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

We would like to thank the two reviewers for their thorough and very constructive comments. We appreciate the input and find that the comments have helped improve clarity of our manuscript. We have addressed the comments in a point-by-point fashion below and revised the manuscript accordingly. Our answers to the comments are given below in blue letters**,** while the reviewer comments are given in *black italics*. Additionally, we added the changes we made in the revised manuscript in **blue bold letters**.

Reviewer 1:

The present study characterises aerosol sources through dispersion modelling on optical and size distribution data measured near the coast of Denmark which overlooks Aarhus Bay.

The study includes literature comparisons of the 1-month-worth of measured data and a focus on three different showcase scenarios. This leads to a complicated and very interesting picture of the different aerosol sources at play in the Aarhus Bay area and with the FLEXPART model authors also investigate their origin.

Gaining better insights into the complex system of atmospheric aerosol is fundamental. This study focuses on coastal regions where the interaction between marine and continental air masses is still largely understudied. I recommend this article be published after the authors address the following list of comments and issues.

We thank the reviewer for this important assessment.

General comments

Environmental data is often distributed following a log-normal distribution. The heavy-tailed nature of log-normal distributions causes relatively large standard deviations compared to the distribution's mean value. This is especially true when considering extensive parameters like σ_{sca}, σ_{abs} and eBC. To make the comparisons of section 3.1 and of the three chosen cases more statistically relevant, the authors should consider including other statistical parameters other than mean±standard deviation in the main text and not just in Table 1, e.g. medians and other percentiles.

The authors should double check all the reported wavelengths in the paper. As an example, the correct triplet of nephelometer (Aurora 3000, Ecotech) wavelengths is 450nm, 525nm and 635nm and not 470nm, 525nm and 630nm as the authors stated in *page 4, line 75, and neither is it 440nm, 525nm and 635nm as stated in page 7, line 185-186. Still, Figure 2(a) and other figures and tables in the paper list the correct triplet of nephelometer wavelengths.*

We thank the reviewer for these comments. We have now explicitly used median values to present our own data and in the comparison of the Aarhus Bay data to other datasets and only used mean and standard deviations when no median values were provided by the other studies. Additionally, the 25% and 75% interquartile ranges were added to Table 1 to improve the statistical analysis.

We also went through the manuscript and checked all wavelengths to be correct.

Specific issues and comments

Page 3, line 73: Figure S1 shows no drying of the sampling line. However, the authors should state it explicitly in the main text.

The following sentence has been added:

The sampled air was dried due to the temperature difference between outdoors and inside where the instruments were located. No additional dryer was employed.

Page 3, line 76: did the authors apply the no cut correction or the sub-μm correction?

The "no-cut" truncation error correction was used. We have changed the text as follows:

The data were corrected for angular truncation errors using the "no-cut" correction according to Müller et al. (2011).

Page 4, line 80: the authors should include what autocalibration setting was used for the nephelometer measurements and how they treated nephelometer zero checks.

No autocalibration setting was used, as the zero and span gas measurements were performed manually once before and once after the measurement campaign. As the values coincided no additional calibration correction was applied. We have adapted the text as follows:

A full calibration, including zero measurement and span gas calibration with carbon dioxide (CO2) (99.9 % purity), was performed prior to and after the measurements. As the results coincided within the measurement uncertainty, no additional correction was applied.

Page 4, line 81: the aethalometer measures the attenuation of light shone through a filter tape which is progressively loaded with atmospheric aerosol. I suggest the authors make this explicit by, e.g., writing "filter light attenuation signal". Furthermore, how to pass from the filter attenuation signal to the absorption coefficient should be clarified.

The aethalometer measurement explanation has been changed to:

The aethalometer measured the attenuation of light through a filter tape progressively loaded with atmospheric aerosol particles, providing a "filter light attenuated signal" ….

To get absorption coefficients we used Eq.1, rather than calculating them from the attenuation. To make this more clear we have rephrased the sentence:

The absorption coefficients were calculated using Eq.1 by its relation to the equivalent black carbon (eBC) mass concentrations, where MAC is the mass absorption cross section (Hansen et al. 1984).

