THE PARTICLE PROJECT 2018

Scientific Report from DCE – Danish Centre for Environment and Energy No. 338 2019

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Jacob Klenø Nøjgaard
Andreas Massling
Thomas Ellermann

Aarhus University, Department of Environmental Science

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Data sheet

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Abstract: The Particle Project 2018 continues the record of particle mass and number measurements on urban and rural locations. Monitoring of Elemental Carbon (EC) has been extended from rural location and curbside station in Copenhagen (2009/2010) to include an urban background location (2014) and a suburban location (2015). 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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Abbreviations and definitions

AARHG	Urban curbside measurement site in Aarhus.
BC	Black carbon, which is roughly equivalent to elemental carbon.
°C	Degrees Celsius.
DMPS	Differential Mobility Particle Sizer.
Dp	Particle diameter.
EC	Elemental carbon, roughly equivalent to black carbon (BC).
HCAB	Urban curbside measurement site in Copenhagen.
HCØ	Urban background measurement site at H.C. Ørsted Institute in Copenhagen.
HVID	Suburban measurement site at Fjelstedvej in Hvidovre.
LVS	Low Volume Sampler for atmospheric particles.
OC	Organic carbon, only the mass of carbon itself.
PM	Particle mass in ambient air.
$PM_{2.5}$	Mass concentration of particles smaller than 2.5 micrometer in diameter, i.e. fine particles.
PM_{10}	Mass concentration of particles smaller than 10 micrometer 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

Particulate matter by mass (PM), number of particles and Elemental Carbon (EC) are measured at selected monitoring stations, which are not entirely covered by the National Air Quality Program, NOVANA LMP (Ellermann et al., 2017). Particle number concentration, their size distribution and particle mass concentration are physical parameters for particle size classes PM_{10}, $PM_{2.5}$ and PM_1, i.e. particles with diameter less than or equal to 10, 2.5 and 1 µm, respectively. Chemical parameters include Organic Carbon (OC) and EC in $PM_{2.5}$.

Sammenfatning

I Partikelprojektet 2018 analyseres partikelantal og partikelstørrelsesfordeling på den landlige målestation RISØ, i bybaggrund i København (HCØ), målestationen HVID i forstaden Hvidovre samt på en trafikeret gade i København (HCAB). Endvidere bestemmes måletidsserier af $PM_{2.5}$ og PM_{10}, dvs. massekoncentrationer af partikler med diameter mindre end hhv. 2.5 og 10 µm. $PM_{2.5}$ og PM_{10} måles med Tapered Element Oscillating Microbalance (TEOM) på gadestationerne HCAB og AARHG i hhv. København og Aarhus. Herudover måles også elementært kulstof, EC, i $PM_{2.5}$ på RISØ, HVID, HCAB og HCØ.

Regionale og langtransporterede partikler antages at bidrage i udtalt grad til partikelantallet i PM_1, dvs. partikler med diameter mindre end 1 µm. Den relative forekomst af disse partikler forventes at være størst i landlig baggrund, dvs. på RISØ. Langtransporterede partikler bidrager imidlertid på alle typer målestationer, også målestationer tættere på lokale kilder. Andelen af langtransporterede partikler er dog mindre på HVID, HCØ og især HCAB, hvor partikelforureningen fra brændeovne i fyringssæsonen, og trafikken især på HCAB bidrager med lokale forbrændingspartikler. Over en 10-årsperiode er der observeret faldende partikelantal på alle målestationer.

Antallet af partikler er steget fra 2017 til 2018 på HVID og HCAB, som er direkte påvirket af lokale menneskeskabte kilder. Omvendt er partikelantallet faldet på RISØ og HCØ, der ikke er direkte påvirket af lokale kilder. For begge effekter gælder det, at ændringer kan ses på antallet af både ultrafine partikler (i dette studie 41 - 110 nm), samt størrelsesklassen fra 110 – 478 / 550 nm. Partikelantallet var højere i forstaden HVID end i bybaggrund på HCØ i 2016 og 2017. Forskellen skyldes sandsynligvis flere brændeovne i området, der især påvirker målingerne i den koldere del af året. Det omvendte er tilfældet i 2018, hvilket formodentlig er et artifakt som følge af den ukomplette tidsserie, måleusikkerhed og variationer i meteorologi. Tilsvarende findes det, at partikelantallet på HCØ overstiger det på RISØ, samt at partikelantallet på HCAB overstiger HCØ og HVID.

