 2019 doubled of what is found at the rural location RISØ ( $13\ \mu\text{g}/\text{m}^3$ ). Analyses of PM data with high temporal resolution is used to determine contribution from long-range transport and to analyze weekly variation, which is the result of the variation in traffic at the Copenhagen curbside.

The annual mean EC concentration at HCØ in 2019 was  $0.32\ \mu\text{g}/\text{m}^3$ , which was 22% higher than at the rural site RISØ ( $0.26\ \mu\text{g}/\text{m}^3$ ). The annual mean EC concentration at the suburban location HVID was 43% higher than the rural site. Thus, ambient  $\text{PM}_{2.5}$  contained more EC at the suburban site than in Copenhagen urban background. EC averaged  $1.0\ \mu\text{g}/\text{m}^3$  at urban curbside HCAB corresponding to an increment of about a factor of 4 from the rural site RISØ. EC changed 3-6% at RISØ, HCØ and HVID from 2017 to 2019, whereas EC decreased by 13% at HCAB during the same time period.

# 1 Measurements

## 1.1 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 distribution in the submicrometer size regime. Particle number size distributions of diameters 6 - 700 nm were measured at the rural station RISØ, 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). 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 2018, but was exchanged in the beginning of 2019 with a new SMPS system. 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 are 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 the course of 2019, these inlets have been exchanged, implying that data from smaller size regimes can be used from 2020 and onwards.

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 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 and discussed in the former Particle Project, were not solved until primo 2020. Hence, this report is based on size ranges from 41 to 478 nm and particle numbers within this range.

**Table 1.1.** Data coverage for DMPS/SMPS measurements at the rural location (RISØ), suburban location (HVID), urban background location (HCØ) and urban curbside (HCAB).

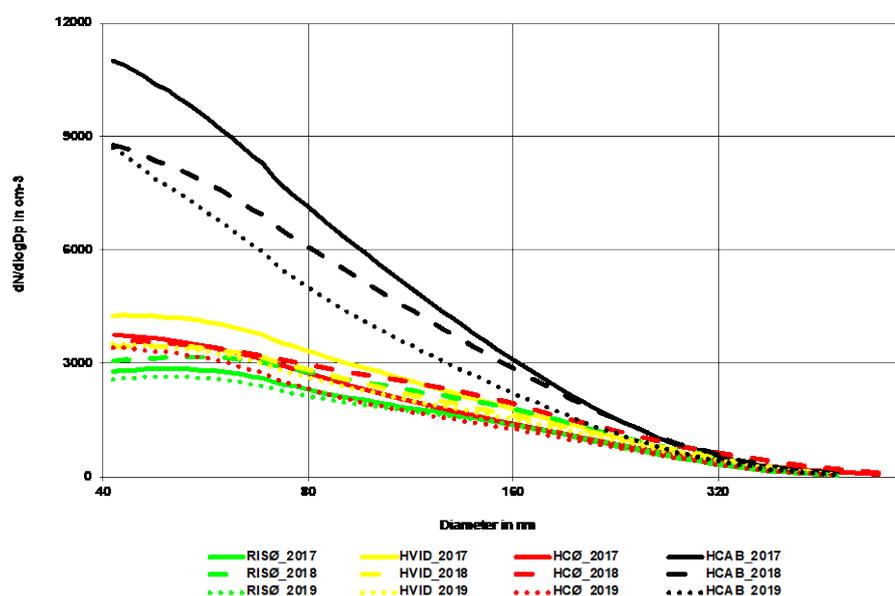
	Rural (RISØ)	Suburban (HVID)	Urban background (HCØ)	Urban curbside (HCAB)
<b>2019</b>	74%	80%	64%	75%

**Table 1.2.** Operation of instruments (old and new) at the four measurement stations during the years 2002 to 2019.

	Rural background (RISØ)	Suburban background (HVID)	Urban background (HCØ)	Urban curbside (HCAB)
<b>Old instrument 6 nm – 700nm</b>	2005-2016	-	2002-2018	2002-2016
<b>New instrument 11 nm – 478 nm</b>	2017-2019	2015-2019	2019	2017-2019

Particle number size distributions are obtained with 30 minutes time resolution. In Figure 1.1, annual mean particle number size distributions at RISØ, HVID, HCØ and HCAB are shown for the years from 2017-2019 in the size range from 41 – 478/550nm. Particle number size distributions were measured at HCØ with the old instruments and are reported in the size range 41 – 550 nm. Similarly, particle number size distributions are measured in the range 11 – 478 nm for the years 2017 – 2019, but reported in the size range 41 – 478 nm at RISØ, HVID and HCAB. In Figure 1.1 the derivative  $dN/d(\log D_p)$  is plotted against the logarithm of the particle diameter  $D_p$ , 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 has decreased from 2017 to 2018 and again from 2018 to 2019.