MAC values for the different wavelengths were used as suggested in the manual (e.g., MAC= 7.77 $m^2 g^{-1}$ for λ = 880 nm).

Page 5, line 102-103: what percentage of data was eliminated due to RH above 40%? Furthermore, the authors should include a comment as to why this data filter was applied as it is the only data filter which refers to environmental values and not inherently bad data due to tube disconnections and instrument self-calibrations.

0.78 % of the data were removed due to an RH above 40 %.

As described above no dryer was employed, instead the aerosols were directly dried by the temperature difference between outdoors and the location of where the instruments were located. To ensure that all reported data complied with the suggested RH < 40 %, we analysed the inlet RH and flagged the data points recorded at RH > 40 % which could be significantly affected by hygroscopic growth.

The following sentence has been revised accordingly:

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.

Page 5, line 107: the authors should include what hypotheses were used for the particle loss calculator (inertial impaction, gravitation deposition, diffusion, …). Moreover, maximum inlet efficiencies are usually found for particles with diameters of approximately 100-200nm and aerosol particles of interest for this paper (such as BC, BrC and secondary aerosols) can be much smaller than 1µm. I suggest the authors consider switching to a logarithmic scale on the x-axis of Figure S2 as this can help the reader better view the inlet efficiency plot at all diameters of interest.

Figure S2 in the SI has been updated with a logarithmic scale on the x-axes.

The following sentence has been added:

The calculation accounts for particle losses due to Brownian diffusion, gravitational settling, and inertial impaction in straight tubing and bends.

Section 2.4: I suggest writing a more general definition of AAE, SAE and ΔSAE (as was done for SSA, b and g in equations (2), (6) and (7)) and anticipate those definitions to the *beginning of the section, e.g. in line 121. Indeed, the authors throughout the whole paper use and show results found in the literature for AAE and SAE values calculated at different pairs of wavelengths to the ones of equations (3) and (4).*

We thank the reviewer for this helpful suggestion! The equations defining AAE, SAE, and ΔSAE have been changed to be more general, e.g., denoting wavelength as λ1, λ2 to be generally applicable.

Page 6, line 138: isn't the Ångström matrix obtained by subdividing the AAE-SAE biplot? I suggest the authors explain what the Ångström matrix is more thoroughly and add a reference herein to Figure S17 to better help the reader.

We thank the reviewer and have now clarified that the Ångström matrix is a bivariate plot of AAE and SAE subdivided into regions representative of different particle types and sources. A reference to the Angstrom matrix in the SI (earlier S17 now S3) has been added (and SI figures have been renumbered and references in the main paper have been updated). The revised text reads as follows:

Finally, the Ångström matrix describes the relationship between AAE and SAE values in a bivariate plot, effectively subdividing the AAE-SAE space into regions that are representative of different particle 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.

Page 6, line 155: while it is true that g=0 occurs for aerosol particles in the Rayleigh-scattering regime where scattering is symmetrical for the front and back hemisphere, there are cases in optics where the forward flux of cosine-weighted scattered radiation is equal to the backward one but with no symmetry between the forward and backward scattered light. As such, g=0 is more an indicator of balanced scattering between the forward and backward directions rather than symmetrical scattering.

We thank the reviewer for this comment and the text has been revised to clarify that g = 0 indicates an equal balance between forward and backward scattering rather than symmetric scattering. The new sentence reads as follows:

It ranges from -1 to 1, representing total backscattering and total forward scattering, respectively, whereas 0 represents an equal balance between forward and backward scattering.

Page 11, line 204-208: qualitative terms like "approximately" and "on average" should be replaced by more accurate scientific terminology, are the reported values means, modes or medians?

We agree with the reviewer that it was unclear. The study by Martinsson et al 2014 only presented monthly mean data, while the Alberg et al. 2023 study shared the whole dataset, thus allowing to calculate both median and mean values. In the revised manuscript we have now explicitly mentioned which values were reported both from our dataset and the data we compared to (i.e. mean or median). Additionally, we have replaced the words "approximately" where possible with more accurate terminology.