Koncentrationen af $PM_{2.5}$ på HCAB blev målt med TEOM, hvor årsgennemsnittet for 2018 var det samme som for 2017 (10 µg / m^3), men i de foregående år er aftaget støt siden målingerne blev påbegyndt i 2002 (17 µg / m^3). PM_{10} på HCAB, RISØ og AARHG er derimod aftaget 5-8% fra 2017 – 2018, men ikke over længere tidsserier. Siden fælles TEOM målinger af PM_{10} blev igangsat på RISØ og HCAB i 2011 (hhv. 14 og 31 µg / m^3) er niveauet i målingerne uændret (i 2018 er niveauerne således igen hhv. 14 og 31 µg / m^3), dog med nogen variation årene imellem.

Årsmidlerne for EC udgjorde 0,29 µg / m^3 på RISØ, 0,35 µg / m^3 på HCØ samt 0,40 µg / m^3 på HVID i 2018. En langt højere koncentration blev dog målt på HCAB (1,13 µg / m^3). Både bybaggrund og forstad var forhøjet i forhold til den landlige station RISØ, hhv. 22% og 39% forhøjet i 2018. Mere end 9 års data ligger til grund for tidsserien på RISØ, hvor årsmidlen fra 2010 – 2018 udgør 0,32 µg / m^3, hvilket varierer fra 0,18 µg / m^3 i sommermånederne juni, juli og august til 0,45 µg / m^3 i vintermånederne december, januar og februar. På HCAB er EC i $PM_{2.5}$ aftaget støt siden 2010, mens kun små ændringer er observeret på RISØ og HCØ

Summary

Time series of atmospheric particles smaller than 2.5 µm ($PM_{2.5}$) and particles smaller than 10 µm (PM_{10}) measured by TEOM at the rural site RISØ and curbside stations in Aarhus (AARHG) and Copenhagen (HCAB) are presented in The particle Project 2017-2018. Also covered are submicron particle size distributions are furthermore measured at RISØ, in urban background (HCØ), HCAB and at the suburban site (HVID). Finally, Elemental Carbon (EC) has been analysed in ambient $PM_{2.5}$ at HCØ since September 2014 and compared to RISØ, HCAB and HVID, which was established in October 2015.

Regional and long-range transported aerosols are to a large extent expected to contribute to the particle number concentration in the submicrometer size range. On a relative base the highest contribution of such aerosols are found at rural background sites, e.g. RISØ in Denmark. Long-range transported aerosols also consequently contribute to other sites which are located with smaller distances to local sources.

For Denmark, the relative contribution of long-range transported aerosols therefore decreases when considering the suburban site at HVID, urban background site HCØ and most pronounced at the urban curbside site HCAB, where highest number concentrations are measured due to a strong local traffic source. A trend of decreasing concentrations can be found at all stations when considering time periods of the order of a decade. From 2017 to 2018, particle number concentrations increased at HVID and HCAB, where anthropogenic sources influence the stations. On the other hand, particle numbers decreased in the same period at RISØ and HCØ, which are not directly influenced by local sources. These changes were observed for both ultrafine particles (41 - 110 nm in this study) and particles in the range 110 – 478/550 nm. In 2016 and 2017, higher particle number concentrations have been measured at HVID which is influenced by elevated wood stove emissions during winter-time. In 2018, particle number concentrations at HCØ exceeded those at HVID, which is probably a consequence of the low data coverage at HCØ, variations due to meteorology and within the range of measurement uncertainty.

$PM_{2.5}$ measurements at HCAB in 2018 using TEOM did not change from its annual concentration in 2017 (10 µg / m^3), but has decreased steadily since the onset of the time series in 2002 (17 µg / m^3). On the other hand, PM_{10} at RISØ, AARHG and HCAB decreased 5-8% from 2017 – 2018. Over longer time series, PM_{10} did not change significantly. At RISØ and HCAB, where common time series have existed since 2011, the concentrations in 2018 are similar to those measured in 2011 (14 and 31 µg / m^3, respectively).

EC averaged 0.29 µg / m^3 at RISØ and 0.35 µg / m^3 at HCØ in 2018. Higher concentrations were measured at the suburban station HVID (0.40 µg / m^3) and even higher at HCAB (1.13 µg / m^3. Both suburb and urban background were elevated with respect to EC compared to the rural site RISØ, i.e. 22% at HCØ and 39% and HVID in 2018. For the longest time series at RISØ, the annually averaged EC is 0.32 µg / m^3 based on 9 years of measurements varying from 0.18 µg / m^3 during the 3 summer months and 0.45 averaged over the 3 winter months. EC concentrations have showed an overall decreasing trend at HCAB since 2010, whereas little change has been observed at RISØ, HCØ and HVID.