At the rural location (RISØ), particle numbers have been rather similar over the last three years of observations, although they increased by 21% between 2017 and 2018 and then decreased by a similar fraction between 2018 and 2019. On the contrary, particle numbers at HVID decreased by 13% between 2017 and 2018, and then by 5% from 2018 – 2019 being relatively stable. At HCØ, the particle numbers do show more variations. The numbers increase by 14% between 2017 and 2018, but then decrease by 24% between 2018 and 2019. Because of a relatively low data coverage at HCØ in 2017 and 2018, as well as due to the additional system change at this station in 2019, trends will need to be observed and evaluated over a long time scale. A steady decrease in particle numbers over the three consecutive years is observed for HCAB, which is heavily influenced by emissions from local traffic.



**Figure 1.1.** Annual means of particle number size distributions at the rural location (RISØ), suburban location (HVID), urban background location (HCØ) and urban curbside (HCAB) during 2017 - 2019. Note the logarithmic x-axis.

A slight decrease in particle number between 2018 and 2019 can be observed for smaller particle fractions (41 – 110 nm) at HCAB and HVID, which is in line with what was observed in previous year (Figure 1.2). At other stations, clear changes in levels between recent years cannot be determined. Whether variations and trends at the stations are real or an artefact introduced by the setup of new instruments has to be investigated further when more data is available from the new instruments. It should here be noted, that such differences can

also be caused by meteorological variations from year to year. For this reason, data have to be investigated over many years to draw conclusions and analyse real trends in time series. However, such analyses are carried out for particle fractions in defined size ranges in the following section.

Particle number 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 lead to a general decrease in particle numbers. In general, 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 that the mean diameter is increasing with distance to major sources (Nøjgaard et al., 2015). It is expected, that local sources in urban areas mainly contribute to particle number concentrations, and especially to 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 illustrated in Figure 1.1, when comparing the particle number size distribution at HCAB to the other stations.

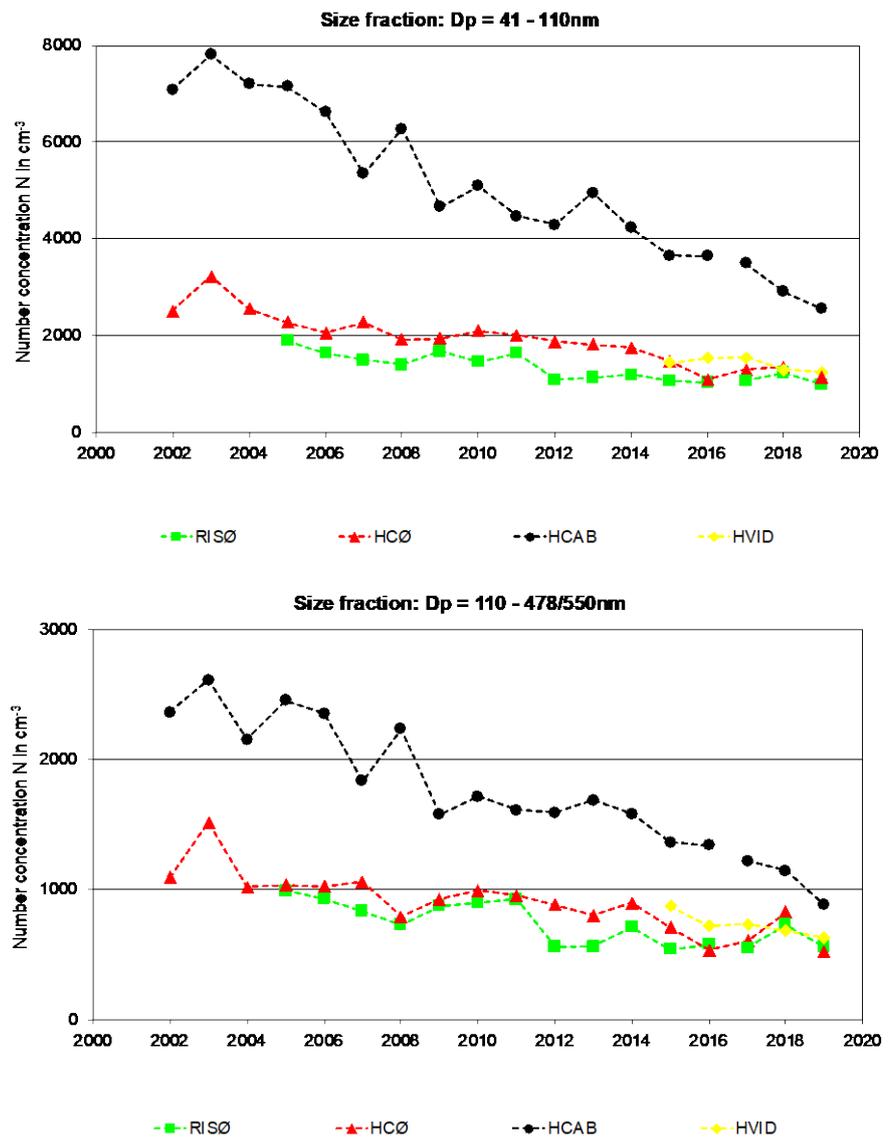
In 2017 and 2019 annual averages at HVID exceeded those at HCØ, which was believed to reflect larger emissions from wood burning in suburban areas during wintertime, and the location of a number of highly busy roads and highways. In 2018, this was not the case, as particle numbers at HCØ slightly exceeded those at HVID. However, the data cannot be used to determine whether particle numbers are highest at HVID or HCØ due to the relatively low data coverage at HCØ in 2017 and 2018.

