

Characterizing aerosol sources based on aerosol optical properties and dispersion modelling in a Scandinavian Coastal Area (Aarhus, Denmark)

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Abstract. Coastal aerosols are formed through the complex mixing between marine air masses and continental emissions, which originate from both natural and anthropogenic sources. The properties of coastal aerosols are decisive for their interaction with sunlight and influence on clouds as well as the potential health implications for the population in these areas. In this study, the aerosol properties and sources at Aarhus Bay, Denmark, were investigated by combining in-situ aerosol light scattering and absorption with size distribution measurements and footprint analysis by FLEXPART. Our analysis demonstrates a considerable contribution of anthropogenic aerosols from both fossil fuel combustion and biomass burning as well as periods with highly scattering aerosols. Furthermore, good agreement was found between in-situ and modelled black carbon data. Combining in-situ measurements and FLEXPART analysis further evidenced a major impact of local emissions as well as a few long-range transport intrusions.

10 1 Introduction

Atmospheric aerosols play a critical role in regional and global climate systems by scattering and absorbing solar radiation and by acting as nuclei for cloud droplets and ice crystals (Masson-Delmotte et al., 2021). Despite their importance, aerosol-radiation and aerosol-cloud interactions remain among the largest sources of uncertainty in current climate predictions (Masson-Delmotte et al., 2021). At the same time aerosols can have negative impacts on human health (Pope III and Dockery, 2006). Reducing uncertainties on aerosol climate effects and improving insights into aerosol adverse health effects require knowledge about ambient aerosol loading and properties across diverse environments. Coastal regions are particularly complex environments where marine and continental air masses interact, leading to highly variable aerosol chemical composition and physical properties (Lewis and Schwartz, 2004). Coastal airmasses are influenced by both anthropogenic and natural sources, including contributions from sea spray, shipping, urban outflow, and biogenic activity. Importantly, coastal zones are also-home to a large fraction of the global population, as approximately 40 % of people live within 100 km of a coastline (Cosby et al.,

2024), ~~highlighting~~. This highlights the importance of understanding aerosol characteristics in these regions for air quality and human health purposes.

The interaction of aerosol particles and solar radiation depends on particle size, shape, and chemical composition as well as the incident wavelengths (Bohren and Huffman, 2008). These are determined by the aerosols' sources and how they have been processed in the atmosphere. In-situ measurements of scattering and absorption coefficients and derived quantities like single scattering albedo and Ångström exponents provide a direct way to probe these characteristics (Cazorla et al., 2013; Cappa et al., 2016; Dubovik et al., 2002). The wavelength dependence of the optical parameters allows to distinguish between particle types such as dust, black carbon (BC), and brown carbon (BrC). Coupling these measurements with auxiliary in-situ data, such as particle size distributions, and air mass back trajectories further constrains source regions thereby enhancing our ability to trace observed aerosol variability back to particular emission sources and transport pathways.

Several long-term atmospheric monitoring stations have provided valuable insights into aerosol behaviour in coastal and marine-influenced environments. Sites such as Mace Head (Ireland), Finokalia (Greece), and the Cape Verde Atmospheric Observatory have contributed ~~decades to~~ decades-worth of in-situ data on aerosol optical, chemical, and microphysical properties (e.g. Dall'Osto et al., 2010; Kalivitis et al., 2019; Fomba et al., 2014). While aerosol optical properties have been widely studied using remote sensing techniques (e.g. Zhang et al., 2025) only a handful of shorter field campaigns have examined the scattering and absorption potential of ambient aerosols particularly in Scandinavia (Ahlberg et al., 2023; Martinsson et al., 2014; Mogo et al., 2012), and only few targeted coastal aerosols (Mogo et al., 2012; Clarke et al., 2003; Donato et al., 2020). Although Denmark has a long coastline compared to the size of the country (Worm, 1997), studies of coastal aerosols in Denmark are scarce (Vignati et al., 1999; Kivekäs et al., 2014), and none have coupled aerosol optical properties with size-distribution measurements and trajectory modelling, limiting our understanding on source attribution and radiative impacts in these settings.

In this study, we measured and analysed the absorption and scattering potential as well as number and surface size distributions of aerosols measured at a coastal site in Denmark during a ~~6.5-week~~ 40-day field campaign in spring 2023. Our site was located near Aarhus, at the Kattegat coast of Jutland in Denmark, offering a unique environment to study the properties of coastal aerosols. Depending on wind direction and synoptic conditions, this site can be influenced by marine air masses from the North Sea or the Baltic Sea, long-range transported aerosols from continental Europe and Asia as well as local influences such as emissions from nearby shipping lanes and urban outflow from the city of Aarhus.

The wavelength dependence of these optical parameters, combined with number and surface size distribution data, was used to assess aerosol types and sources. To further investigate source regions and particle age, we employed the Lagrangian particle dispersion model FLEXPART to simulate air mass histories. Within specific case studies we highlight the variability in aerosol loading and composition. The results contribute to a more detailed understanding of aerosol sources and their characteristics in coastal Denmark.

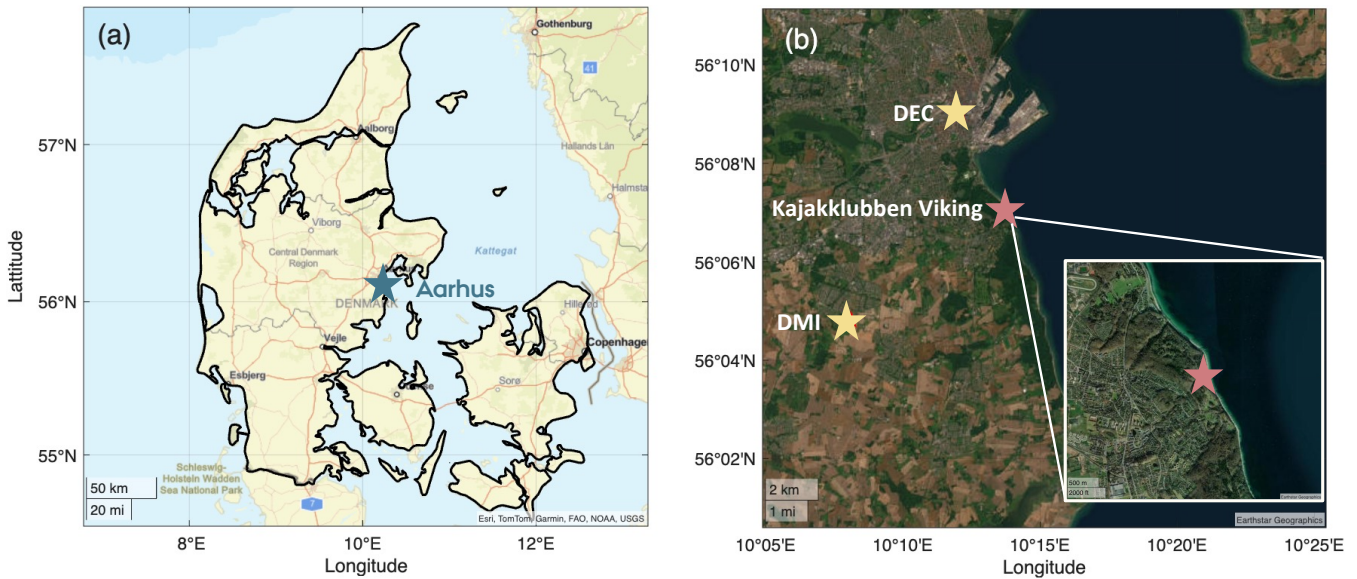


Figure 1. Map of (a): Aarhus (Basemap Sources: Esri, TomTom, Garmin, FAO, NOAA, USGS) and (b): sampling sites (Basemap provided by Earthstar Geographics.). In (b), Kajakklubben Viking is marked with a red star and DMI and DCE stations providing meteorological and air quality data are marked with yellow stars. It was Produced using MATLAB version 9.14 (R2023a). (Matlab, 2021).

2 Methods

2.1 Measurement site

55 The measurements were carries-carried out in Aarhus (56.13° N, 10.22° E) from 3 March to 11 April, 2023. Aarhus is a coastal city, the second largest city in Denmark, and has approximately 360.000 inhabitants and an area of 468 km². As shown in Fig.1, the instruments were placed in the house of a Kayak club ("Kajakklubben Viking"), located on a beach along the southeastern coastline of Aarhus. The coastline is boarded-bordered by a forest area, where there-including a deer park, which is a habitat for a herd of deer and other wildlife. Outside the forest, there are main roads, residential houses and agricultural fields (to the south-west). In addition, Aarhus Port, one of Denmark's largest container ports, is at a distance of 3.83 km from the sampling site (to the north). Thus, the measurement site is potentially influenced by clean marine, forest and anthropogenic emissions from residential activities, traffic both from the continent (i.e. vehicles) and the sea (marine shipping).

65 Wind, radiation and relative humidity (RH) data were obtained from the Danish Meteorological Institute (DMI). The closest DMI measurement site is located in the south of Aarhus (56.1° N, 10.23° E), 7 km from the Kayak club. PM₁₀ (particulate matter with a diameter of less than 10 μm), nitrous oxide (NO_x) and carbon monoxide (CO) were monitored by the Danish Center for Environment and Energy (DCE), from a station about 5 km from the Kayak Club, located at Banegårdsgade (56.15° N, 10.20° E), close to the railway station characterized by heavy traffic (Nordstrøm et al., 2024).

2.2 Instrumentation

A nephelometer (Aurora 3000, Ecotech), an aethalometer (AE33, Aerosol Magee Scientific) and a white-light optical particle spectrometer (Welas® 2300, PROMO 3000, PALAS) were used to obtain the scattering and absorption coefficients as well as the number and surface particle size distributions of the aerosols. The three instruments were placed inside "Kajakklubben Viking" and shared a stainless steel inlet via a three-way splitter to separate the airflow. The inlet was installed through the wall, parallel to the ground, at a height of around 4 m above the beach. A steel net covered the end of the inlet to prevent droplets and bugs from entering the inlet. The sampled air was dried due to the temperature difference between outdoors and inside where the instruments were located. No additional dryer was employed. A schematic is presented in Fig. S1.

The nephelometer measured the integrating intensity of total light scattering and backscattering from aerosols and gas molecules at three wavelengths ($\lambda = 470\text{-}450$ nm, 525 nm, and $630\text{-}635$ nm) with a resolution of 1 minute. The data were corrected for angular truncation errors using the "no-cut" correction according to Müller et al. (2011). The nephelometer operated at a flow rate of 5.0 L min^{-1} . Both total scattering (angles between 10° and 171°) and backscattering (angles between 90° to 171°) were measured. A full calibration, including zero measurement and span gas calibration with carbon dioxide (CO_2) (99.9 % purity), was performed prior to and after the measurements. As the results coincided within the measurement uncertainty, no additional correction was applied. The nephelometer was equipped with a RH sensor in the measurement cell and two temperature sensors, one in the inlet and the other one in the measurement cell.