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 in and around Copenhagen. Locations included the rural station RISØ (previously Lille Valby), urban background HCØ and urban curbside HCAB. From 2017 and onwards, the instruments at HCAB and RISØ were replaced with commercial instruments delivered by TSI (Model 3938). 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. The DMPS system has been 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) is operated at the suburban station HVID in Hvidovre. The new instruments are connected to new inlet systems which introduce losses that affect the general uncertainty of particle number concentrations.

As discussed in *The particle Project 2017-2018 (Nøjgaard et al., 2018)*, the slightly different measurements 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 particle in the range 11-41 nm, which were observed and discussed in the former Particle Project, is still under investigation. Consequently, only data from 41 nm will be included in this specific report.

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

	Rural (RISØ)	**Suburban (**HVID)	**Urban background (**HCØ)	**Urban curbside** (HCAB)
2018	71%	62%	33%	88%

Particle number size distributions are obtained with 30 minutes time resolution. One of the original DMPS instruments is monitoring at HCØ, and unfortunately its data coverage has been affected by frequent service and repair of the instrument. Data acquisition at HCAB and RISØ with new instruments started in February 2017.

1.1.1 Particle number size distribution

In Figure 1.1, annual mean particle number size distributions at RISØ, HVID, HCØ and HCAB are shown for 2018. For comparison, data from 2011 – 2013, 2014 – 2016, and 2017 representing previous *Particle Project* periods are also shown. Particle number size distributions are measured at HCØ in the size range 6 - 700 nm, but reported in the size range 41 - 550 nm for reasons explained in section 1.1. Similarly, particle number size distributions are measured in the range 11 - 478 nm, but reported in the size range 41 - 478 nm at RISØ, HVID and HCAB.

At the rural site RISØ, particles numbers have been rather similar in the first three periods, but increased by 21% from 2017 – 2018. On the contrary, particle numbers at HVID decreased by 13% from 2017 - 2018 following a period with stagnating concentrations. At HCØ, the particle numbers in 2017 and 2018 were lower than the two preceeding periods and especially for the first period from 2011 to 2013. Although the average particle number concentration at HCØ in 2018 is somewhat higher than in 2017, it is associated with high uncertainty due to the low data coverage. A steady decrease in particle numbers through all 4 periods is observed for HCAB, which is heavily influenced by traffic.

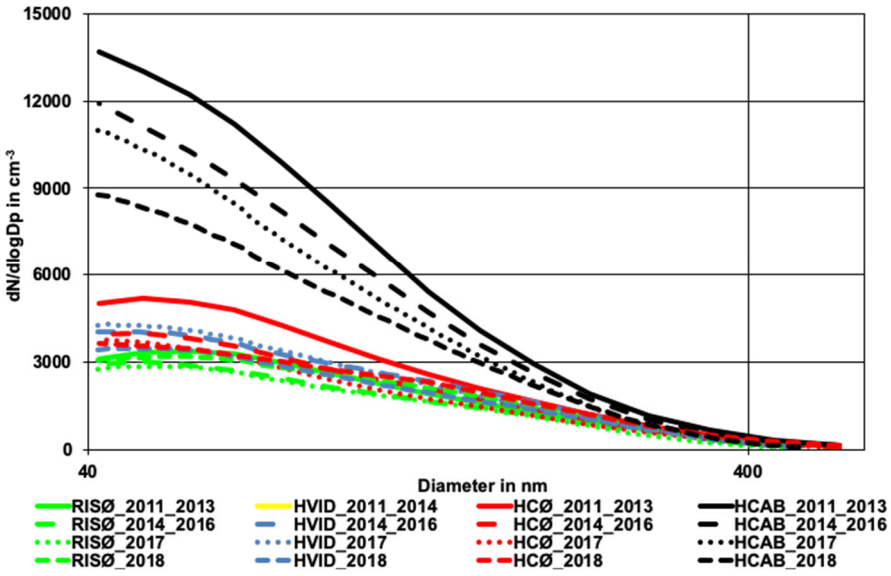

Figure 1.1. Annual means of particle number size distributions at the rural site (RISØ), suburban site (HVID), urban background (HCØ) and urban curbside (HCAB) during 2011 - 2013, 2014 - 2016, 2017 (previous particle projects) and 2018.