Page 12, line 254-256: Figure 4 shows median daily trends where traffic peaks are evident for weekdays and weekends only in the absorption signal. I suggest the authors refrain from using the words "similar trends" as weekday scattering is completely different to any other trend and weekend scattering peaks are not found at the same time as weekend absorption peaks. The difference between the absorption and scattering trends, both for weekdays and the weekend, suggests that non-absorbing and/or weakly-absorbing aerosols play a significant role in measured optical data.

We thank the reviewer for this suggestion. The phrase "similar trends" has been removed, and the text has been revised to more clearly distinguish between the diurnal behavior of absorption and scattering. The revised text now highlights that absorption coefficients show traffic-related peaks on both weekdays and weekends, whereas scattering coefficients exhibit different patterns. Also, a sentence has been added, that the difference between scattering and absorption patterns indicate a substantial contribution from non-absorbing or weakly absorbing aerosol components to the scattering signal. The revised text reads as follows:

The absorption coefficient σabs 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 was generally around 5-9 h, while the evening peak typically lasted 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 at different wavelengths were very similar between 10 and 18 h, indicating that both smaller and larger particles were present. During weekdays, however, σ_{sca} at 450 nm was clearly higher in the same time period, indicating a larger fraction of small particles. Overall, the differing diurnal behaviour in scattering and absorption indicate that non-absorbing or weakly absorbing aerosol components contributed substantially to the observed scattering signal, particularly during weekdays.

Figure 3 shows a shorter measurement period compared to Figure 2. Furthermore, Figure S3 shows daily patterns of 7 different weeks but only 5 full weeks are shown in Figure 2 and Figure 3. Could the authors comment on this? I suggest including the whole period of seven weeks for all figures. If data are not available for the whole seven weeks, FigureS17 should either show the trends of weeks where there is complete data coverage, or the caption should explicitly state which weeks have complete data coverage.

WELAS data was only available from 7 March (3 days after nephelometer and aethalometer), which explains the shorter time period in Figure 3. I have added this explanation to the figure caption. Figure S3 (now Fig. S4) shows diurnal variation for “calendar weeks”, i.e., Monday-Sunday. The measurement period spans 7 different calendar weeks. An explanation to the word “weeks” has been added in the caption for Fig. S3.

The new caption of Fig. 3 reads as follows:

The WELAS measurements were available from 7 March to 11 April 2023.

New figure caption of Fig. S3 (now S4):

Diurnal variation of (a) median value of σ_{sca} at 525 nm and (b) median values of σabs at 520 nm grouped by calendar week (Monday-Sunday) during the measurement period (3 March-11 April).

Page 11, line 216: the authors state that the aethalometer model was used to separate the fossil fuel component from the biomass burning component at the beginning of the text. Why do the authors mention compatibility of measured AAE values to fossil fuel AAE values if they expect to find a biomass burning component and intend to determine its contributing percentage it with the aethalometer model at a later stage?

These comparisons were made to better understand the AAE values found in the Ångström matrix throughout the campaign. As seen in the general Ångström matrix, AAE values ranged from approximately 0.5 to 2.5 thus spanning over a wide region. We aimed at comparing our AAE values to other coastal studies and find out if comparable values were observed.

Page 14, line 283-288: the authors should include a statistical parameter which quantifies the “very good agreement” between modelled and measured BC concentrations. To give more weight and importance to the FLEXPART analysis, the authors should also consider including comments on: (1) why the modelled results of Figure 5 show a smoothed-out trend compared to measured values; (2) why sharp BC peaks are never correctly modelled; (3) why the model underestimates BC concentrations and if this underestimation is found in any other literature.

We thank the reviewer for this comment and have included a more thorough statistical analysis and discussion regarding the comparison of the modelled and observed data.