The aethalometer measured light attenuation—the attenuation of light through a filter tape progressively loaded with atmospheric aerosol particles, providing a "filter light attenuated signal" at 7 wavelengths ($\lambda = 370$ nm, 470 nm, 520 nm, 590 nm, 660 nm, and 880 nm) at 1 minute time resolution and a flow rate of 5 L min^{-1} (Drinovec et al., 2015). The absorption coefficients are related—were calculated using Eq. (1) by its relation to the equivalent black carbon (eBC) mass concentrations and are calculated using Eq. (1), where MAC is the mass absorption cross section (Hansen et al., 1984):

$$\sigma_{abs}(\lambda) = eBC(\lambda) \cdot MAC(\lambda) \quad (1)$$

A MAC value of 7.77 (mMAC values for the different wavelengths were used as suggested in the manual (e.g. $MAC = 7.77 \text{ g}^{-1}$) was used for $\lambda = 880$ nm, as suggested in the manual). This value was previously deployed at 880 nm was previously employed for aerosols measured in southern Sweden (Ahlberg et al., 2023). Black carbon source apportionment, to subdivide the fractions from fossil fuel combustion and biomass burning, was performed according to Sandradewi et al. (2008), where the portion from biomass burning is referred to as BB%. This model is based on the assumption that the absorption signal is due to only two absorbing aerosol ensembles, one with a low wavelength dependence (fossil fuel combustion aerosols) and one with a high wavelength dependence (biomass burning aerosols), and that no other types of aerosols influence the absorption signal. A detailed explanation of how these sources can be subdivided is presented in the SI. There is a multiple scattering effect caused by light scattering between the particles and the filter fibres. It depends on both the type of filter and the single scattering albedo of the aerosol. Therefore, we followed the approach by Drinovec et al. (2015) including the single scattering albedo of aerosol the aerosols to improve the correction factor value of 1.57 for TFE-coated glass filter (M8020) tape for the aethalometer. Using

a maximum value of 0.025 for the cross-sensitivity of absorption to scattering, as stated in (Drinovec et al., 2015), we find that the maximum total correction factors for both scattering from the aerosol and the filter fibres were 1.76, 1.77, and 1.79 for $\lambda = 470$ nm, 520 nm and 660 nm. This results in a maximum relative error on the absorption coefficients due to the scattering of the particles of up to 12.25 %, 12.99 % and 14.47 % for $\lambda = 470$ nm, 520 nm and 660 nm, respectively. This indicates that the scattering by the aerosols in our specific case did not have a significant effect on the measured absorption signal.

The white-light optical particle spectrometer WELAS was used to measure number and surface size distributions of particles with optical diameters from approximately 0.2 μm to 10 μm (in polystyrene latex (PSL) equivalent particle diameters). The presented total particle number concentrations are thus only representative for this size range, only partly capturing the nucleation and Aitken mode. A flow rate of 5 L min^{-1} was used. The WELAS sensors were calibrated with MonoDust 1500 as provided by PALAS prior to the measurements.

Prior to data processing, all raw data were cleaned for instances with RH above 40 % (0.78% of the total dataset), tubing disconnection and tape advancement of the aethalometer.

2.3 Inlet and sampling efficiency calculation

A schematic illustrating the tubing used for the inlet and instrumental setup at the measurement site are presented in Fig. S1. To assess losses of particles in the sampling probe and inlet tubing, we calculated overall inlet efficiency as a function of particle diameter using an open-source particle loss calculator (von der Weiden et al., 2009). The calculation accounts for particle losses due to Brownian diffusion, gravitational settling, and inertial impaction in straight tubing and bends. This was done for each instrument by inputting length, diameter, angle of inclination, angle of curvature, and sample flow for each tubing piece between the instrument and the sampling inlet. For all three instruments we found decreasing inlet efficiencies with increasing particle size. The nephelometer and aethalometer have a 50 % inlet efficiency for a particle diameter of 8 μm , whereas the WELAS has a 50 % inlet efficiency for a particle diameter of approximately 6.5 μm . We also calculated the sampling efficiency of the sampling probe at five different wind speeds: 0 m s^{-1} , 2.5 m s^{-1} , 5 m s^{-1} , 7.5 m s^{-1} and 10 m s^{-1} . At wind speeds 0 and 2.5 m s^{-1} there is a depletion in the sampling efficiency of larger particles, whereas at wind speeds 7.5 and 10 m s^{-1} , there is an enrichment in the sampling efficiencies of larger particles. Efficiency curves are presented in Fig. S2 and tubing parameter tables in Tables S1-S3. The results were not used to correct the data.

2.4 Optical parameters

The Single Scattering Albedo (SSA), Absorption Ångström Exponent (AAE), Scattering Ångström Exponent (SAE), spectral variation of SAE (ΔSAE), backscattering ratio (b) and asymmetry parameter (g) were derived from the scattering and absorption coefficients measured by the nephelometer and aethalometer.

SSA was calculated using Eq. (2) as the ratio of scattering coefficient (σ_{sca}) to extinction coefficient, which is the sum of the scattering coefficient and absorption coefficient (σ_{abs}). SSA values at wavelengths (λ) of 470 nm, 520 nm, 660 nm were calculated; thus the scattering coefficients were recalculated for these three wavelengths based on a power law fitting

(Ångström, 1929).

$$SSA(\lambda) = \frac{\sigma_{sca}(\lambda)}{\sigma_{sca}(\lambda) + \sigma_{abs}(\lambda)} \quad (2)$$

135 Typical values of SSA for sea salt and sulfate aerosol are expected to be close to 1 at $\lambda = 550$ nm (Takemura et al., 2002), while fresh BC is highly absorbing and has been shown to have SSA values close to 0.3 at $\lambda = 554$ nm (Bergstrom et al., 2002).

SAE and AAE reflect the spectral variability of light scattering and absorption and were ~~found~~ calculated using Eq. (3) and Eq. (4). SAE was ~~derived from the wavelength pair~~ calculated for the nephelometer wavelength pair (450 / 635 nm) and additionally for the wavelength pair of 450 nm and 550 nm to compare to Cappa et al. (2016). Values that were not directly
140 measured, were recalculated based on power-law fitting.

$$SAE(450nm, 550nm)_{\lambda_1, \lambda_2} = -\frac{\ln[\sigma_{sca}(450nm)/\sigma_{sca}(550nm)]}{\ln(450nm/550nm)} \frac{\ln(\frac{\sigma_{sca}(\lambda_1)}{\sigma_{sca}(\lambda_2)})}{\ln(\frac{\lambda_1}{\lambda_2})} \quad (3)$$

$$AAE(532nm, 660nm)_{\lambda_1, \lambda_2} = -\frac{\ln[\sigma_{abs}(532nm)/\sigma_{abs}(660nm)]}{\ln(532nm/660nm)} \frac{\ln(\frac{\sigma_{abs}(\lambda_1)}{\sigma_{abs}(\lambda_2)})}{\ln(\frac{\lambda_1}{\lambda_2})} \quad (4)$$

SAE is inversely related to particle size: ~~aerosol particles typically found in the coarse mode.~~ Coarse-mode aerosol particles, such as dust, pollen and sea salt, have been reported to show SAE values of 0.5 (Costabile et al., 2013), ~~while small particles.~~
145 Small particles, such as carbonaceous particles ~~originating~~ from fossil fuel combustion or biomass burning, ~~have typical~~ typically show SAE values greater than 1.5 (Cazorla et al., 2013). AAE is related to the chemical composition of the particles: ~~BC.~~ BC (emitted during both fossil fuel and biomass combustion) is known to absorb radiation equally well at all wavelengths, ~~thus having and therefore exhibits~~ AAE values close to 1 (Bond and Bergstrom, 2006), ~~while dust.~~ Dust (e.g. from soil or volcanic eruptions) and brown carbon (BrC; ~~from biomass burning~~) ~~show a wavelength dependency with higher,~~
150 however, exhibit wavelength-dependent absorption, with stronger absorption in the ultraviolet and blue spectral regions (Kirchstetter et al., 2004) ~~(i.e. AAE > 1.5).~~ corresponding to AAE values greater than 1.5. Finally, the Ångström matrix, ~~which combines~~ describes the relationship between AAE and SAE values, ~~can be subdivided into several regions representative for different aerosol~~ in a bivariate plot, effectively subdividing the AAE-SAE space into regions that are representative of different particle
155 types and sources (Cazorla et al., 2013; Cappa et al., 2016). Each point in the matrix corresponds to a pair of AAE and SAE values, and clustering of points in specific regions can be used to identify dominant aerosol types (e.g., large coarse particles, carbonaceous particles, or mixed particles). For a visualization of the full dataset, see Fig. S3.

ΔSAE describes the spectral variation of SAE, calculated as Perrone et al. (2018):

$$\Delta SAE = SAE(450nm, 550nm)_{(\lambda_1, \lambda_2)} - SAE(550nm, 700nm)_{(\lambda_2, \lambda_3)} \quad (5)$$

Here, $\lambda_1 < \lambda_2 < \lambda_3$. While SAE is related to the mean size and relative concentrations of the accumulation and coarse mode aerosol, ΔSAE provides information on the contribution of fine and coarse mode particles leading to a better understanding of the SAE values (Perrone et al., 2018). A negative value indicates a large contribution from a single, fine mode, while a positive value indicates a contribution from both the fine and coarse modes, where higher values represent a larger contribution of the coarse mode particles.

The backscattering ratio, b , is defined as the ratio of backscattering coefficients ($\sigma_{bsca,\lambda}$) to total scattering coefficients ($\sigma_{sca,\lambda}$):

$$b(\lambda) = \frac{\sigma_{bsca}(\lambda)}{\sigma_{sca}(\lambda)} \quad (6)$$

b values range from 0 to 1, where a high fraction of larger particles results in a smaller value of b , as large particles (size larger than wavelength) predominantly scatter radiation in the forward direction (Bohren and Huffman, 2008).

b can also be used to estimate the asymmetry parameter, g (Andrews et al., 2006). g is the cosine-weighted average-mean of the phase function, which describes the angular distribution of the-scattered radiation. It ranges from -1 to 1, representing total backscattering and total forward scattering, respectively, whereas 0 represents symmetric-an equal balance between forward and backward scattering. It is a crucial parameter in radiative transfer models, climate models, and general circulation models used to estimate the direct radiative effect of aerosols (Bellouin et al., 2005; Barman et al., 2024). g was calculated using Eq. (7):

$$g = -7.143889 \cdot b_\lambda^3 + 7.464439 \cdot b_\lambda^2 - 3.96356 \cdot b_\lambda + 0.9893 \quad (7)$$

2.5 Source-receptor modelling with FLEXPART

The origins of the airmasses arriving at the receptor site, where sampling instruments were placed, were simulated with the Lagrangian particle dispersion model FLEXPART version 10.4 (Pisso et al., 2019). The model emits computational particles at the surface that are tracked backward in time by the ambient flow, as defined by meteorological fields that are used as input to the model. In the present study, the meteorological input was based on hourly reanalysis meteorological fields (ERA5) from the European Centre for Medium-Range Weather Forecasts (ECMWF) with 137 vertical levels (up to approximately 80 km) and a horizontal resolution of $0.5^\circ \times 0.5^\circ$ (Hersbach et al., 2020). Simulations extended over 30 days backward in time, a sufficient period to capture more than 98 % of most BC emissions arriving at the receptor, given a typical BC lifetime of 1 week (Bond et al., 2013). FLEXPART includes gravitational settling for spherical particles, dry and wet deposition of aerosols (Grythe et al., 2017), turbulence (Cassiani et al., 2015), unresolved mesoscale motions (Stohl et al., 2005), and deep convection (Forster et al., 2007). The model output consists of a footprint emission sensitivity that expresses the probability of any emission occurring in any grid-cell to arrive at the receptor site and can be converted to model concentration, when coupled with gridded emissions.