A slight decrease in particle number can be observed for smaller particle fractions (41 – 110 nm) at HCAB in 2018 as it was also observed for previous periods. At other stations clear trends cannot be determined. Whether these variations at other stations are real or an artefact introduced with the setup of new instruments has to be investigated further in future data analysis, and when more data is available. Also, such differences can be caused by meteorological variations that occur from one year to the next year. For this reason data have to be investigated over a series of many years to draw conclusions and analyse real trends.

Particle number generally decrease with distance to major sources of aerosol particles. This is the case as polluted air becomes diluted with cleaner air during aging processes resulting in a general decrease of particle numbers in the respective air mass. Also, the mean diameters are increasing for aerosols observed with larger distance to major sources (Nøjgaard et al., 2015). It is expected that local sources in urban areas mostly contribute to ultrafine particles being smaller than 100 nm in diameter and in this way particle size distributions close to major sources are characterized by smaller mean diameters in general.

Over the timeperiod 2015 - 2018 where common time series are available for both HCØ and HVID, only small differences in particle number concentrations have been observed at the two stations. Annual averages at HVID exceeded those at HCØ in 2016 and 2017, which was belied to reflect larger emissions from wood stoves in suburban areas during wintertime, and the location of a number of highly busy roads and highways. In 2018, particle numbers at HCØ slightly exceeded that at HVID. However, the data cannot been used to determine whether particle numbers are highest at HVID or HCØ due to the low data coverage.

At HVID the particle number size distribution looks quite similar in the periods 2014 - 2016 and 2017. In 2018, particle numbers decrease by 13% which is roughly equal to the decrease measured at HCAB (14%). The largest number of small particles at all stations is expected at the urban curbside station HCAB also in 2018. This large number is mostly a result of ultrafine particles originating from vehicle exhaust emissions close to trafficked roads.

Interestingly, decreases are observed from 2017 – 2018, where anthropogenic sources influence particle number concentrations at HVID and HCAB, whereas inceases in particle number concentrations are observed at locations with no apparent local sources at RISØ and HCØ.

1.1.2 Particle number fractions

For a more detailed analysis, particle number concentrations were determined in specific size regimes, in this case particles with diameters of DpF1 = 41 – 110 nm and DpF2 = 110 – 550 nm for old instruments and DpF1 = 41 – 110 nm and DpF2 = 110 – 478 nm for new instruments at all four stations. In Figure 1.2 averages of the annual means for the years 2011 - 2013, 2014 – 2016, 2017 and 2018 are calculated. At HVID, no data is available prior to late 2015.

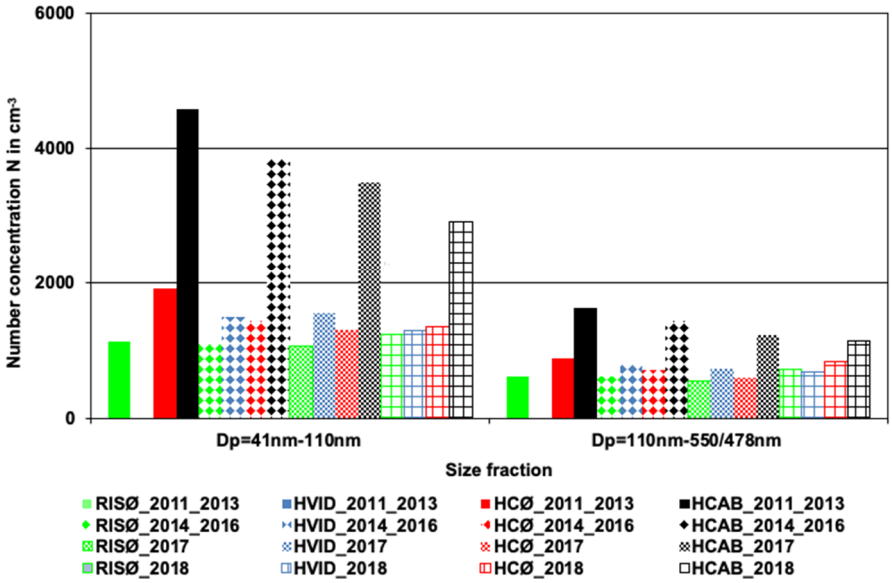

Figure 1.2. Annual mean particle number concentrations in specific size regimes DpF1 = 41 - 110 nm and DpF2 = 110 - 550 nm (old instruments) and DpF1 = 41 - 110 nm and DpF2 = 110 - 478 nm (new instruments) at the rual site (RISØ), suburban site (HVID), urban background (HCØ) and urban curbside (HCAB) during 2011 - 2013, 2014 - 2016, 2017 (previous particle projects) and 2018.