(1) There are several studies in which FLEXPART modelled BC have been compared to measured BC, either under- or overestimating observations (Popovicheva et al. 2025, Fang et al. 2025, Yttri et al. 2024, Fang et al. 2023, Popovicheva et al. 2022, Platt et al. 2022). These studies were mostly performed in background sites and found good agreement between modelled and observed data. In urban sites such as in Aarhus, FLEXPART has not been used often, due to difficulties arising when local emissions have to be considered. Such emissions are sometimes located within the same grid cell with the receptor (in the present study, the receptor is Aarhus), and then transport cannot be properly resolved. The resulting superposition of transported BC and emitted BC makes

a direct comparison with FLEXPART data difficult. This is clearly the case in the present study with smoothed lines as compared to the observations. The same was observed in urban Vilnius (Lithuania) and Warsaw (Poland) in a paper that was published a few weeks ago. The problem of global models to resolve local emissions is further discussed there (Hey et al. 2026).

(2+3) An immediate result of the problem of global models like FLEXPART to resolve local emissions is underestimation of observations. Since local sources cannot be resolved by the model, it means that they are not properly transported to the receptor (Aarhus). Local pollution is mainly shown with sharp BC peaks in Fig.5, and the lack of these from the modelled BC indicates missing sources in the emission inventories used by the model. The larger the magnitude of the local pollution, the larger the underestimation by the model. As regards to Aarhus, which is a rather small city with limited local pollution, we only see a relatively small underestimation by the model, while the trend/tendency of the observed concentrations is clearly reproduced; this is the reason why we decided to use FLEXPART in the present study.

Other relevant studies where FLEXPART BC was underestimated are:
Hey et al 2026: https://doi.org/10.1016/j.atmosres.2025.108731
Arora et al 2025:https://doi.org/10.1016/j.atmosenv.2025.121367
Zhu et al 2020: https://doi.org/10.5194/acp-20-1641-2020
Evangeliou et al 2018: https://doi.org/10.5194/acp-18-963-2018
Wininger et al 2017: https://doi.org/10.1073/pnas.1613401114
Evangeliou et al 2016: https://doi.org/10.5194/acp-16-7587-2016
Eckhardt et al 2015: https://doi.org/10.5194/acp-15-9413-2015

The revised text reads as follows:

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. 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 %) while minor contributions from IND (~ 3 %), FLR (~2 %), SHP and WSD (~ 1 %) and ENE (~ 0 %) were found.

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. 2022, Platt et al. 2022), 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.

Pages 14-15, lines 290-304: could the authors also include the same statistical parameters of the previous point for the three showcase scenarios and comment on how the modelled results of the selected cases compare to the modelled results of the whole period? Is there a better or a worse agreement for Case1, Case2 and Case3 compared to the whole period?

We have included a statistical analysis for the three discussed cases and added the following parts to the manuscript:

Case 1:

FLEXPART footprint analysis for Case 1 revealed a clear continental contribution (see Fig. S7) and an age of 3 days for on average 76 % of BC data, where 91 % of BC originated from Europe (see Fig. S11), 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.

Case 2:

FLEXPART footprint analysis for Case 2 revealed a contribution from air masses arriving from the ocean (see Fig. S7), thus supporting an impact by marine particles. The trend of the modelled and observed black carbon is well comparable, with FLEXPART being able to capture the short BC peak (see Fig. 5), while the overall values were underestimated by the model (MFB = -78.7 %, RMSE = 128 ng m^{-3}).

Case 3:

Figure 8(d) demonstrates a strong agreement between black carbon concentrations simulated by FLEXPART and those measured by the aethalometer during Case 3. The model slightly overestimated the observations by 15.8 %, with RMSE of 132 ng m^{-3}. This good performance is consistent with the dominance of long-range transport during Case 3, a process that FLEXPART is well suited to represent.