Anthropogenic emissions were adopted from the latest version (v6b) of the ECLIPSE (Evaluating the CLimate and Air Quality ImPacts of ShortlivEd Pollutants) dataset, an upgraded version of the previous version (Klimont et al., 2017). The inventory includes emissions from industrial combustion (IND), from the energy production sector (ENE), residential and

commercial emissions (DOM), emissions from waste treatment and disposal sector (WST), transportation (TRA), shipping activities (SHP) and gas flaring emissions (FLR). The methodology for obtaining emissions from FLR specifically over the Russian territories has been improved in ECLIPSEv6 (Böttcher et al., 2021). Biomass burning (BB) from wildfires was adopted from the Copernicus Global Fire Assimilated System (CAM5-GFAS) (Kaiser et al., 2012), a product that provides estimates of the injection altitude of the fire emissions that is crucial for accurate simulation of the simulations of BB dispersion.

3 Results and Discussion

3.1 Main features of optical and physical properties

Figure 2 presents an overview of the aerosol optical properties during the measurement period, each data point represents an average a mean of 10 min, while Table 1 presents numerical values of statistical parameters. Figure 2(a) shows scattering coefficients (σ_{sca}) at three different wavelengths. Two periods with particularly high σ_{sca} are visible, the first from 18 to 20 March and the second from 30 to 31 March. Apart from these two periods, the values of σ_{sca} were below 100 Mm^{-1} . The average median σ_{sca} at 440-450 nm, 525 nm and 635 nm were found to be $35.2-24.42 \text{ Mm}^{-1}$, $29.0-20.24 \text{ Mm}^{-1}$ and $23.7-17.29 \text{ Mm}^{-1}$, respectively. In addition, the σ_{sca} at 440-450 nm was generally higher than at 525 nm and 635 nm, indicating that fine-mode particles contributed significantly to the aerosols. The period with the highest absorption coefficients (σ_{abs}) was observed from 17 to 20 March (Fig. 2(b)). Many sharp peaks were only visible in σ_{abs} and were absent in σ_{sca} , indicating the presence of high-absorbing strongly absorbing particles for short periods of times. These were likely emitted by traffic or biomass burning near the site. Therefore, they are not considered representative of the regional aerosols. The average median σ_{abs} at 370 nm, 470 nm, 520 nm, 590 nm, and 660 nm, 880 nm, 950 nm were $7.41-3.61 \text{ Mm}^{-1}$, 5.32 Mm^{-1} , $4.53-3.06 \text{ Mm}^{-1}$ and 2.20 Mm^{-1} , 3.86 Mm^{-1} , 3.26 Mm^{-1} , 2.37 Mm^{-1} , 2.22 Mm^{-1} , respectively. In general, the absorption coefficients decreased with increasing wavelengths.

Compared to Vavihill, a background site in southern Sweden, median σ_{sca} values at 525 nm in this study ($29.02 \pm 27.39-20.24 \text{ Mm}^{-1}$) were lower than in the Swedish study, where values amounted to 48.0 ± 44.6 medians amounted to 31.4 Mm^{-1} for a similar period of the year (1 March - 1 April 2011). Also, median σ_{abs} values at 520 nm from this study ($4.53 \pm 4.43-3.06 \text{ Mm}^{-1}$) were slightly lower compared to those medians measured at Vavihill, amounting to $6.1 \pm 6.5-3.7 \text{ Mm}^{-1}$ at 520 nm, for the same season (Roldin, 2012; Genberg, 2012). A follow-up study in Vavihill in April 2013 reported a similar value monthly mean of approximately 5 Mm^{-1} at 520 nm (Martinsson et al., 2014), which is similar to our mean values ($4.53 \pm 4.43 \text{ Mm}^{-1}$). Compared to a coastal, Mediterranean city (Lamezia Terme, Italy), characterized by anthropogenic pollution emissions related to traffic and agriculture (Donateo et al., 2020), the mean, the σ_{sca} values at 550 nm during the cold season (October to March from 2015 to 2018) were found higher than those from this study with mean values of 39 median values of 30.93 Mm^{-1} (Donateo et al., 2020).

A median eBC value of $318 \pm 270-231.3 \text{ ng m}^{-3}$ was found during the whole measurement period at Aarhus Bay, as calculated from the σ_{abs} value at 880 nm. Furthermore, the median value of BB%, characterizing the fraction of biomass burning to the eBC load, was 22.9% with a large standard deviation of 11.9% 22.4% . This indicates that biomass burning

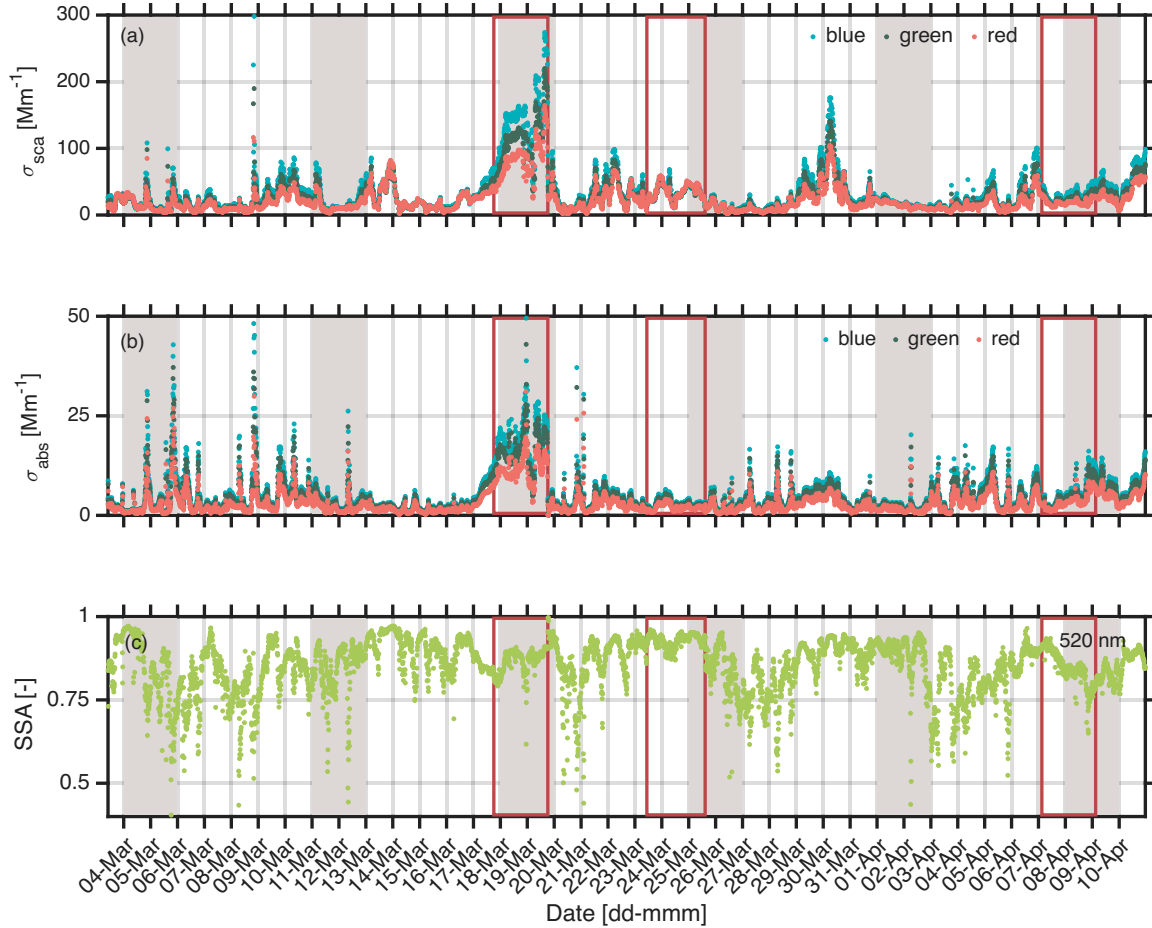


Figure 2. 10 min averages-means of optical properties during the whole measurement period with weekends highlighted in grey, and the three selected cases highlighted with red squares. (a) scattering coefficient σ_{sca} at three wavelengths (blue: 450 nm, green: 525 nm, red: 635 nm); (b) absorption coefficient σ_{abs} at three wavelengths (blue: 470 nm, green: 520 nm, red: 660 nm); (c) single scattering albedo SSA at 520 nm; σ_{sca} at 520 nm was recalculated using power law fitting.

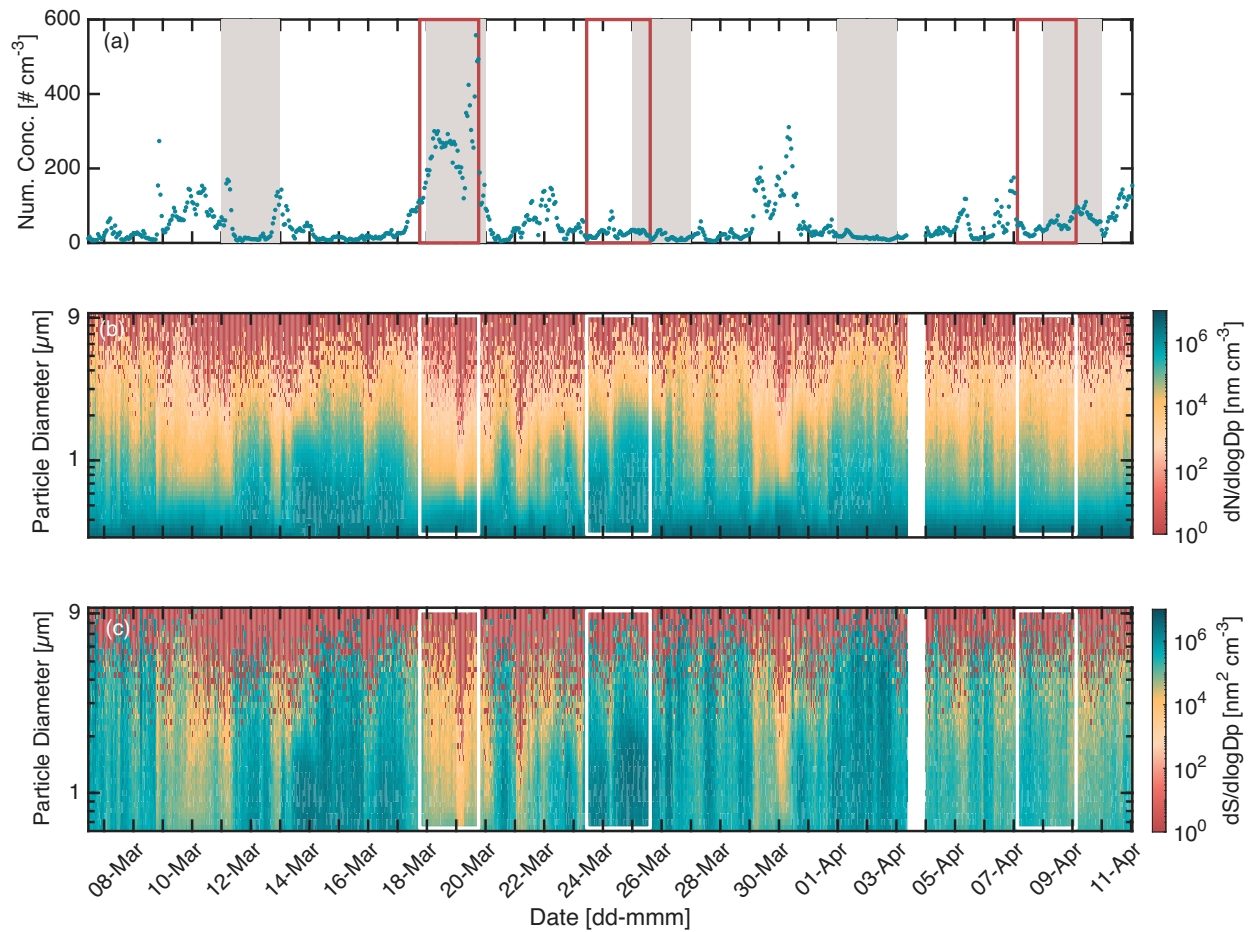


Figure 3. Time series of particle concentration and size distribution during the whole measurement period, with weekends highlighted in grey and the three selected cases with red rectangles. [The WELAS measurements were available from 7 March to 11 April 2023.](#) (a) total number concentration N_{TOT} , (b) number size distribution $dN/d\log D_p$, and (c) surface area size distribution $dS/d\log D_p$.