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 all size regimes and all years. A general decrease in particle number is in accordance with the European trend of avoiding particulate emissions especially from road traffic where new environmental regulations and cleaner technologies are continuously introduced.

For other especially more rural stations, it is difficult to identify clear trends. Nevertheless, some trends are expected as the background aerosol is affected by general emissions in Europe and worldwide based on anthropogenic activities. On the other hand also natural processes result in relatively stable values over time. Small variations within the last years might be based on meteorological factors that can highly impact on the particle numbers observed for a single year.

1.2 Particle mass concentration using TEOM

Measurements of Particulate Matter (PM) are typically specified for a given particle diameter, e.g. an aerodynamic diameter smaller than 2.5 micrometre ($PM_{2.5}$) or 10 micrometre (PM_{10}). The reference method is gravimetry. In addition to gravimetric measurements in NOVANA LMP, PM is measured using a Tapered-Element Oscillating Microbalance (TEOM) instrument at urban curbside (PM_{10} and $PM_{2.5}$ at HCAB), Aarhus curbside (PM_{10} at AARGH) and at the rural station RISØ (PM_{10}) in the Particle project 2018. Time resolution is 30 minutes (Table 1.2 and 1.3), which enables near real time reporting of data to the public. During sampling the collected particles are heated to 50°C. Consequently, semivolatile particulate species may evaporate.

Table 1.2. 2018 annual statistics for PM_{10} measured by TEOM. The values are based on 30 minutes averages.

Unit: µg/m^3	Data coverage	Average	Average x 1.3
Street			
HCAB	99%	31	40
AARHG	83%	18	23
Rural			
RISØ	97%	14	18
Limit value			40

Table 1.3. 2018 annual statistics for $PM_{2.5}$ measured by TEOM. The values are based on 30 minutes averages

Unit: µg/m^3	Data coverage	Average	Average x 1.3
Street			
HCAB	98%	10	13
2015 Limit value (2020 in brackets)			25 (20)

Examples are Secondary Organic Aerosols (SOA) and in particular ammonium nitrate, NH_4NO_3. Evaporative losses depend on the chemical composition of the aerosols. The European Commission has permitted that measurements of PM using TEOM may be applied with a default correction factor of 1.3. However, the correction factor depends e.g. on the specific measurement

site and seasonality. Correction of PM using a correction factor of 1.3 does therefore have considerable uncertainty.

The time series of TEOM $PM_{2.5}$ at RISO was discontinued in 2017, leaving HCAB the sole monitoring station left with $PM_{2.5}$ measurements by TEOM. At the same time, measurements of PM_{10} by TEOM were initiated at a curbside station in Aarhus (AARHG) in 2017. Thus, PM_{10} is measured by TEOM at the curbside stations AARHG and HCAB, and at the rural site RISØ from 2017.

The annual average of $PM_{2.5}$ in 2018 was identical to 2017 at HCAB , whereas PM_{10} decreased from 34 µg / m³ (2017) to 31 µg / m³ in 2018, which matched the annual average in 2016 (Figures 1.3 and 1.4). At HCAB, RISØ and AARHG, TEOM PM_{10} decreased 5-8% compared to 2017. Annual averages of TEOM PM_{10} at RISØ and HCAB in 2018 are identical to 2011, where measurements were initiated at RISØ. In this period, TEOM $PM_{2.5}$ has decreased from 14 to 10 µg / m³.

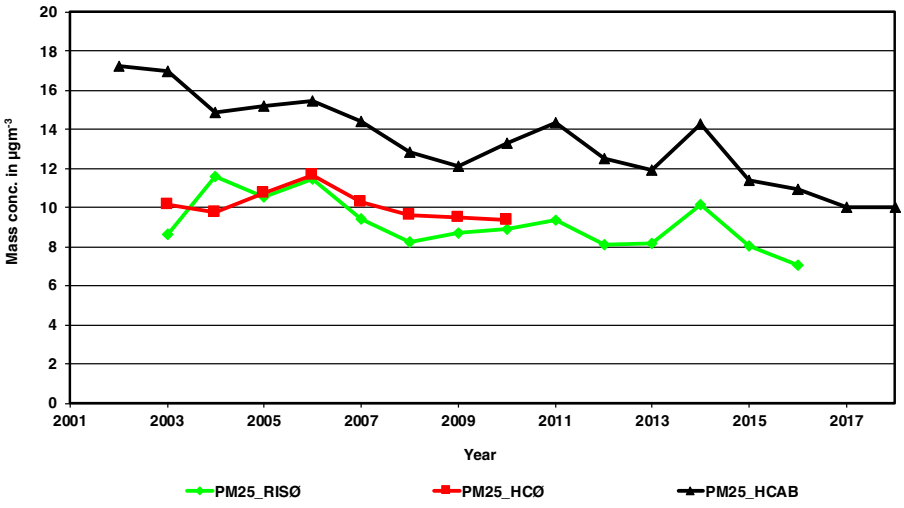

Figure 1.3. Trend in annual means of TEOM $PM_{2.5}$ at rural site (RISØ), urban background (HCØ) and urban curbside (HCAB) during 2002 – 2018.