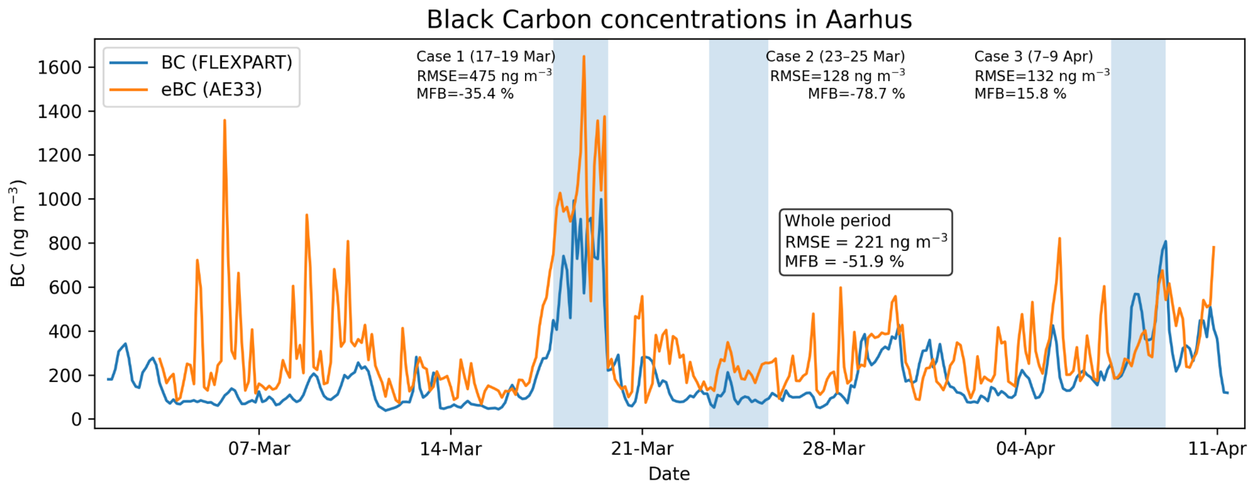

Figure S8 and Figure S10 show missing NOx data for a considerable portion of the Case 1 scenario (which is the local pollution case) and NOx is a tracer of local emissions as stated by the authors in page 15, line 310. Maybe this is a stupid question, but can the lack of NOx data influence modelled results substantially? If this is the case, please add a comment in the paper.

We thank the reviewer for this (not at all stupid) question. The FLEXPART analysis does not use the measured NOx data presented in Fig. S8 and S10.

Page 14, line 287: I would consider a percentage of 3% for the industrial emissions as being a minor contribution too.

We agree with the reviewer and have changed the contribution from IND to be “minor” instead of “main”.

Page 15, line 308-309: I would revise the phrase as follows “NOx and CO are often used as tracers of pollution as they are mainly emitted in fossil fuel combustion and biomass burning”. Any type of open-air combustion occurring at a high temperature can break the bonds of N_2 and O_2 (which are the main atmospheric gases) thus forming NO which subsequently can oxidise to NO_2. Moreover, nitrogen is essential for plant growth and is found abundantly in any plant, for these reasons NOx can be emitted in wood-burning processes too and should also be included as possible contributing sources on page 16, line 316. One should also note that BC and not only BrC is emitted in wood burning processes too.

We thank the reviewer for this comment.

Page 15, lines 308-309 have been revised to clarify that both NOx and CO are mainly emitted from fossil fuel combustion and biomass burning.

Page 16, line 316 has been changed to the following:

Elevated NOx and CO levels indicate dominance of local emissions, most likely from ships, local traffic, or other high-temperature combustion sources such as wood burning.

A note about BC also being emitted by biomass burning has been added in the methods section where we mention BrC is from biomass burning.

Page 17, line 344: averages are a more general definition than mean. I suggest that authors use more accurate statistical terminology.

We thank the reviewer for this comment and have changed all instances of "average" to the more precise mean or median term.

Page 19, line 392-394: This is the first occurrence where the authors discuss possible limitations of the aethalometer model. Indeed, the aethalometer model assumes that the absorption signal is due to only two absorbing aerosol ensembles, one with a high wavelength dependence and the other with a low wavelength dependence (AAE=2 and AAE=1 was the pair of Ångström exponents employed by the authors respectively for biomass burning and fossil-fuel combustion). Model limitations list the need to find site-specific AAE values to fit the data correctly, that particles with low AAE values can be emitted in wood burning processes too and that there are other absorbing aerosols other than BB and FF aerosol in the atmosphere such as mineral dust. For the above reasons, the aethalometer model offers only a coarse view of what is truly found in the atmosphere and its results should be taken with a pinch of salt. I suggest the authors explain the aethalometer model more thoroughly and discuss some of its limitations in the main text (e.g. when it is introduced in section 2.2 in page 4, line 88-89) as the aethalometer model is extensively used in the study.