Table 1. Statistics of optical properties. Mean \pm standard deviation (std) and median, 25th and 75th percentiles (25th, median, 75th) values-of 10 min averaged optical parameters and total number concentration for the whole period (3 March to 11 April) and the three cases.

Parameter	λ (nm)	Whole period			Case 1			Case 2			Case 3		
		mean \pm std	[25,50,75]	mean \pm std	[25,50,75]	mean \pm std	[25,50,75]	mean \pm std	[25,50,75]	mean \pm std	[25,50,75]	mean \pm std	[25,50,75]
$\sigma_{\text{SCA}} [\text{Mm}^{-1}]$	450	35.16 \pm 34.43	[13.65, 24.42, 43.84]	137.64 \pm 46.99	[102.54, 142.48, 154.96]	35.86 \pm 11.13	[27.85, 36.97, 44.03]	30.84 \pm 8.66	[24.21, 29.54, 35.46]				
	525	29.02 \pm 27.39	[11.40, 20.24, 36.31]	109.54 \pm 37.53	[84.18, 114.26, 123.77]	34.93 \pm 10.73	[26.46, 34.90, 43.29]	24.46 \pm 6.30	[19.95, 23.40, 27.73]				
	635	23.71 \pm 20.77	[10.20, 17.29, 29.24]	81.08 \pm 28.11	[61.10, 84.98, 93.39]	35.16 \pm 11.51	[26.34, 34.40, 44.63]	19.14 \pm 4.46	[15.83, 18.85, 20.81]				
$\sigma_{\text{abs}} [\text{Mm}^{-1}]$	470	5.32 \pm 5.31	[2.11, 3.61, 6.49]	19.14 \pm 5.46	[15.84, 18.60, 22.27]	3.09 \pm 1.20	[2.16, 2.88, 3.93]	5.66 \pm 3.22	[3.83, 4.92, 6.48]				
	520	4.53 \pm 4.43	[1.81, 3.06, 5.61]	16.32 \pm 4.71	[13.54, 15.80, 19.29]	2.66 \pm 1.02	[1.87, 2.48, 3.34]	4.83 \pm 2.66	[3.24, 4.23, 5.57]				
	660	3.26 \pm 3.09	[1.35, 2.20, 4.03]	11.60 \pm 3.40	[9.58, 11.21, 13.97]	2.01 \pm 0.74	[1.46, 1.87, 2.49]	3.45 \pm 1.84	[2.30, 3.01, 4.02]				
SSA	470	0.85 \pm 0.08	[0.81, 0.87, 0.91]	0.87 \pm 0.04	[0.84, 0.88, 0.89]	0.92 \pm 0.02	[0.91, 0.92, 0.94]	0.84 \pm 0.05	[0.83, 0.84, 0.89]				
	520	0.85 \pm 0.08	[0.81, 0.87, 0.91]	0.87 \pm 0.04	[0.84, 0.88, 0.89]	0.93 \pm 0.02	[0.92, 0.93, 0.94]	0.85 \pm 0.05	[0.82, 0.84, 0.89]				
	660	0.82 \pm 0.10	[0.76, 0.84, 0.89]	0.82 \pm 0.05	[0.78, 0.84, 0.85]	0.93 \pm 0.03	[0.91, 0.94, 0.95]	0.80 \pm 0.07	[0.76, 0.80, 0.86]				
AAE	470/660	1.37 \pm 0.26	[1.27, 1.39, 1.50]	1.48 \pm 0.10	[1.42, 1.47, 1.55]	1.25 \pm 0.15	[1.17, 1.27, 1.35]	1.41 \pm 0.15	[1.32, 1.41, 1.53]				
	532/660	1.41 \pm 0.28	[1.27, 1.41, 1.55]	1.52 \pm 0.13	[1.43, 1.49, 1.60]	1.32 \pm 0.14	[1.24, 1.34, 1.42]	1.44 \pm 0.17	[1.31, 1.42, 1.57]				
SAE	450/635	1.03 \pm 0.68	[0.45, 1.19, 1.58]	1.55 \pm 0.15	[1.46, 1.56, 1.65]	0.08 \pm 0.45	[-0.18, -0.07, 0.19]	1.35 \pm 0.33	[1.18, 1.35, 1.64]				
	450/550	1.06 \pm 0.70	[0.50, 1.23, 1.61]	1.53 \pm 0.17	[1.43, 1.54, 1.64]	0.10 \pm 0.46	[-0.17, -0.04, 0.22]	1.38 \pm 0.33	[1.20, 1.38, 1.67]				
ΔSAE	450/525/635	0.22 \pm 0.42	[-0.00, 0.19, 0.42]	-0.10 \pm 0.17	[-0.16, -0.10, -0.01]	0.16 \pm 0.33	[0.01, 0.15, 0.31]	0.20 \pm 0.30	[0.07, 0.22, 0.39]				
b	450	0.12 \pm 0.02	[0.11, 0.12, 0.14]	0.10 \pm 0.01	[0.10, 0.10, 0.10]	0.12 \pm 0.01	[0.12, 0.12, 0.13]	0.14 \pm 0.01	[0.13, 0.14, 0.14]				
	525	0.13 \pm 0.02	[0.12, 0.13, 0.14]	0.11 \pm 0.01	[0.10, 0.10, 0.11]	0.12 \pm 0.01	[0.11, 0.12, 0.12]	0.15 \pm 0.01	[0.14, 0.15, 0.15]				
	635	0.14 \pm 0.02	[0.12, 0.13, 0.15]	0.12 \pm 0.01	[0.11, 0.11, 0.12]	0.11 \pm 0.01	[0.10, 0.11, 0.11]	0.16 \pm 0.01	[0.15, 0.16, 0.16]				
g	450	0.60 \pm 0.04	[0.58, 0.60, 0.63]	0.66 \pm 0.02	[0.66, 0.67, 0.67]	0.61 \pm 0.02	[0.60, 0.61, 0.62]	0.58 \pm 0.02	[0.56, 0.57, 0.59]				
	525	0.59 \pm 0.04	[0.55, 0.58, 0.62]	0.65 \pm 0.03	[0.64, 0.66, 0.66]	0.62 \pm 0.01	[0.61, 0.62, 0.63]	0.55 \pm 0.02	[0.54, 0.55, 0.56]				
	635	0.58 \pm 0.05	[0.54, 0.58, 0.62]	0.62 \pm 0.03	[0.62, 0.63, 0.64]	0.64 \pm 0.02	[0.63, 0.65, 0.65]	0.53 \pm 0.02	[0.52, 0.52, 0.54]				
eBC [ng m ⁻³]	880	317.53 \pm 269.97	[155.20, 231.30, 374.70]	1042.85 \pm 312.78	[860.50, 997.20, 1263.80]	218.98 \pm 65.08	[168.43, 213.70, 262.88]	337.48 \pm 151.33	[240.47, 294.40, 371.65]				
BB%	-	22.80 \pm 11.93	[15.19, 22.40, 29.75]	25.61 \pm 5.69	[21.71, 24.79, 28.96]	15.19 \pm 8.26	[10.68, 13.75, 18.62]	24.01 \pm 9.05	[18.34, 24.55, 30.15]				
$N_{\text{TOT}} (\# \text{ cm}^{-3})$	-	58.04 \pm 70.15	[15.19, 22.40, 29.75]	225.53 \pm 111.43	[120.77, 231.77, 275.15]	29.40 \pm 14.27	[20.03, 27.66, 34.63]	52.96 \pm 22.04	[36.92, 51.66, 67.39]				

also contributed significantly to the absorbing aerosols at Aarhus Bay. eBC values at Vavihill, as calculated from presented
225 σ_{abs} values at 880 nm during April 2013 (monthly means), were found to be approximately 380 ng m^{-3} (Martinsson et al.,
2014), which is in the same range as our measurements at Aarhus Bay. ~~On the other hand, (Aarhus: mean of $317.53 \pm$
 269.97 ng m^{-3}).~~ eBC values in Malmö, a coastal city Hyltemossa Research Station, a rural station about 65 km northeast of
Malmö–Copenhagen located in southern Sweden ~~with a similar population to Aarhus, were higher with on average about 500,~~
~~were comparable with a median of 227.5 ng m^{-3} in autumn during March~~ 2018 (Ahlberg et al., 2023). Interestingly, the median
230 eBC concentration at Lamezia Terme ~~was during the cold season was 490 ng m^{-3} (mean: $690 \pm 0 \text{ ng m}^{-3}$)~~ (Donateo et al.,
2020) and thus significantly higher than the ~~mean value in Aarhus during the cold season~~ median and mean values in Aarhus.

Figure 2(c) depicts the single scattering albedo (SSA) at 520 nm, which ranged from 0.28 to 0.99 with a ~~mean and standard~~
~~deviation of 0.85 ± 0.08~~ median of 0.87. Oceanic aerosols are generally expected to have high SSA values close to 1 at
wavelengths from 440 to 1020 nm, while urban aerosols and biomass burning aerosols are typically described by lower values
235 (Dubovik et al., 2002). Median SSA values at 520 nm in Aarhus (this study) and Vavihill were very similar (~~0.85 ± 0.08~~ 0.87
in Aarhus vs. ~~0.88 ± 0.05~~ 0.89 in Vavihill) (Genberg, 2012; Roldin, 2012; Tørseth et al., 2012). The median SSA value at
Lamezia Terme using a wavelength of 637 nm during the ~~same period cold season~~ was 0.78 (min: 0.2; max: 0.99) (Donateo
et al., 2020), which is slightly lower ~~but within the variability of the~~ than the values from this study (~~0.82 ± 0.10~~ Aarhus
median: 0.84 at 660 nm).