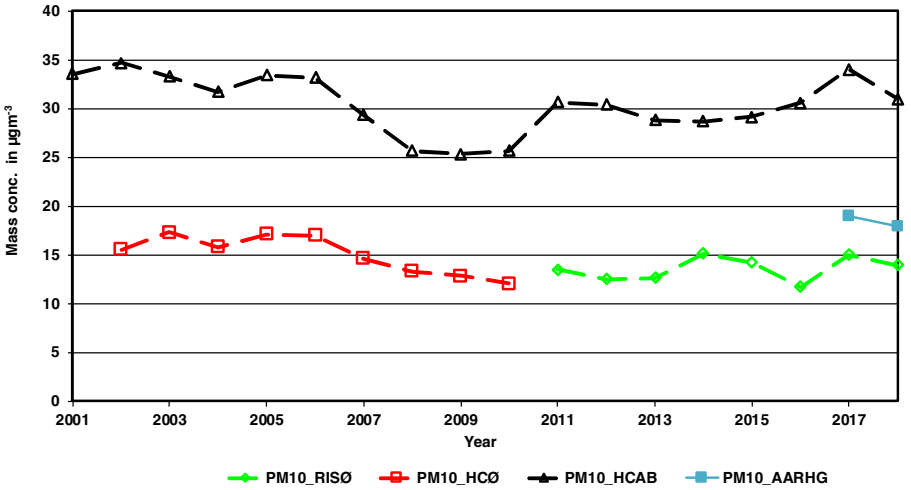

Figure 1.4. Trend in annual means of TEOM PM_{10} at rural site (RISØ), urban background (HCOE) and urban curbside (HCAB) during 2002 – 2018.

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 chemically inertness as elemental carbon (EC). EC has been measured using the European standard thermal optical protocol EUSAAR2 (Cavalli et al., 2010).

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

EC and particular organic carbon (OC) have 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 Fjelstedvej in Hvidovre by October 2015.

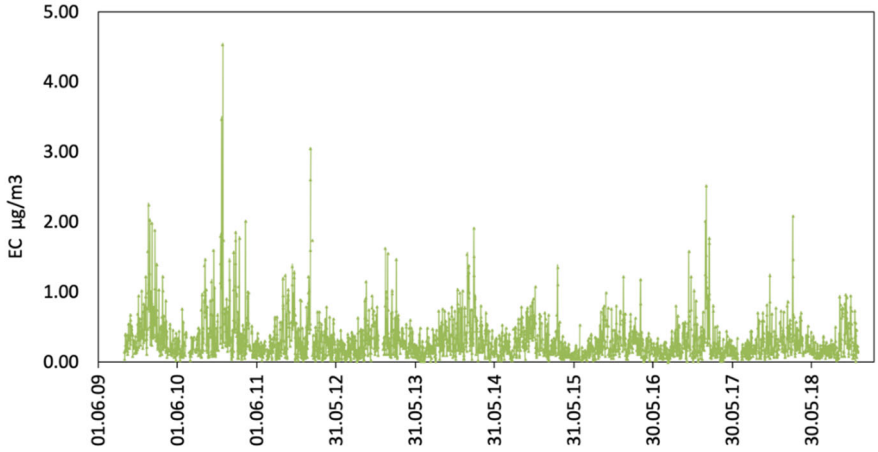

Figure 1.5. Trend in annual EC concentrations at the rural site RISØ 2009 - 2018.

Figure 1.5 shows EC measurements from 2009 – 2018 at RISØ. The 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 concentrations during the heating season (Figure 1.5 and 1.6). Based on 9 years of measurements, winter concentrations average 0.45 µg / m^3 during December-January-February, while summer concentrations average 0.18 µg / m^3 during May–June-July. Autumn concentrations are slightly higher than Spring concentrations, i.e. 0.36 vs. 0.28 µg / m^3 (Figure 1.6). Peak concentrations of 2 - 5 µg / m^3 are occasionally observed (Figure 1.5), but has only a minor influence on the annual mean of 0.32 µg / m^3. Ambient EC concentrations are lowest in rural areas and highest at urban curbside (Figure 1.7). In 2018, annual averages were: rural EC: 0.29 µg / m^3 < urban background EC: 0.35 µg / m^3 < sub-urban EC: 0.40 µg / m^3 < urban curbside EC: 1.13 µg / m^3.