We thank the reviewer for this suggestion. We have added more information on the aethalometer model and linked limitations in the methods section:

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.

Typographical and grammatical issues

The following is a list of revised phrases where typos and grammatical errors were revised.

We thank the reviewer for the revisions and all typographical and grammatical errors have been corrected throughout the manuscript.

- *Page 2, line 33: please revise the last phrase. An option could be the following "have contributed to decades-worth of in-situ aerosol optical …".*

- *Page 3, line 54: please revise “carries out” to “carried out”*
- *Page 3, line 57: please revise to “habitats which include wildlife such as herds of deer and…” possibly other wildlife.*
- *Page 4, line 86: “deployed” should be changed to “employed”.*
- *Page 6, line 154: please eliminate "the" and revise to “angular distribution of scattered radiation”.*
- *Page 7, line 178: please eliminate "the" and revise to “of fire emissions” and “of BB dispersion”.*
- *Page 7, line 178: “crucial for an accurate simulation of …” or “accurate simulations of …” or “crucial for the accurate simulation of …”.*
- *Page 7 line, 187-188: “Many sharp peaks were only visible in σ_{abs} and were absent in σ_{sca}, indicating the presence of strongly absorbing particles for short periods of time”.*
- *Page 14, line 273: “during weekends”.*
- *Page 14, line 277: “a lower PBL hinders dispersion thus favouring higher concentrations”.*
- *Page 18, line 358: I would revise to “thus supporting the hypothesis of marine-particles intrusion”.*
- *Page 18, line 362: the wording “this is also clearly visible” suggests that a different analysis was carried out and confirmed the FLEXPART results. I suggest the authors revise the phrase to “The FLEXPART footprint analysis for Case 3 is shown in Figure S7” as the authors are still talking about FLEXPART model results.*
- *Page 19, line 385-386: “reconstructed by the model” or “extracted from the model”.*
- *Page 20, line 408: “with the FLEXPART model was carried out”.*

<u>Reviewer 2:</u>

General Comments

The manuscript presents an ambitious and multifaceted characterization of aerosol optical and microphysical properties in a Scandinavian coastal environment, supported by dispersion modelling using FLEXPART. The combination of in-situ observations with source–receptor modelling is valuable, and the multi-instrument dataset collected over several weeks offers substantial scientific potential.

However, some methodological aspects require clarification—particularly the inlet configuration, humidity handling, nephelometer corrections, aethalometer assumptions, and instrument calibration procedures. Since these factors directly affect σ_{sca}, σ_{abs} and eBC, their explicit and consistent reporting in the main text is essential.

In addition, several optical parameters and time series are summarized using mean ± standard deviation, but the underlying distributions are likely skewed. Including median and percentile metrics directly in the main text would improve the scientific robustness of comparisons among the three case studies.

Finally, the selection criteria for the defined periods, the interpretation of optical exponents, and the model–measurement comparisons would benefit from clearer justification and more transparent presentation. With these refinements, the study would provide a more solid contribution to coastal aerosol research.

We thank the reviewer for this assessment and have improved the mentioned sections in the revised manuscript.

Specific Comments

Page 2, line 58–63: the inlet description does not clearly state whether the sampling line included drying or conditioning. Since RH strongly affects σ_{sca}, this should be explicitly clarified in the main text.

The following sentence has been added:

The sampled air was dried due to the temperature difference between outdoors and inside where the instruments were located. No additional dryer was employed.

Page 3, line 65: the Introduction refers to a field campaign lasting 6.5 weeks. However, the Methods section specifies measurement dates from March 3rd to April 11th, 2023, which corresponds to approximately 5.7 weeks (40 days). The authors should revise the reported study duration in the Introduction to ensure consistency with the dates provided in the Methods section.