240 The ~~average value of median~~ AAE_{470/660 nm} throughout the campaign was 1.39 (mean: 1.37 ± 0.26), which is well com-
parable to ~~values reported previously reported values~~ for aerosols from fossil fuel combustion of 1 to 1.5 (Lee et al., 2012).
The ~~mean value of median~~ SAE_{450/550 nm} was ~~1.15 with a relatively large~~ 1.32 with a mean and standard deviation of $1.15 \pm$
0.74, indicating more variability in the wavelength dependence of the scattering coefficient. This variability can typically be
explained by a change in the dominant aerosol particle mode as larger particles are described by smaller SAE values and vice
245 versa. SAE_{440/675 nm} values for aerosols from fossil fuel combustion were previously reported larger than 1.2 (Bahadur et al.,
2012) indicating smaller particles, while SAE_{440/675 nm} of dust and SAE_{467-660 nm} of sea salt were reported to be less than 0.5
(Bahadur et al., 2012; Costabile et al., 2013), indicating larger particles. This suggests that both fine and coarse mode particles
occurred during this Aarhus Bay campaign, which is also in agreement with the median value of ΔSAE of ~~0.220~~ 0.19, suggest-
ing the presence of both modes (Perrone et al., 2018; Suchánková et al., 2024). Few other studies exist reporting similar data
250 for coastal environments: a monthly mean AAE_{370/950 nm} for April 2013 of approximately 1.3 was found in Vavihill (Sweden)
(Martinsson et al., 2014), in-line with our data, while Donateo et al. (2020) reported a median SAE_{450/700 nm} of ~~1.95~~ 2.14 during
the cold season (October to March from 2015 to 2018) in the coastal city Lamezia Terme (Italy). The substantially lower SAE
value from this study indicates the presence of more large particles in Aarhus Bay compared to the Mediterranean coastal city.

The median backscatter ratios, b , at 450 nm, 525 nm and 635 nm were 0.12, 0.13 and ~~0.14 with the same standard deviations~~
255 ~~of 0.020~~ 0.13. As larger particles scatter predominately in the forward direction, such small b values support the SAE and
 ΔSAE results highlighting ~~the~~ presence of in general larger particles. These values are in line with ~~those found median~~
values measured in Lamezia Terme during the cold season (October to March from 2015 to 2018) of 0.10, 0.11, ~~0.11~~ 0.12 at
450 nm, 550 nm and 700 nm (Donateo et al., 2020), respectively. The ~~mean values and standard deviations~~ median values of

the asymmetry parameter, g , in Aarhus were 0.60 ± 0.04 , 0.59 ± 0.04 , 0.58 ± 0.05 and 0.58 at 450 nm, 525 nm and 635 nm, respectively, which is comparable to ~~those median values~~ at similar wavelengths in Lamezia Terme during ~~the~~ cold season, found to be ~~0.66-0.67~~ (450 nm), 0.63 (550 nm) and ~~0.65-0.62~~ (700 nm) (Donateo et al., 2020).

Figure 3 depicts time series of the total number concentration (N_{TOT} ; Fig. 3(a)), number size distribution (dN/dlogDp; Fig. 3(b)), and surface area size distribution (dS/dlogDp; Fig. 3(c)) measured at Aarhus Bay. Both the total number concentration and size distributions are depicted as hourly ~~averages~~ means. The size distributions are plotted for particles above 300 nm. The changes in N_{TOT} follow the same trend as was observed for σ_{sca} and σ_{abs} ; i.e. the periods that were found to have high σ_{sca} and σ_{abs} ~~have had~~ also high N_{TOT} as seen in Fig. 3(a). From 17 March to 20 March, the highest total particle number concentration was observed, with a maximum concentration of $\sim 550 \text{ \# cm}^{-3}$. Elevated total particle number concentrations were also detected from 9 March to 11 March and from 29 March to 31 March. The ~~average median~~ total particle number concentration throughout the whole period was ~~31.48 # cm⁻³~~ (mean: $58.04 \pm 70.15 \text{ \# cm}^{-3}$). Number size distributions highlight particles between the detection limit of the WELAS and approximately 1 \mu m and surface size distributions more clearly depict the contribution of particles up to 7 \mu m . When comparing the evolution of N_{TOT} with the number and surface size distributions (Fig. 3(b) and 3(c)), it is evident that the period with the highest particle number concentration coincided with the number and surface size distributions dominated by smaller particle sizes, indicating an enhanced abundance of small particles during this period. Generally, when the number and surface size distributions show a mode above 0.5 \mu m in particle diameter, the total number concentration remained below 200 \# cm^{-3} and was typically less than 100 \# cm^{-3} .

3.2 Daily patterns of scattering and absorption coefficient and eBC

Figure 4 illustrates the median diurnal variation of σ_{sca} , σ_{abs} and eBC concentrations during weekdays (4(a), 4(c), and 4(e)) and weekends (4(b), 4(d), and 4(f)). Median values were calculated excluding the data from 17 to 19 March, which showed the highest concentrations. ~~Except for σ_{sca} during weekdays,~~

~~The absorption coefficient σ_{abs} and σ_{sca} show similar trends~~ shows clear diurnal patterns on both weekdays and weekends, with two major peaks, one in the morning and one during the evening/night, highlighting the typical rush hours. The morning peak is generally around 5-9 h, while the evening peak typically lasts from 17-23 h.

~~The morning peak appeared earlier and sharper during weekdays than during weekends, coinciding with expected traffic patterns during weekdays and weekends, respectively. The evening peak was slightly lower during weekdays than during weekends (Fig. 4(c) and 4(d)).~~ The scattering signal did not show a clear pattern during weekdays, however, two distinct peaks could be observed during the weekend (Fig. 4(a) and 4(b)). Furthermore, the difference between the three wavelengths was larger on weekdays than on weekends. During weekends, σ_{sca} values ~~were very close to each other at different wavelengths~~ were very similar between 10 and 18 h, indicating that both smaller and larger particles were present, ~~while the signal.~~ During weekdays, however, σ_{sca} at 450 nm was clearly higher during weekdays in the same time period, indicating a larger fraction of small particles. ~~The absorption signal in the morning appeared earlier and sharper during weekdays than during weekends, coinciding with expected traffic patterns during weekdays and weekends, respectively. The evening peak was slightly lower during weekdays than during weekends (Fig. 4(e) and 4(d)).~~ Figures S3

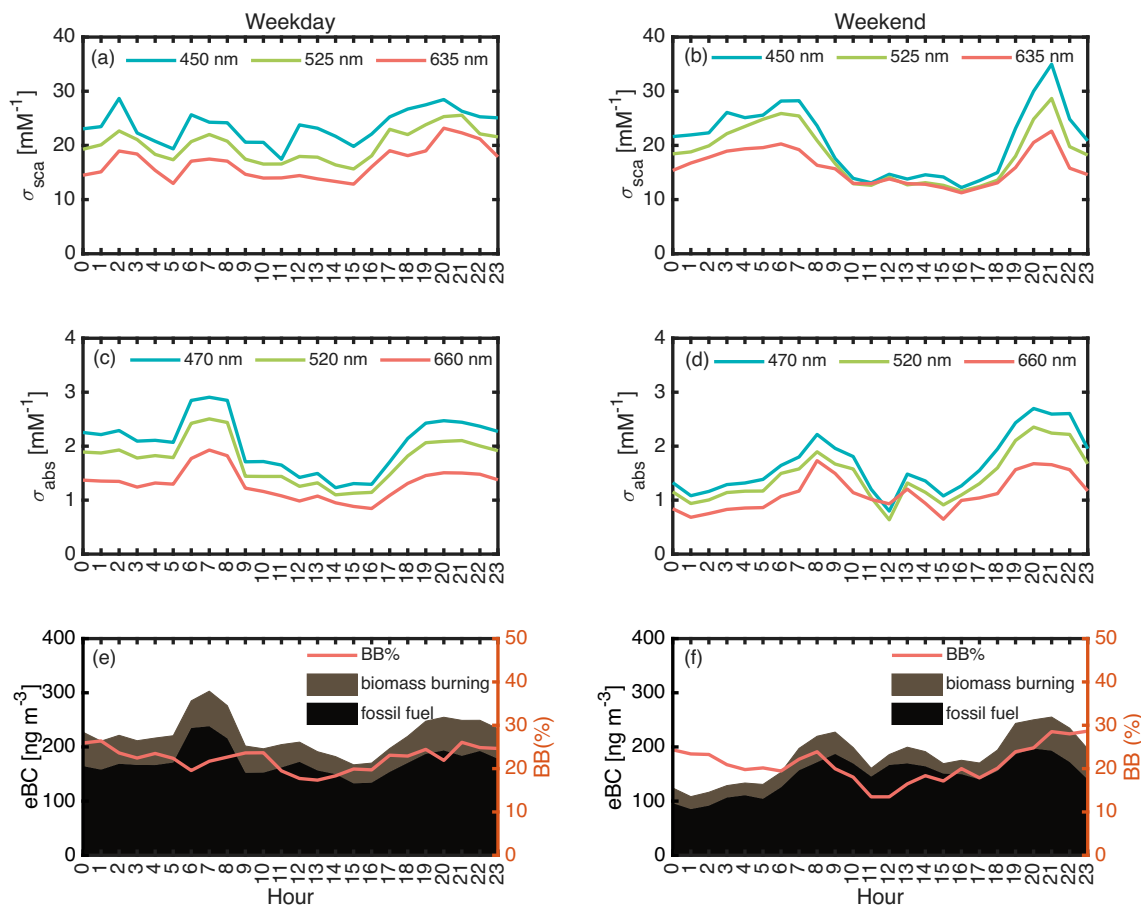


Figure 4. Diurnal variation of (a) median σ_{sca} values at three wavelengths during weekdays, (b) σ_{sca} at three wavelengths during weekends, (c) median values of σ_{abs} at three wavelengths during weekdays, (d) median values of σ_{abs} at three wavelengths during weekends, (e) median values of eBC during weekdays, (f) median values of eBC during weekends. In (e) and (f), brown colours represent contribution from biomass burning and black colours from fossil fuels. Source apportionment is based on BB%, see text for more information.

Overall, the differing diurnal behaviour in scattering and absorption indicate that non-absorbing or weakly absorbing aerosol components contribute substantially to the observed scattering signal, particularly during weekdays. Figures S4(a) and S3S4(b) show the temporal evolution of ~~the diurnal patterns~~ diurnal pattern of scattering and absorption coefficients, highlighting that except for the last week, the daily patterns were quite comparable throughout the course of the campaign.

The contribution from combustion of fossil fuels was calculated by subtracting the contribution from biomass burning from the total eBC. Figure 4(e) and 4(f) present eBC values subdivided in fossil fuel (black) and biomass burning (brown) contributions. Fossil fuel contributed the most to eBC concentrations, above 70 % during both weekdays and weekends. BB% varied from 17 % to 26 % during weekdays, and from 13 % to 29 % during weekends. During weekdays, a prominent peak during rush hour was observed from around 5 to 8 h, underlining the important influence of traffic emissions at the site. In particular, the increase on weekdays was larger than that on weekends. During the evening, an increase in both fossil fuel and biomass burning contributions was found on weekends and weekdays. Notably, the contribution from biomass burning was higher during weekend, where it increased from 20 % to 29 % between 18 and 23 h. This could arise from the increased use of domestic fireplaces during weekends and bonfires.