At RISØ, HVID, and HCØ, the particle number size distributions look rather similar over the years 2017 - 2019 with only smaller variations. As expected, largest number of small particles is found at the urban curbside station HCAB also in all three years. This large number is mainly a result of ultrafine particles originating from vehicle exhaust emissions in busy roads.

Interestingly, decreases are observed at HCAB, and to a smaller extent at HVID, over the years 2017 to 2019, where anthropogenic sources influence particle number concentrations. In contrast, small variations in particle number concentrations throughout the years 2017 to 2019 are observed at the locations RISØ and HCØ for which local sources are a little further away.

### **1.1.1 Particle number fractions**

For a more detailed analysis, particle number concentrations were determined in specific size regimes, in this case particles with diameters of  $D_p = 41 - 110$  nm (medium size regime) and  $D_p = 110 - 550$  nm (large size regime) from the old instruments operated from 2002 to 2016 (HCAB), 2005 to 2016 (RISØ) and 2002 to 2018 (HCØ), and likewise for  $D_p = 41 - 110$  nm (medium size regime) and  $D_p = 110 - 478$  nm (large size regime) from new instruments operated from 2017 to 2019 (HCAB), 2019 (HCØ), 2015 to 2019 (HVID) and 2017 to 2019 (RISØ). Figure 1.2 show annual means for  $D_p = 41 - 110$  nm for the years 2002 to 2019.



**Figure 1.2.** Annual mean particle number concentrations in specific size regimes  $D_p = 41 - 110\text{nm}$  (upper Figure) and  $D_p = 110 - 478/550\text{nm}$  (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 2019.

For HCAB, the largest particle number is typically found in the smallest size regime from 11 to 41 nm (Wåhlin, 2009), which is not presented here. However, a tendency of decreasing particle number is evident at HCAB for the medium and large size regimes 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 introduced over the years. We observe a slight decreasing trend for both size regimes at HVID, a finding that has to be investigated in more detail when longer time series are available at the semi-urban background station.

For other stations like RISØ and HCØ, a clear trend for the different size regimes is also observed with respect to the entire measurement period from 2002/2005 to 2019. The trend is more pronounced for the medium size regime compared to the large size regime. Nevertheless, some variations are observed for some years, and especially in 2018 increases are observed at RISØ

and HCØ for the large size regime which might 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. 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.

## 1.2 Particle mass concentration using TEOM

Measurements of Particulate Matter (PM) are typically specified for PM<sub>2.5</sub> and PM<sub>10</sub>, i.e. particles with aerodynamic diameters smaller than 2.5 micrometre or 10 micrometre, respectively. The reference method is gravimetry. In addition to gravimetric measurements in the National Monitoring Programme under NOVANA, PM is measured using a Tapered-Element Oscillating Microbalance (TEOM) instrument at urban curbside in Copenhagen (PM<sub>10</sub> and PM<sub>2.5</sub> at HCAB), Aarhus curbside (PM<sub>10</sub> at AARGH) and at the rural station RISØ (PM<sub>10</sub>). The TEOM technique enables near real time reporting of data to the public with a time resolution of 30 minutes (Tables 1.3 and 1.4). During sampling, the collected particles are heated to 50°C, which consequently may cause semivolatile particulate species to evaporate. In particular, ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) and some semivolatile organic compounds (SVOC) evaporate in the TEOM, but not in the reference method.