We thank the reviewer for this comment. This was a mistake. The duration of the campaign was 5.7 weeks (40 days) as stated and has been changed in the introduction.

Page 3, line 72–74: please specify whether the nephelometer data were processed using the "no-cut" correction or the "sub-micron" correction. This choice significantly affects scattering coefficients and Ångström exponents.

The "no-cut" truncation error correction was used. We have changed the text as follows:

The data were corrected for angular truncation errors using the "no-cut" correction according to Müller et al. (2011).

Page 4, line 78–82: the autocalibration configuration for the Aurora 3000 should be described, along with the treatment of zero checks (frequency, duration, and how zero offsets were applied).

No autocalibration setting was used, as the zero and span gas measurements were performed manually once before and once after the measurement campaign. As the values coincided no additional calibration correction was applied. We have adapted the text as follows:

A full calibration, including zero measurement and span gas calibration with carbon dioxide (CO2) (99.9 % purity), was performed prior to and after the measurements. As the results coincided within the measurement uncertainty, no additional correction was applied.

Page 4, line 85–88: the wavelengths reported for the nephelometer appear inconsistent across the manuscript. Please verify and harmonize all wavelength triplets for scattering and absorption instruments.

We thank the reviewer for this comment, indeed some of the wavelengths were not correct. We have changed them throughout the manuscript.

Page 4, line 95–97: the WELAS Optical Particle Spectrometer (OPS) measures particle optical diameters in the range of approximately 0.2 µm to 10 µm. This lower limit 0.2 µm excludes the nucleation mode and a substantial fraction of the Aitken mode (ultrafine particles). Given the study's focus on combustion-related sources (traffic, domestic heating), which are major contributors to the ultrafine fraction, the authors must explicitly discuss the potential impact of this 0.2 µm cut-off on the derived total particle number concentrations and the source apportionment results, acknowledging the likely underestimation of ultrafine particle counts.

We thank the reviewer for this comment. Indeed the WELAS is not an adequate instrument to track nucleation and Aitken mode particles. However, the strength of the WELAS is the detection of larger particles up to several µm in diameter. The size distributions and number concentrations presented in the paper were predominately used to capture a possible sea spray aerosol contribution, typically described by larger particles. The discussion on particles from combustion-related sources, therefore mainly relies on the scattering and absorption coefficients together with the FLEXPART analysis. In order to make this more clear we have added the following statement to the methods section about the WELAS:

The presented total particle number concentrations are thus only representative for this size range, only partly capturing the nucleation and Aitken mode.

Page 5, line 115–120: clarify whether the sampling system and tubing efficiency calculations were used to correct measured concentrations, or only to estimate potential biases. Currently, the text does not explicitly state how inlet losses were applied.

The tubing efficiency calculations were carried out to estimate potential biases only. The following sentence has been added to the end of the inlet and sampling efficiency paragraph to clarify this:

The results were not used to correct the data.

Page 6, line 150–160: a more detailed explanation of the criteria used to define the three case periods would improve reproducibility. Thresholds for SSA, SAE, AAE or FLEXPART footprint characteristics should be reported.

The criteria are now listed more clearly in the manuscript under each case:

Case 1: The start time was determined to be when $\sigma_{sca,525}$ and $\sigma_{abs,520}$ were higher than their campaign means and SSA_{520} lower. The end time was set to be when $\sigma_{abs,520}$ decreased below the campaign mean.

Case 2: The start time was determined to be when both $\sigma_{sca,525}$ and SSA_{520} exceeded their campaign means and the end time when SSA_{520} decreased below its mean value for the whole period.

Case 3: 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.

Page 7, line 185–195: optical parameters such as AAE and SAE are interpreted as indicators of dominant particle size and source. Consider including a short discussion of the sensitivity of these quantities to mixing state and chemical composition.

We thank the reviewer for this suggestion. The relation of AAE and chemical composition is included in section 3.1 where the AAE and SAE values are compared to other data.