In addition to human activities such as traffic and biomass burning, changes in the planetary boundary layer (PBL) height may impact daily patterns. A high PBL typically leads to dilution of surface emissions and in general lower concentrations, while a lower PBL ~~leads to up-concentrations of emissions~~ hinders dispersion thus favoring higher concentrations. Solar radiation is the major driver for PBL height and can be used as a proxy. Figure S4-S5 shows the overall radiation time per day and Fig. S5-S6 illustrates the diurnal variation of solar radiation during the campaign. The typical radiation pattern showed an increase from around 7 to 8 h and a decrease around 19 to 21 h, where the decrease shifts to later hours at the end of the campaign. This coincides well with the time the morning peaks start to decrease and the time the evening peaks start to increase.

3.3 Comparison between measured and modelled black carbon

Figure 5 highlights a very good agreement between the temporal evolution of total eBC, as measured by the aethalometer, and BC, as modelled by FLEXPART analysis. FLEXPART separates the contribution from black carbon into eight different sources (see Fig. S6-S7 for the time series of the individual sources). On average, the main BC sources contributing to the air masses in Aarhus Bay during the campaign were DOM (~ 49 %), BB (~ 26 %), and TRA (~ 19 %) ~~and while minor contributions from IND (~ 3 %), while minor contributions from FLR (~ 2 %), SHP and WSD (~ 1 %) and ENE (~ 0 %) were found. Overall modelled values underestimated the BC mass concentrations by ~ 60 %.~~

An overall comparison to FLEXPART data yielded a mean fractional bias (MFB) of -51.9 % and a root mean squared error (RMSE) of 221 ng m⁻³. While several previous studies comparing FLEXPART BC concentrations with observations in background stations found good agreement (Popovicheva et al., 2025; Fang et al., 2025; Yttri et al., 2024; Fang et al., 2023; Popovicheva et al., 2023), a comparison in urban areas is more challenging due to difficulties arising when local emissions have to be considered. The superposition of transported and emitted BC makes a direct comparison with FLEXPART data difficult leading to more smoothed modelled lines as compared to the observations. Local pollution is mainly characterized by sharp BC peaks, as seen in Fig. 5, and the lack of these in the modelled BC indicates missing sources in the emission inventories thus leading to an

underestimation by the model. Similar results were also found in the cases of urban Vilnius (Lithuania) and Warsaw (Poland) (Hey et al., 2026). The BC trend in Aarhus bay was however captured very well by FLEXPART.

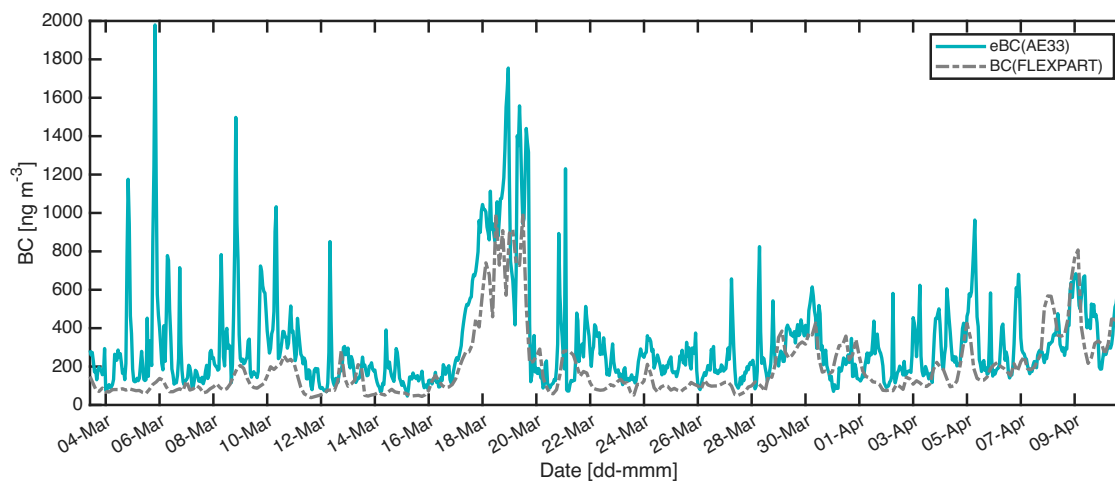


Figure 5. Black carbon mass concentrations measured by the aethalometer (blue solid line) and modelled using FLEXPART (grey stippled line) combining the all source contributions.

3.4 Characterization of three special air mass types

330 We selected three special cases from the entire measurement period ~~that were characterized,~~ characterised by different air mass types:-

1. Case 1: "Local pollution": This period lasted 48 hours, from 17 March 18:10 to 19 March 18:20. It was characterized by high scattering and absorption coefficients, reaching the highest values during the campaign. The measured wind speed (~~2.23-1.48~~ $\pm 1.72 \text{ m s}^{-1}$; median: 3.10 m s^{-1}) and radiation were relatively low during this period, as recorded at the DMI station, providing favourable conditions for particle accumulation. The start time was determined to be when $\sigma_{\text{sca},525}$ and $\sigma_{\text{abs},520}$ were higher than their campaign means and SSA_{520} lower. The end time was set to be when $\sigma_{\text{abs},520}$ decreased below the campaign mean.
2. Case 2: "High Single Scattering Albedo": This period lasted 52 hours, from 23 March ~~10:40-11:00~~ to 25 March ~~14:40-15:00~~. During this period, highest SSA values were reached, as well as higher wind speeds (~~5.42-5.40~~ $\pm 1.87-1.88 \text{ m s}^{-1}$; median: 5.40 m s^{-1}), with a maximum of 10.2 m s^{-1} (from DMI station). This combination could favour a marine emission source. The start time was determined to be when both $\sigma_{\text{sca},525}$ and SSA_{520} exceeded their campaign means and the end time when SSA_{520} decreased below its mean value for the whole period.
3. Case 3: "Long-range transport": This case lasted 48 hours, from 7 April 3:00 to 9 April 3:00. Based on the results of the FLEXPART model, significant contributions to BC originated from the "Asia" and "Russia" sectors. Wind speed

345 was low with a mean value of $2.65 \pm 1.66-1.22 \text{ m s}^{-1}$ and a median value of 2.80 m s^{-1} , also the wind direction mainly originated from Northeast and East-Northeast (from DMI station), in agreement with the FLEXPART results. The start time was determined to be when the contribution from Asia or Russia was higher than its mean contribution during the whole measurement period and the end time when both contributions decreased below their mean values throughout the campaign.

350 Figure S7-S8 presents footprint analysis for each of the selected cases while Fig. S8-S9 shows CO, PM₁₀, and NO_x data from DCE station, highlighting the case periods. Figure S9-S10 shows time series of wind speed and wind direction during the entire measurement period.

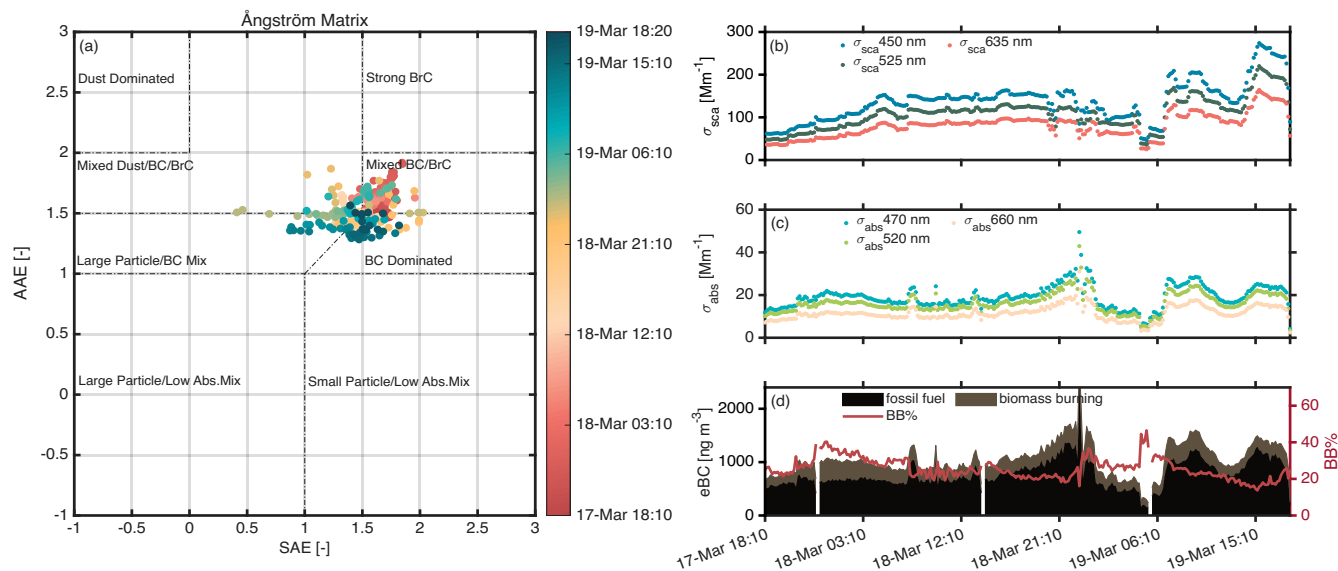


Figure 6. Optical properties and eBC concentration during Case 1. (a) Ångström matrix colour-coded by time; (b) time series of scattering coefficients; (c) time series of absorption coefficients; (d) source apportionment of eBC based on the biomass burning percentage (BB%) provided by the aethalometer.

3.4.1 Case 1:

Both σ_{sca} and σ_{abs} were around three to four times higher than the mean of the entire measurement period, as seen in Table 355 ~~??~~. Also, the median total number concentration was $231.77 \# \text{ cm}^{-3}$ (mean: $225.53 \pm 111.43 \# \text{ cm}^{-3}$ and thus almost four) and thus about seven times higher than the median value during the whole period (almost four times higher than mean value). NO_x and CO are often used as tracers of pollution as they are mainly emitted from fossil fuel combustion and CO may also originate from biomass burning. NO_x has a short lifetime of around one day and a limited travel range usually below one km Seinfeld and Pandis (2016) (Seinfeld and Pandis, 2016) and is thus indicative of local emissions. CO, on the other hand, has a 360 lifetime of below 1 year and a travel range of about 50 km Seinfeld and Pandis (2016), thus representing more regional air

masses. NO_x concentrations varied from approximately 15 to 109 ppb with ~~an average value of 49 ppb and a mean value of 48.22 ppb~~, a standard deviation of ~~23 ppb~~ 22.67 ppb and median value is 46.32 ppb (Fig. [S10S11](#)). This is 160 % higher than the mean NO_x mixing ratios (17.96 ± 17.31 ppb; median: 12.25 ppb) measured during the entire measurement period at that location. Meanwhile, the mean CO concentration during Case 1 was 0.29 ± 0.09 ppm ~~with a median of 0.27 ppm~~ ranging from 0.16 ppm to 0.59 ppm. This is 55 % higher than the mean value over the whole measurement period of 0.18 ± 0.05 ppm. Thus, ~~both~~ the elevated levels of NO_x and CO indicate that Case 1 was dominated by local emissions most probably originating from ships or local traffic or other high-temperature combustion sources such as biomass burning. FLEXPART footprint analysis for Case 1 revealed a clear continental contribution (see Fig. [S7S8](#)) and an age of 3 days for on average 76 % of BC data, where 91 % of BC originated from Europe (see Fig. [S11S12](#)), suggesting that the aerosols were rather fresh and probably emitted from local sources. FLEXPART simulated BC mass concentrations were 35.4 % (RMSE = 475 ng m⁻³) lower than the observations, representing a smaller deviation than the average bias observed over the full campaign.