**Table 1.3.** 2019 annual statistics for PM<sub>10</sub> measured by TEOM. The values are based on 30 minutes averages.

Unit: µg/m <sup>3</sup>	Data coverage	Average
<b>Street</b>		
HCAB	95%	26.7
AARGH	84%	16.8
<b>Rural</b>		
RISØ	98%	13.4

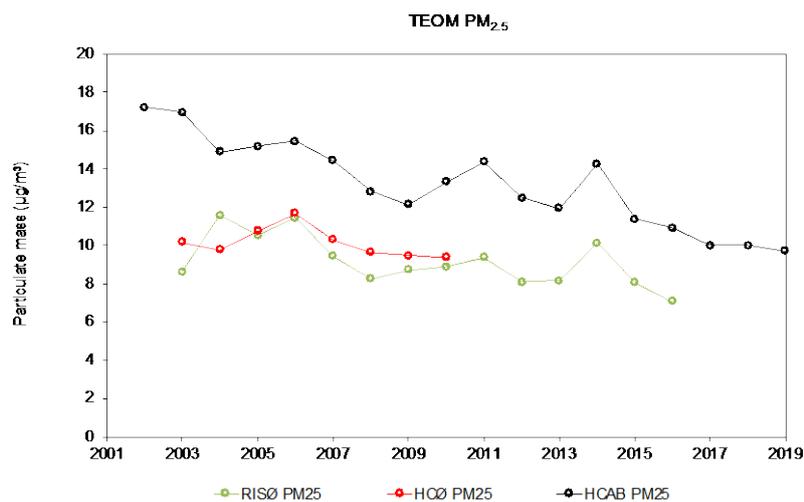
**Table 1.4.** 2019 annual statistics for PM<sub>2.5</sub> measured by TEOM. The values are based on 30 minutes averages.

Unit: µg/m <sup>3</sup>	Data coverage	Average
<b>Street</b>		
HCAB	93%	9.7

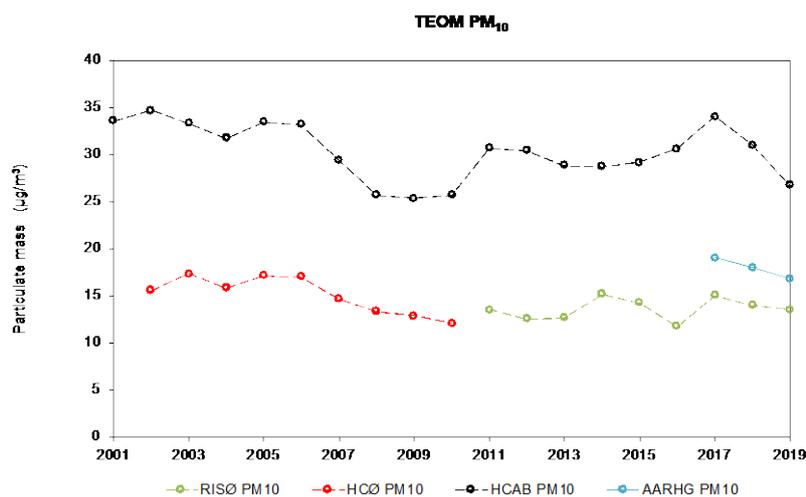
Evaporative losses depend on the chemical composition of the aerosols. The TEOM concentrations can therefore not be directly compared to the reference method, but TEOM data can be utilized to study processes on a short time scale.

The time series of TEOM PM<sub>2.5</sub> at RISØ was terminated in 2017, leaving HCAB as the sole monitoring station left with TEOM PM<sub>2.5</sub>. Simultaneously, measurements of PM<sub>10</sub> by TEOM were initiated at a curbside station in Aarhus (AARGH) in 2017. Thus, PM<sub>10</sub> is measured by TEOM at the curbside stations (AARGH and HCAB), and at the rural location (RISØ) from 2017.

From 2002 to 2019, PM<sub>2.5</sub> and PM<sub>10</sub> decreased 7.5 and 8.0 µg/m<sup>3</sup> at HCAB, respectively (Figures 1.3 and 1.4). This is in accordance with expectations, since HCAB is heavily affected by road traffic of which tail pipe emissions has been intensively regulated within EU. Part of the observed decrease is due to regulations affecting local emission, but long-range transport of fine particles has also decreased. PM<sub>2.5</sub> is part of PM<sub>10</sub>, for which reason a similar reduction should be reflected in both measures provided that no further reductions have occurred in the particle mass with diameters between 2.5 µm and 10 µm. Although a similar reduction is found in both measures, PM<sub>10</sub> clearly shows more variation over the time period 2002 to 2019, than what is seen for PM<sub>2.5</sub>. This will be evaluated in future Particle Projects.