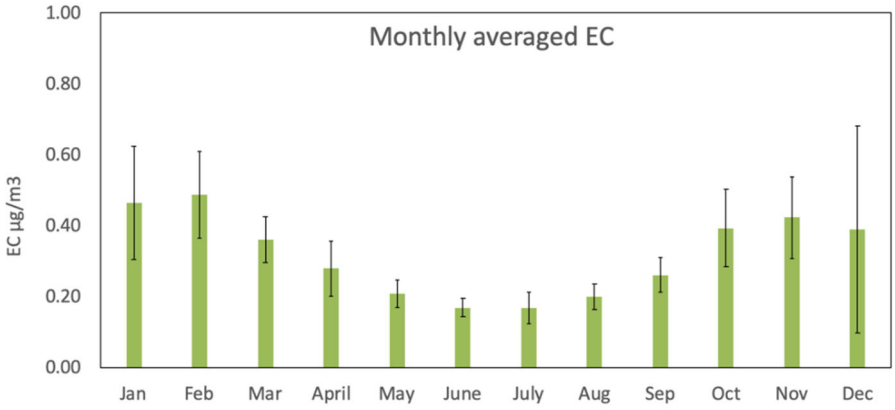

Figure 1.6. Monthly averaged EC based on the period 01.01.2010 – 31.12.2018. Vertical bars illustrate +/- 1 standard deviation between monthly means.

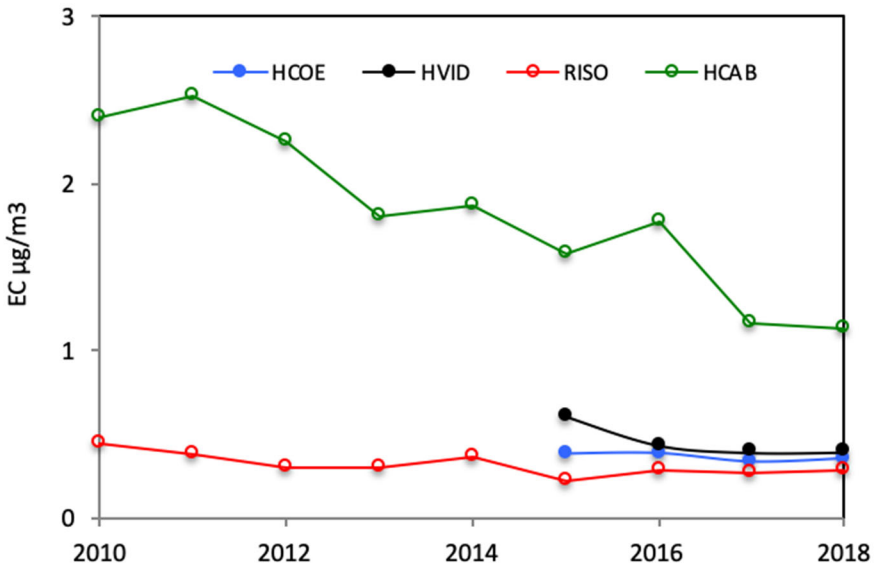

Figure 1.7. Trend in annual EC concentrations at urban background (HCØ), suburban (HVID) and rural locations (RISØ) and urban curbside (HCAB).

Ambient EC has shown a pronounced reduction of 53% (1.26 µg / m^3) at HCAB in the period 2010 – 2018, predominantly due to a reduction in traffic emissions as improved combustion technology in general, and e.g. 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*)*. However, EC has decreased by 39% (0.16 µg / m^3) at RISØ from 2010 – 2018. Therefore, the reduction in EC at HCAB is mainly ascribedto the local traffic source assuming the trends in rural EC and urban background EC being similar.

Only little change is observed in ambient EC at RISØ (0.27 +/- 0.03 µg / m^3) and HCØ (0.36 +/- 0.02 µg / m^3) from 2015 – 2018 (Figure 1.7). Measurements of ambient EC at HVID were initiated in October 2015, and therefore not influenced by low summer levels, for which reason EC is markedly higher in 2015 compared to the average of 2016 - 2018 (0.41 +/- 0.02 µg / m^3).