As illustrated in Fig.6(a), most data points were concentrated in the "BC-dominated" and "mixed BC/BrC" zones. The ~~average-mean~~ values of AAE_{532/660 nm} and SAE_{450/550 nm} were 1.52 and 1.49, respectively. Figure 6(d) shows that the contribution of fossil fuel combustion to eBC was significant during this period. Aerosols from fossil fuel combustion represented 75.03 % of the eBC concentration, ranging from 52.51 % to 86.31 %, which was slightly higher than during the entire measurement period. Figures 6(b) and (c) present the temporal evolution of σ_{sca} and σ_{abs} during Case 1. It is evident that σ_{sca} increased slightly until 18 March 5:00, while σ_{abs} first increased but then stopped increasing earlier on 17 March 23:00 and subsequently decreased until 18 March 5:00. During this period, aerosols gathered in the "mixed BC/BrC" range as biomass burning had a significant impact on eBC during this time (d). After 18 March 5:00, both σ_{sca} and σ_{abs} remained at a constant level until 18 March 20:00. From 18 March 20:00 to midnight of 19 March, σ_{sca} and σ_{abs} show different trends: while σ_{sca} shows two dips, σ_{abs} , on the contrary, increased to reach the highest values. During the σ_{abs} peak, the data points were gathered in the "BC dominated" zone (a) and a clear signal from fossil fuels is visible in the source distribution plot (d). After midnight on 19 March, both scattering and absorption coefficients started to decrease, possibly due to a change in the meteorology, as all data recorded at the DCE station decreased simultaneously with the optical signals as seen in Fig. [S10S11](#). At the end of Case 1, both σ_{sca} and σ_{abs} increased again and then stabilized. Notably, σ_{sca} reached the highest values during this end phase of Case 1. Here, particles were gathered in the "large particle/BC Mix" zone and moved towards the "BC dominated" zone (a). Figure 6 (d), illustrates that BB% continuously decreased during this period, while traffic emissions dominated the signal. Figure S13 displays the size distributions as dN/dlogD and dS/dlogD during Case 1.

3.4.2 Case 2:

During Case 2, the highest SSA values during the whole campaign with a median and mean value of 0.93 at 520 nm were recorded, indicating that highly scattering particles constituted a significant portion of the aerosols throughout this period. As shown in Table ~~??, the~~ 1, the median and mean scattering coefficients were higher in Case 2 (34.62 ± 10.91 median: 34.90 Mm⁻¹ at 525 nm) than during the entire period (29.02 ± 27.39 20.24 Mm⁻¹ at 525 nm). Conversely, the absorption coefficients were lower during Case 2 (2.64 ± 1.03 median: 2.48 Mm⁻¹ at 520 nm) than during the whole measurement period ($4.53 \pm$

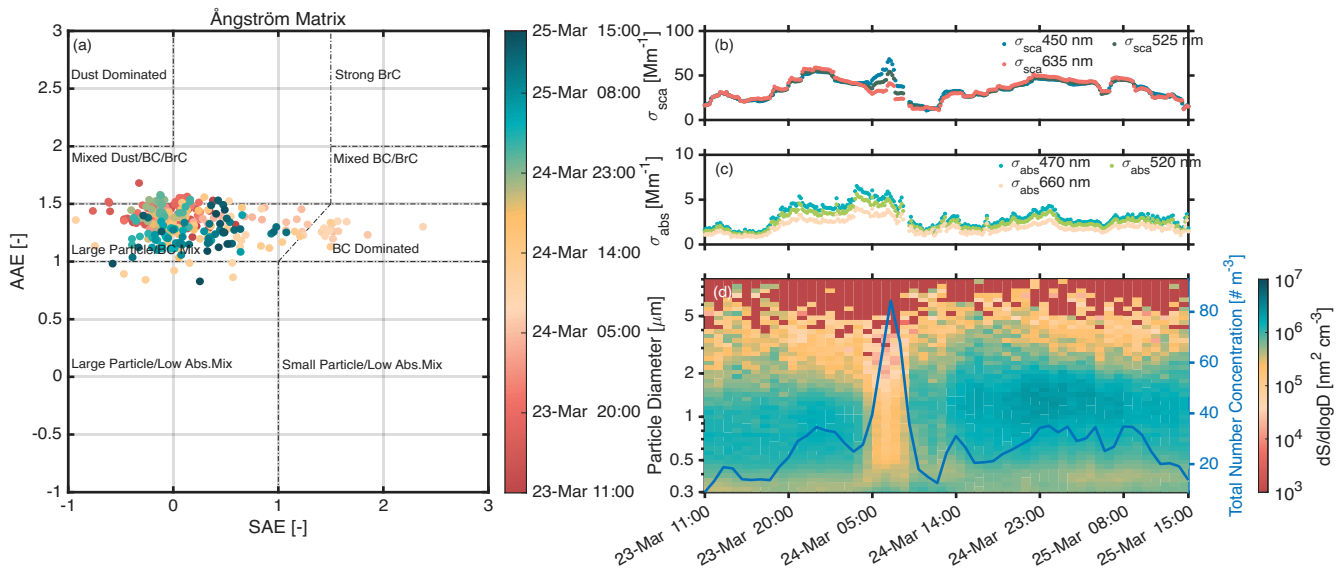


Figure 7. Optical and physical properties during Case 2. (a) Ångström matrix colour-coded by time; (b) time series of scattering coefficients; (c) time series of absorption coefficients; (d) time evolution of surface area size distribution and total number concentration.

395 4.43 median: 3.06 Mm⁻¹ at 520 nm). Consequently, the SSA values were comparatively high in relation to the values during the entire period (median: 0.93 ± 0.02 at 520 nm).

The AAE median and mean AAE_{532/660 nm} values during Case 2 (median: 1.34; mean: 1.32 ± 0.14) were slightly lower than the median and mean of the entire period (median: 1.41; mean: 1.41 ± 0.28). The average SAE median and mean SAE_{450/550 nm} values during Case 2 (0.17 median: -0.04; mean: 0.10 ± 0.520.46) were significantly lower than the average median and mean values for the entire period (1.15 median: 1.23; mean: 1.06 ± 0.740.70), suggesting that large particles dominated the overall aerosol population. Thus, in the Ångström matrix (Fig. 7(a)), the data are clustered together in the "Large particle / BC mixture" zone. Only during midnight on 24 March (orange points), data points were shifted to the "BC dominated" zone before shifting back to the "Large particle/ Low Abs mix" zone (Fig. S14 displays NO_x, CO and eBC data).

Figure 7(b) shows that, during this shift, σ_{sca} at different wavelengths was clearly different, while the signal mostly overlapped during the rest of Case 2, leading to low SAE values that are typical for large particles. The absorption coefficients (c) generally followed the trend of scattering, except for the special short period observed on 24 March, which lasted approximately 3 hours.

Figure 7(d) highlights that in terms of surface area, particles of 0.35 to 5 μ m in diameter dominated, with a mode diameter around 1 μ m. Clarke et al. (2003) reported that breaking waves produce aerosols in all sizes from 10 nm to greater than 10 μ m. Additionally, the size distributions clearly point out that during the special period on (Fig. S15 additionally illustrates the size distribution as dN/dlogD). On 24 March, the particle number concentration strongly increased to approximately 75 # cm⁻³ and

sizes below 0.3 μm dominated, when consistent with when data points shifted to the "BC dominated" emissions were detected.
The higher wind speeds recorded zone in the Ångström matrix.

415 Breaking waves can produce aerosols across a wide size range, from 10 nm to > 10 μm (Clarke et al., 2003), supporting the interpretation that marine coarse particles could have contributed to the observed high scattering. The elevated scattering coefficient during Case 2, together with the low SAE values and measured larger particle sizes suggest a marine influence. The values that suddenly appeared in the "BC dominated" zone are likely influenced by these coarse-mode particles, including sea-salt aerosols. However, the observed contributions by large coarse particles above approximately 8 μm may be underestimated due to the losses in the inlet and tubing.

420 Supporting evidence for marine influence includes higher wind speeds during Case 2 and FLEXPART footprint analysis indicating air masses arriving from the ocean (see Fig. S8). Periods of elevated SSA and scattering are therefore consistent with a contribution from coarse sea-salt particles, while the short BC-dominated event could have been due to ship or local local ship or traffic emissions. FLEXPART footprint analysis for Case 2 revealed a contribution from air masses arriving from the ocean. The trend of the modelled and observed black carbon is well comparable, with FLEXPART being able to capture the short BC peak (see Fig. S7), thus supporting an impact by marine particles5), while the overall values were underestimated by the model (MFB = -78.7 %, RMSE = 128 ng m^{-3}).

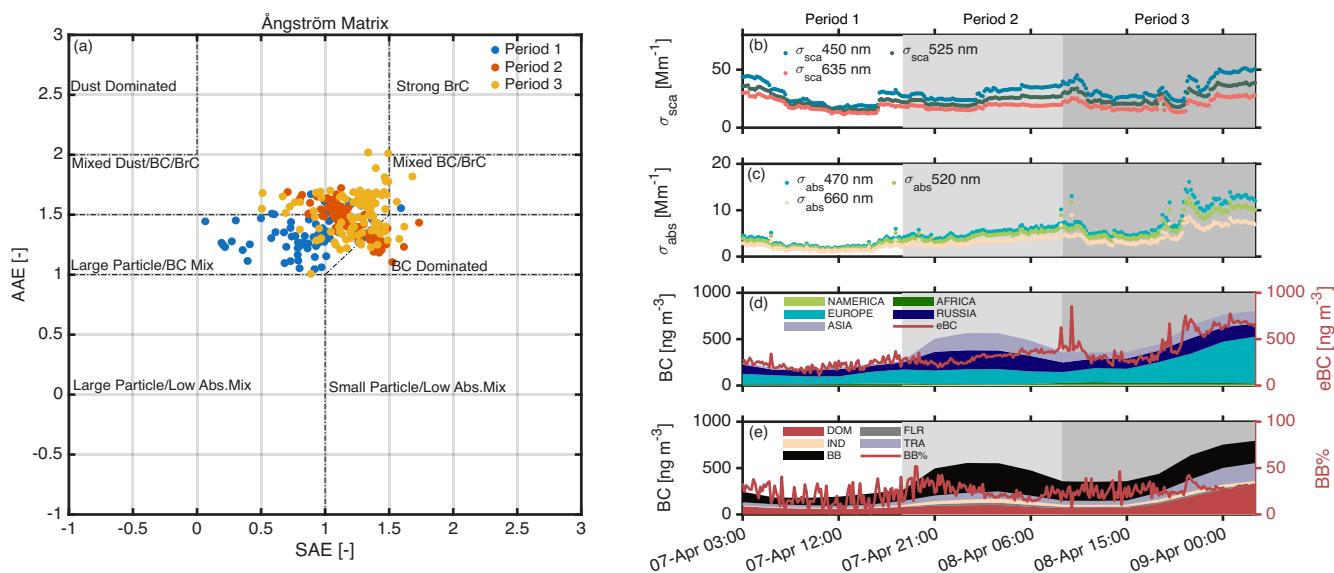


Figure 8. Optical properties, eBC concentrations and FLEXPART results during Case 3. (a) Ångström matrix colour-coded by time; (b) time series of scattering coefficients; (c) time series of absorption coefficients; (d) continental contribution to the BC concentration from FLEXPART and eBC from aethalometer; (e) source contributions of BC from FLEXPART (DOM: residential and commercial combustion, FLR: gas flaring, SHP: shipping activities, TRA: surface transportation, ENE: power plants and energy conversion, IND: industrial combustion, WST: waste burning, BB: biomass burning.) and BB% from aethalometer shown with the red line.