**Figure 1.3.** Trend in annual means of TEOM PM<sub>2.5</sub> at rural location (RISØ), urban background (HCØ) and urban curbside (HCAB) during 2002 – 2019.

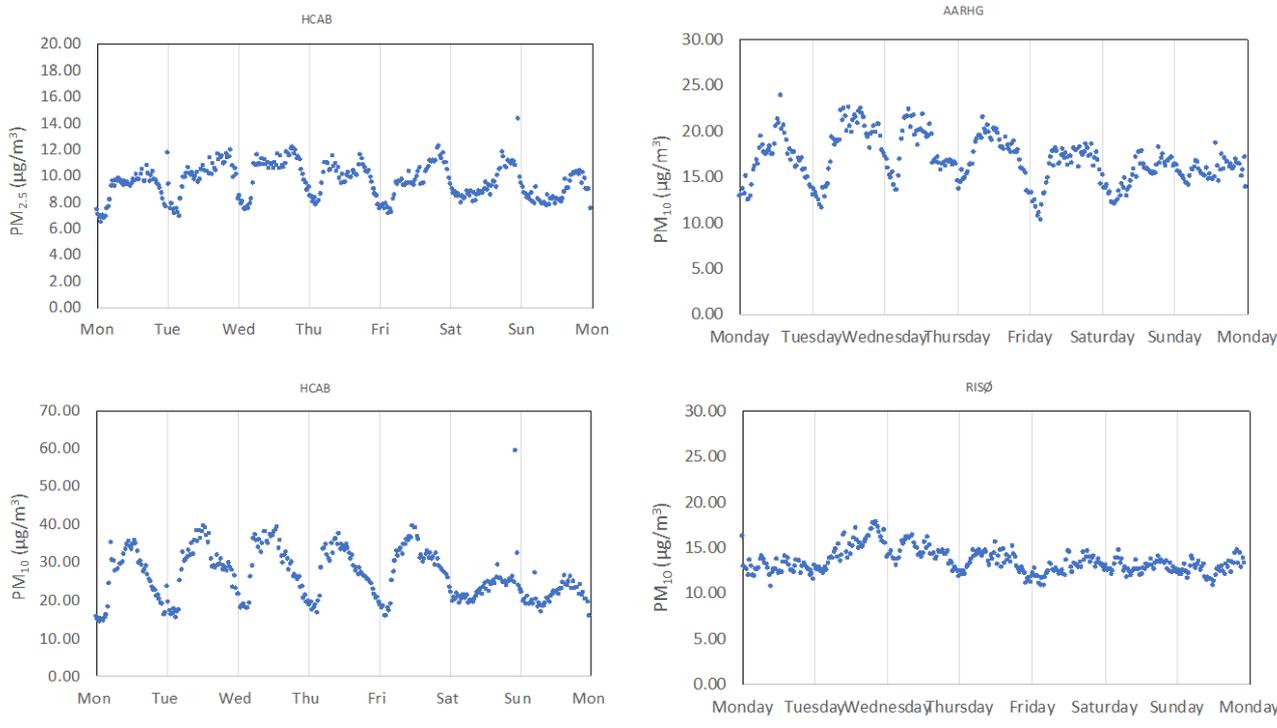


**Figure 1.4.** Trends in annual means of TEOM PM<sub>10</sub> at rural location (RISØ), urban background (HCOE) and urban curbside (HCAB) during 2001 – 2019.