HCØ appears to follow the same trend as the rural site, though only based on 4 years of data. In 2018, HCØ (0.35 µg / m^3) and HVID (0.40 µg / m^3) experienced EC concentrations 22 and 39% higher than at RISØ.

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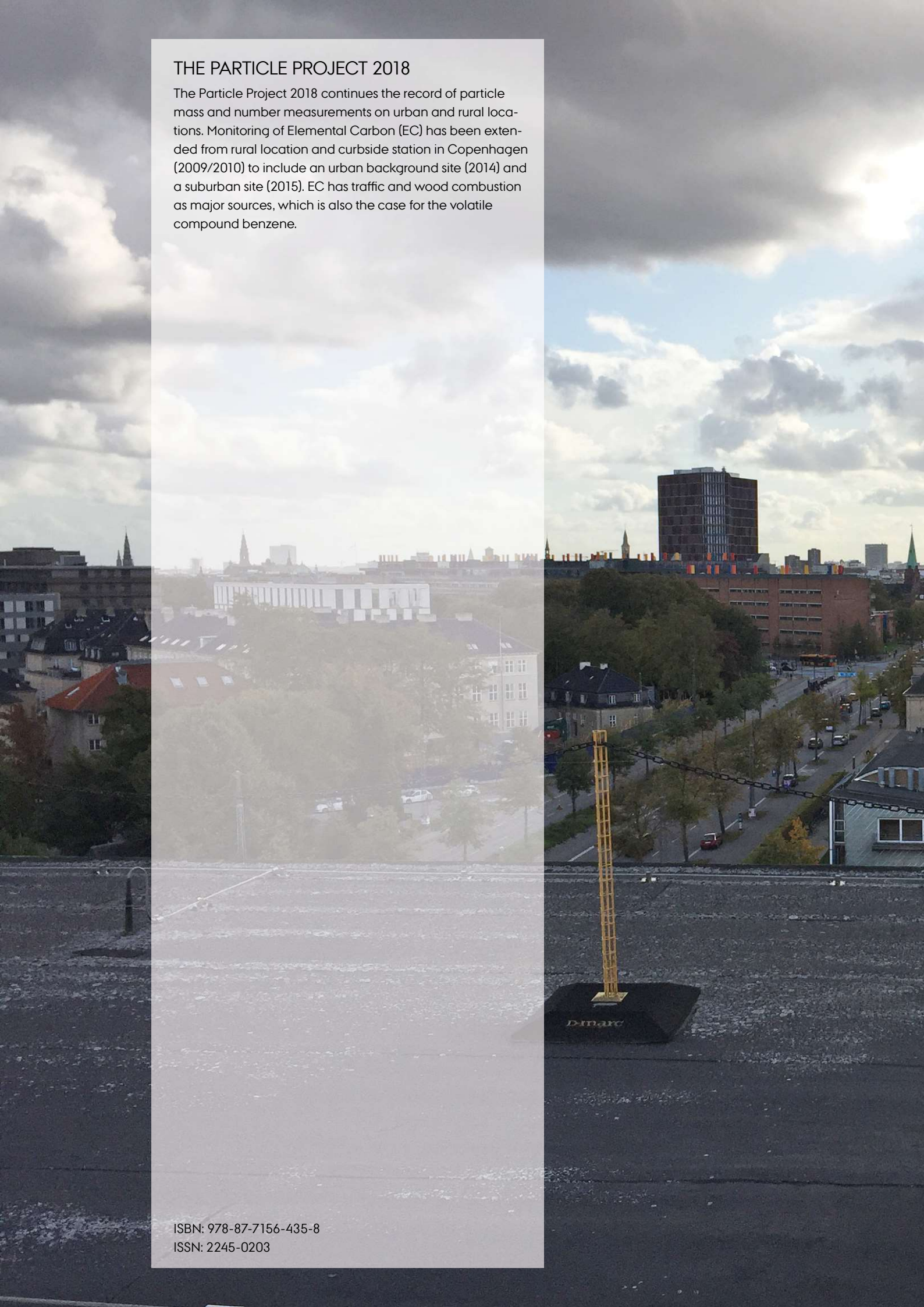

THE PARTICLE PROJECT 2018

The Particle Project 2018 continues the record of particle mass and number measurements on urban and rural locations. Monitoring of Elemental Carbon (EC) has been extended from rural location and curbside station in Copenhagen (2009/2010) to include an urban background site (2014) and a suburban site (2015). EC has traffic and wood combustion as major sources, which is also the case for the volatile compound benzene.

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