3.4.3 Case 3:

According to the FLEXPART model results, Russia and Asia contributed significantly to the BC concentrations during Case 3, leading to a more "long-range" dominated aerosol. ~~This is also clearly visible in the~~ The FLEXPART footprint analysis for
430 Case 3 (see Fig. S7) is shown in Fig. S8.

Figure 8 shows the measured optical properties and source apportionment, and continental contributions of BC from the FLEXPART model results. Based on Fig. 8(d), Case 3 was subdivided into three periods: PERIOD 1: From 3:00 to 19:00 on 7 April, with a contribution from Russia of around 34 %, 50 % from Europe and 8 % from Asia. PERIOD 2: From 19:00 on 7 April to 9:00 on 8 April, with increased contributions from Asia and Russia, reaching around 27 % and 34 %, respectively,
435 while contributions from Europe were 34 %. PERIOD 3: From 9:00 on 8 April to 3:00 on 9 April, with contributions from Asia and Russia remaining constant (approximately 20 % and 26 %) while the contribution from Europe increased over time to reach 48 %.

Figure 8(a) shows the Ångström matrix, which is colour-coded by the 3 periods introduced above. The points are initially located in the "Large Particle/BC Mix" zone (PERIOD 1), then shift to the "Mixed Dust/BC/BrC" and "BC Dominated" zones
440 (PERIOD 2), and finally spread between the "Mixed Dust/BC/BrC", "BC Dominated", "Mixed BC/BrC" and "Large Particle /BC Mix" zones during PERIOD 3. This indicates that PERIOD 1 contained a high fraction of large particles, then the average mean particle size decreased while the BC fraction increased during PERIOD ~~2-2~~ (Fig. S17 illustrates size distributions during Case 3). During PERIOD 3, the BC and BrC fractions increased while SAE values increased, highlighting smaller particles.

Figure 8(b) illustrates the scattering and (c) absorption coefficients at different wavelengths over time. During PERIOD 1,
445 the scattering coefficient at 525 nm decreased from 40 to 10 Mm^{-1} , and the absorption coefficient at 525 nm decreased from 4 to 2 Mm^{-1} . The average mean SSA value was 0.90. The similar σ_{sca} values at the three wavelengths, lead to the observed clustering in the "Large Particle/BC Mix" zone (Fig. 8(a)). During PERIOD 2, the scattering and absorption coefficients began to increase. The SSA dropped to 0.84, indicating an increase in the contribution by absorbing aerosols. During PERIOD 3, the scattering and absorption coefficients first briefly decreased and subsequently increased again, fluctuating but both following
450 similar trends, while SSA values decreased to 0.80.

Figure 8(d) ~~highlights that the temporal evolution of BC concentration from FLEXPART and eBC concentration aethalometer measurements agreed well during case demonstrates a strong agreement between black carbon concentrations simulated by FLEXPART and those measured by the aethalometer during Case 3. Only during PERIOD 2 the modelled BC concentrations appeared higher than the measured eBC concentrations. The mean BC concentration obtained from the FLEXPART model~~
455 ~~during Case 3 was 423.6~~ The model slightly overestimated the observations by 15.8 %, with RMSE of 132 ng m^{-3} , while the aethalometer's mean eBC concentration. This good performance is consistent with the dominance of long-range transport during Case 3 was 376.2 ng m^{-3} , a process that FLEXPART is well suited to represent.

Figure 8(e) divided BC into different emission sources. According to FLEXPART, biomass burning (BB) significantly contributed to PERIODS 1-3, especially PERIOD 2. BC concentrations from BB reached highest values with increasing contribu-
460 tions from Asia and Russia, while transport (TRA) and domestic (DOM) emissions increased during PERIOD 3 simultaneously

with an increasing contribution from Europe (Fig. 8(d)). The median and mean BB% from the aethalometer was approximately 24 were 24.55 % and 24.01 ± 9 % during the whole case study 9.05 % during Case 3 and its evolution during PERIODS 1-3 appeared rather constant, not following the FLEXPART BB signal, ~~which is 48 ± 9 %~~. On the other hand, the evolution of the eBC concentration is well comparable with the FLEXPART signal from the domestic sector. A discrepancy between BB% and
465 FLEXPARTs' BB concentration could potentially arise from the fact that while FLEXPART clearly separates between BB and DOM, the signal ~~might be~~ is most probably mixed in the aethalometer measurement.

The mean NO_x mixing ratio measured at the DCE station during this case, was 13.30 ± 8.46 ppb with a median value of 10.94 ppb, which was lower than the whole campaign period ~~average of~~ (17.96 ± 17.31 ppb; median: 12.25 ppb). Similarly, the mean CO concentration was 0.16 ± 0.03 ppm, the median value is 0.152 ppb and thus comparable to the mean value of the
470 whole period (0.18 ± 0.055 ppm; median: 0.165 ppm). Figure ~~S15-S16~~ shows that NO_x decreased sharply below 5 ppm around 00:00 on 8 April (PERIOD 2), and then increased again. However, the eBC concentration continually increased while the NO_x concentration decreased. This suggests that the BC was likely transported from other regions, as FLEXPART suggested, such as Asia and Russia, instead of being emitted locally. During PERIOD 3, the NO_x and eBC concentrations increased drastically, indicating that the pollutants were derived from local emissions. This is consistent with the BC results from the FLEXPART
475 model and the increased contribution from Europe during this period.

4 Conclusions

Aerosol properties were investigated during five weeks from March to mid April 2023 at the coastal site - Aarhus Bay, Denmark. Aerosol absorption and scattering coefficients as well as size distributions were continuously monitored to investigate the aerosol type and infer the main aerosol sources during the selected period. Specifically, the study aimed to assess the importance
480 of anthropogenic vs. natural, including marine, sources at this coastal area. Additionally, a comparison to footprint, air mass source and age analysis ~~from~~ with the FLEXPART model was carried out.

Scattering and absorption coefficients were relatively low during the whole period ($\sigma_{sca} = 29.02 \pm 27.39$; median: 20.24 Mm⁻¹, $\sigma_{abs} = 4.53 \pm 4.43$; median: 3.06 Mm⁻¹ at $\lambda = 525$ nm), similar to what was found in other Scandinavian locations (Genberg, 2012; Roldin, 2012; Tørseth et al., 2012) and lower compared to Mediterranean coastal cities (Donateo et al.,
485 2020). SSA varied from 0.28 to 0.9, pointing to both, periods with high absorbing, most likely anthropogenic, and highly scattering, potentially marine, influences. The wavelength dependence of absorption, highlighted a substantial impact from biomass burning aerosols ~~of on average 22.90 ± 11.93 % (BB%) of 22.40 % (median)~~. This can be attributed to emissions from domestic fireplaces which are common in the Aarhus area (Plejdstrup et al., 2021). The wavelength dependence of the scattering coefficient showed large variability due to changes in the particle size distribution. This was corroborated by the
490 recorded size spectra showing periods with high number concentrations and mostly, small sizes and several periods with lower number concentrations and particle diameters above 1 μm. Compared to a Mediterranean coastal city during a similar period, particles in Aarhus appeared on average larger (Donateo et al., 2020). Daily patterns in Aarhus Bay revealed a diurnal variation related to traffic and biomass burning emissions detected in the absorption coefficients, both during weekdays and weekends.

495 Interestingly, scattering coefficients revealed a pattern only during weekends, with highest scattering coefficients during the evening hours between 18 and 23 h.

Wavelength dependences in scattering and absorption were explored by the Ångström matrix and combined with FLEXPART analysis to pinpoint major aerosol sources in Aarhus Bay. Periods with local anthropogenic pollution, highly scattering aerosols, and particles from long-range transport were observed. Using the Ångström classification scheme proposed by (Cappa et al., 2016), aerosols during the whole campaign were to 48 % characterized by 'BC dominated', 31 % by 'Large Particles/BC mix', 500 7.7 % by 'Large Particles/Low Abs. Mix', 4.3 % by 'Small Particles/Low Abs. Mix', 4.2 % by 'Mixed Dust/BC/BrC', 4.1 % by 'Mixed BC/BrC' and 0.85 % by 'Strong BrC' (see Fig. S17S3). According to FLEXPART the ~~average-mean~~ continental contribution to BC concentrations was ~ 71 % from Europe, ~ 10 % from Russia, ~ 9 % Asia, ~ 6 % North America, ~ 2 % Africa, ~ 1 % Central America and the Ocean. Besides, the air mass age as modelled by FLEXPART underscores the mixture of both freshly emitted and several week-old air masses present in the Aarhus Bay area (see Fig. S18).

505 In particular, three distinct cases were chosen to describe typical aerosol properties in Aarhus Bay. "Case 1" ~~describes depicts~~ a typical local pollution event, characterized by highest scattering and absorption coefficients and simultaneously strongly elevated NO_x and CO mixing ratios. According to the Ångström matrix, most data could be described as 'BC-dominated' and 'mixed BC/BrC', with ~~on-average-a mean of~~ 75 % originating from fossil fuel combustion. Particle number concentrations reached highest values but contained predominately particle sizes below 500 nm. eBC data compared well with modelled 510 values from FLEXPART highlighting the large contribution from fossil fuels. FLEXPART analysis showed a clear influence by continental air masses. "Case 2" highlights the period with the highest SSA values during the measurement campaign. Both SAE and size distribution measurements indicate the presence of large particles that are mainly described by the 'Large Particles/BC mix' area in the Ångström matrix. The higher wind speeds during the "Case 2" period and lower absorption coefficients, could indicate a larger contribution by marine particles during this period, which is also suggested by FLEXPART 515 footprint analysis. "Case 3" denotes an event with air masses from long-range transport. FLEXPART analysis yielded a larger contribution by the sectors "Russia" and "Asia" impacting BC concentrations particularly at the beginning of "Case 3", which were characterized by smaller particles, as evidenced by smaller SAE values, compared to local events dominated by the "Europe" sector.

Data availability. Data will be available on Zenodo.

520 *Author contributions.* B.R. designed and supervised the research; Z.T., J.S.T. and B.R. conducted field experiments; Z.T. analysed the scattering and absorption measurements, J.T.S. analysed the spectrometer measurements and performed loss calculations; N.E. and S.E. performed FLEXPART simulations; B.R., A.M., H.S. and M.B. contributed to interpreting the results. Z.T., J.T.S. and B.R. wrote the paper with contributions from all co-authors. All authors read and reviewed the manuscript.

Competing interests. The authors declare no conflict of interest.

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