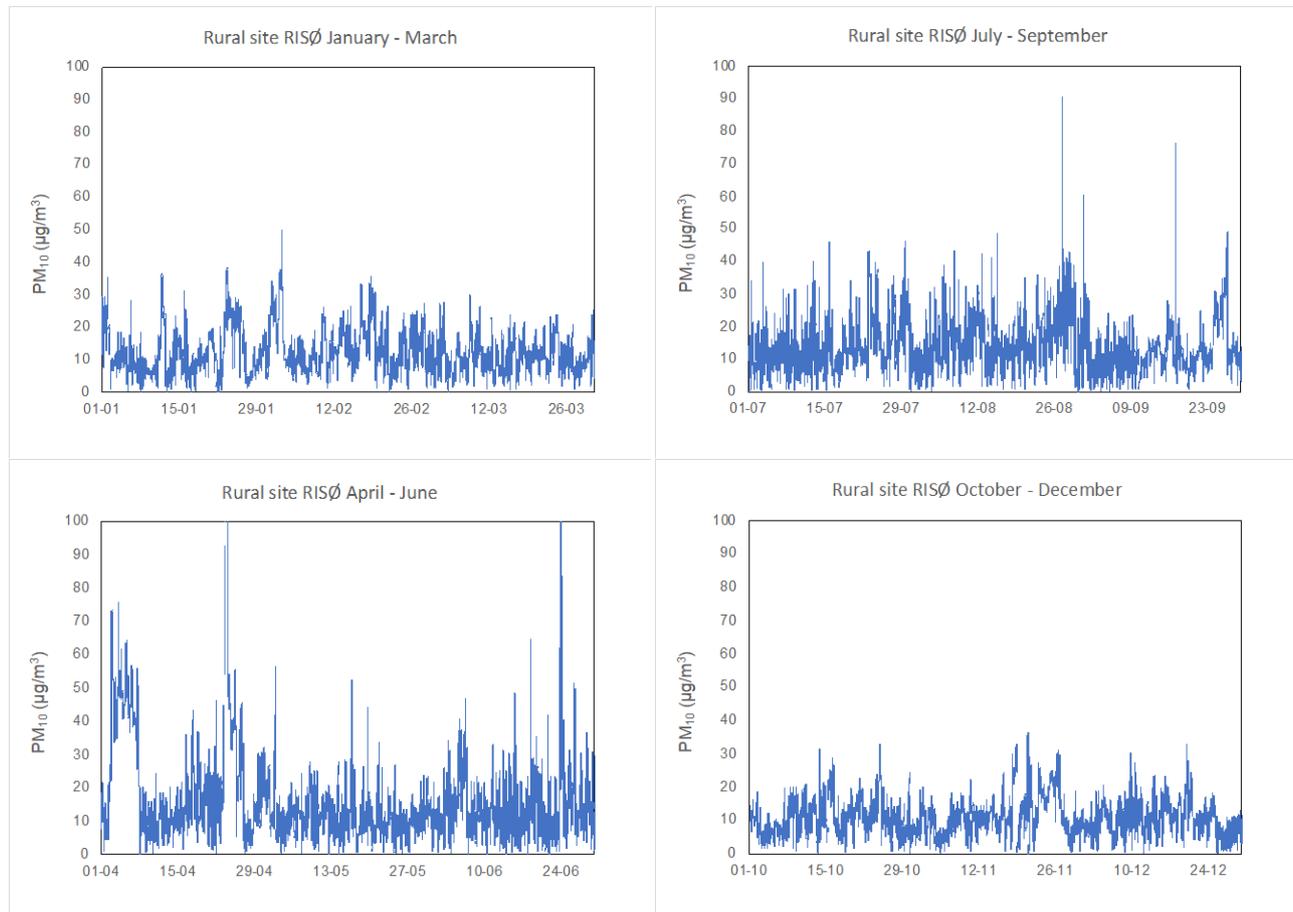
At RISØ, PM<sub>10</sub> decreased by only 4% compared to the previous year 2018. This is roughly the same reduction as observed for PM<sub>2.5</sub> at HCAB, whereas reductions of 7% and 14% were observed for PM<sub>10</sub> at the curbside stations AARHG and HCAB, respectively.

High temporal resolution data allows for studies of diurnal profiles in various environments, e.g. urban curbside. In Figure 1.5, 2019 data for PM<sub>2.5</sub> at HCAB and PM<sub>10</sub> data for HCAB, AARHG and RISØ are sorted by weekday and hour. Each data point represents an average of 52 weeks, and week to week variation in e.g. seasonal and day to day variations in traffic load and meteorology are averaged out. HCAB is among the busiest road in Copenhagen and PM<sub>2.5</sub> is heavily influenced by tailpipe emissions. A morning rush hour peak of PM<sub>2.5</sub> is evident on weekdays, where concentrations typically decrease somewhat in the afternoon and rises to a maximum in the late afternoon/evening, possibly with a contribution from wood stoves. Lowest concentrations are found during the night on weekdays. On weekends, night time concentrations of PM<sub>2.5</sub> are higher than on week days evidencing more night time traffic. Furthermore, there are no rush hour peaks on Saturday and Sunday mornings. A low level prevails during the night until noon on Saturdays and until the afternoon on Sundays. At slightly different pattern is found for PM<sub>10</sub> curbside stations. PM<sub>10</sub> at the street station HCAB and AARHG build-up during the morning to reach a daily maximum. A bimodal curve is still observed with a peak in the afternoon/evening, but the bimodal curve is not as evident as observed for PM<sub>2.5</sub>, which may be a consequence of only one year of data. Longer timeseries will be a subject for further studies in the *Particle Project 2020*. As for PM<sub>2.5</sub>, minimum concentrations are observed shortly after midnight, and lower peak concentrations are observed during weekends compared to weekdays. Peaks and minima reflect a diurnal pattern which for weekdays are different from weekends.

In Figure 1.6, high time resolution TEOM PM<sub>10</sub> data for RISØ is shown for the entire calendar year of 2019. Contrary to HCAB, there is no evident impact from local sources at the rural site. Rather, the substantial day to day variation as well as the seasonal variation reflects mainly variations in meteorology, national activities and long-range transport. By inspection of monthly averages of 2019, higher concentrations are monitored at all stations HCAB, AARHG and RISØ in April 2019 (data not shown). Figure 1.6 reveals that these higher concentrations in April are due to events in the periods 4 to 7 April and 22 to 27 April. In fact, it is possible to examine the onset of long-range transport events, for which average daily concentrations on April 4 to 5 exceeded 50 µg/m<sup>3</sup>, which is much higher than the annual mean of 13.4 µg/m<sup>3</sup>. This is most likely a result of farmers applying manure on their fields.



**Figure 1.5.** Weekly high-resolution 30-min (2019) data of TEOM PM<sub>2.5</sub> at curbside station (HCAB) and TEOM PM<sub>10</sub> at curbside stations AARHG and HCAB, and rural station RISØ.



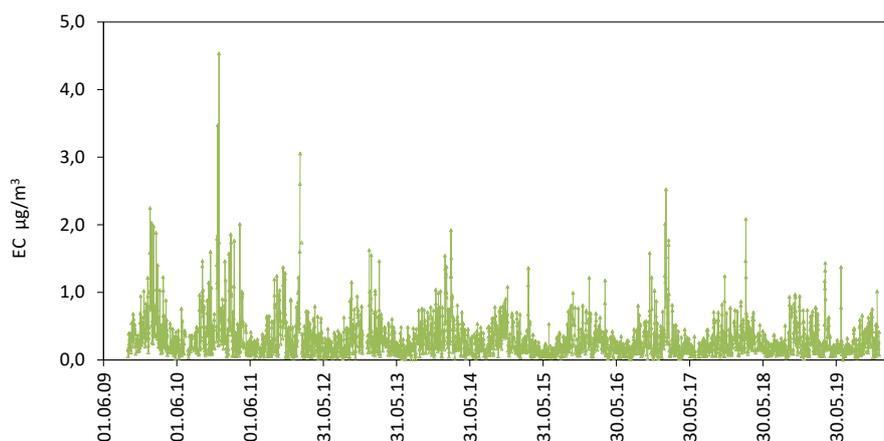
**Figure 1.6.** High-resolution 30-min data of TEOM PM<sub>10</sub> at rural location (RISØ) in 2019.

### 1.3 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 chemical 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).

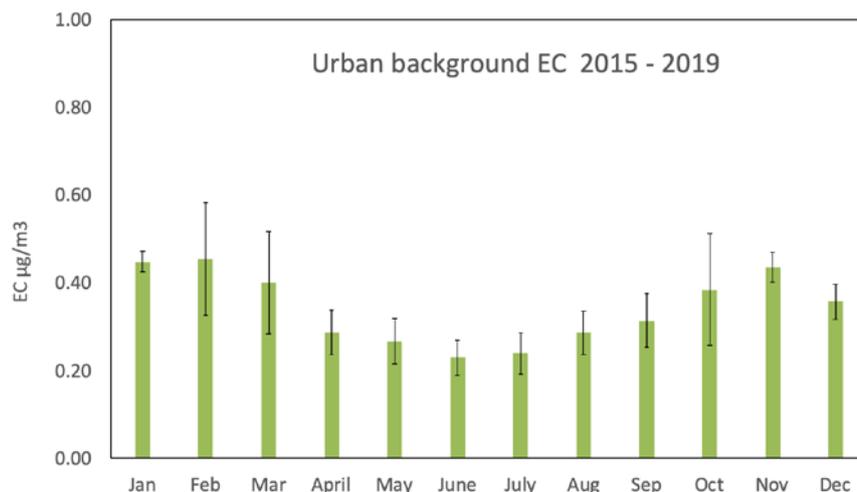
Low Volume Samplers (LVS) equipped with PM<sub>2.5</sub> inlets are located at urban curbside (HCAB) and urban background (HCØ) in addition to rural (RISØ) and suburban (HVID) locations. Atmospheric aerosols are collected on quartz fiber filters using 24-hour time resolution. The filters are subsequently weighted to measure PM<sub>2.5</sub> 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 at Fjlstedvej in Hvidovre from October 2015.

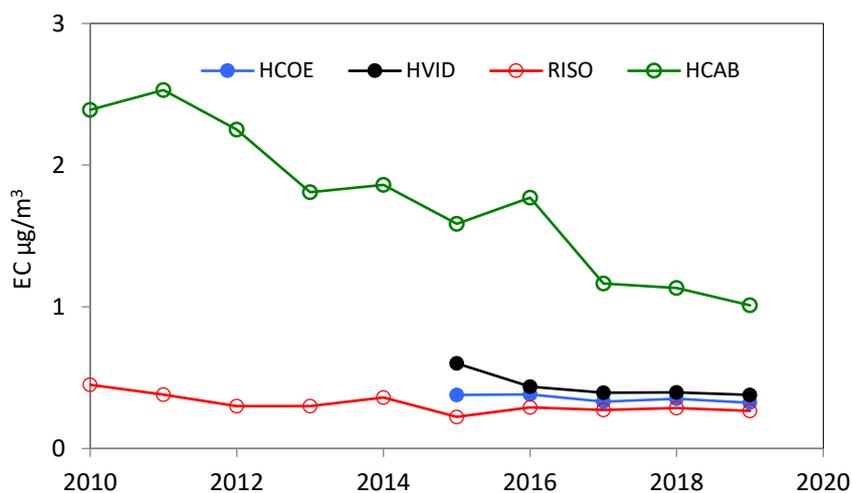


**Figure 1.7.** Trend in annual EC concentrations at the rural location (RISØ) 2009 - 2019

Figure 1.7 shows rural EC measurements from 2009 -2019 at RISØ. Annual variation is mainly caused by biomass combustion, including Residential Wood Combustion (RWC), as concluded in *The Particle Project 2017-2018* (Nøjgaard et al., 2018). Note the strong seasonal variation, with highest observed concentrations during the heating season (Figures 1.7 and 1.8). Based on 10 years of measurements, mean winter concentrations average 0.43 µg/m<sup>3</sup> during December-January-February, while summer concentrations show mean values of 0.18 µg/m<sup>3</sup> during June-July-August. Autumn concentrations are 25% higher than spring concentrations, i.e. 0.35 vs. 0.28 µg/m<sup>3</sup>. Peak concentrations of 2 to 5 µg/m<sup>3</sup> are occasionally observed (Figure 1.7). In 2019, annual mean values were: rural EC 0.26 µg/m<sup>3</sup> < urban background EC 0.32 µg/m<sup>3</sup> < suburban EC: 0.38 µg/m<sup>3</sup> < urban curbside EC 1.01 µg/m<sup>3</sup> (Table 1.5).



**Figure 1.8.** Monthly averaged EC based on the period 01.01.2015 – 31.12.2019. Vertical bars illustrate +/- 1 standard deviation between monthly means.



**Figure 1.9.** Annual trends in annual EC concentrations at urban background (HCØ), sub-urban (HVID) and rural location (RISØ) and urban curbside (HCAB).

Ambient EC has demonstrated a pronounced reduction of 58% ( $1.38 \mu\text{g}/\text{m}^3$ ) at HCAB over the time period 2010 to 2019, 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 emissions standards for newer vehicles. Part of this reduction is ascribed to a general reduction in urban background EC in Copenhagen and Europe in general. Urban background measurements of EC were not available until 2015 when initiated in *The Particle Project 2014-2016* (Nøjgaard et al., 2017). EC has decreased by 41% ( $0.19 \mu\text{g}/\text{m}^3$ ) at RISØ over the time period 2010 to 2019. Therefore, the reduction in EC at HCAB is mainly ascribed to reduction in the local road traffic assuming that trends in rural EC and urban background EC are similar.

**Table 1.5.** 2019 annual statistics for EC at Danish measurement sites. Increments are relative to rural site Risø.

<b>Unit: <math>\mu\text{g}/\text{m}^3</math></b>	<b>Data coverage</b>	<b>EC</b>	<b>EC increment</b>
<b>Urban Street</b>			
HCAB	94%	1.01	282%
<b>Suburban site</b>			
HVID	98%	0.38	43%
<b>Urban background</b>			
HCØ	99%	0.32	22%
<b>Rural site</b>			
RISØ	93%	0.26	

EC concentrations at the rural site Risø have decreased markedly since the onset of measurements in the beginning of the last decade. However, EC has changed 3-6% at RISØ, HCØ and HVID since 2017 (Figure 1.9), whereas a reduction in EC of 13% has been measured at HCAB.

Overall, rural, suburban and urban background appears to follow the same stagnating trend, where EC at urban street continues to decrease. Note that the increment from rural EC levels are higher at the suburban location compared to the urban background, probably due to residential wood combustion. That is, EC is on average 43% higher than rural concentrations in the suburb, and 22% higher than rural concentrations in urban background during 2019.

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## THE PARTICLE PROJECT 2019

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<sub>2.5</sub> and PM<sub>10</sub> 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.