Page 8, line 210–220: the interpretation of high scattering periods should consider the role of sea-salt coarse particles. Because the inlet figures show size-dependent losses above ~6–8 µm, these limitations should be acknowledged in the discussion.

We thank the reviewer for this comment. As Case 2 presents the period with the highest scattering signals that are most likely linked to sea spray, we have included the following sentence to the Case 2 description:

However, the observed contributions by large coarse particles above approximately 8 µm may be underestimated due to the losses in the inlet and tubing.

Page 9, line 240–255: the comparison between measured eBC and FLEXPART BC would benefit from including additional statistical indicators (median difference, percentiles), since eBC distributions are typically log-normal.

We thank the reviewer for this comment. Including also the comment by reviewer 1 we have revised the comparison about modelled and observed BC data in the following way:

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. 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 %) while minor contributions from IND (~ 3 %), FLR (~2 %), SHP and WSD (~ 1 %) and ENE (~ 0 %) were found.

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 data 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. 2022, Platt et al. 2022), 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.

Page 10, line 265–275: if FLEXPART under- or overestimates BC for certain case studies, potential reasons (emission inventory uncertainties, domestic heating patterns, atmospheric mixing) should be explored more explicitly.

We thank the reviewer for this comment. This discussion has been implemented in the revised paragraph above.

There are several studies in which FLEXPART modelled BC have been compared to measured BC, either under- or overestimating observations (Popovicheva et al. 2025, Fang et al. 2025, Yttri et al. 2024, Fang et al. 2023, Popovicheva et al. 2022, Platt et al. 2022). These studies were mostly performed in background sites and found good agreement between modelled and observed data. In urban sites such as in Aarhus, FLEXPART has not been used often, due to difficulties arising when local emissions have to be considered. Such emissions are sometimes located within the same grid cell with the receptor (in the present study, the receptor is Aarhus), and then transport cannot be properly resolved. The resulting superposition of transported BC and emitted BC makes a direct comparison with FLEXPART data difficult. This is clearly the case in the present study with smoothed lines as compared to the observations. The same was observed in urban Vilnius (Lithuania) and Warsaw (Poland) in a paper that was published a few weeks ago. The problem of global models to resolve local emissions is further discussed there (Hey et al. 2026).

An immediate result of the problem of global models like FLEXPART to resolve local emissions is underestimation of observations. Since local sources cannot be resolved by the model, it means that they are not properly transported to the receptor (Aarhus). Local pollution is mainly shown with sharp BC peaks in Fig.5, and the lack of these from the modelled BC indicates missing sources in the emission inventories used by the model. The larger the magnitude of the local pollution, the larger the underestimation by the model. As regards to Aarhus, which is a rather small city with limited local pollution, we only see a relatively small underestimation by the model, while the trend/tendency of the observed concentrations is clearly reproduced; this is the reason why we decided to use FLEXPART in the present study.

Other relevant studies where FLEXPART BC was underestimated are:
Hey et al 2026: https://doi.org/10.1016/j.atmosres.2025.108731
Arora et al 2025:https://doi.org/10.1016/j.atmosenv.2025.121367
Zhu et al 2020: https://doi.org/10.5194/acp-20-1641-2020
Evangeliou et al 2018: https://doi.org/10.5194/acp-18-963-2018
Wininger et al 2017: https://doi.org/10.1073/pnas.1613401114
Evangeliou et al 2016: https://doi.org/10.5194/acp-16-7587-2016
Eckhardt et al 2015: https://doi.org/10.5194/acp-15-9413-2015

Technical Corrections

Page 1, line 20–22: minor grammar refinement recommended to improve flow of introductory paragraph.

The sentence has been split into two for better flow.

Page 2, line 55: replace "was carries out" with was carried out.

Done.

Page 3, line 90–92: ensure consistency in unit formatting (e.g., Mm^{-1}, $\mu g\ m^{-3}$, # cm^{-3}).

We have checked all units to have the right formatting.

Page 4, line 75: correct wavelength notation for nephelometer channels; ensure format is uniform throughout the text.

We have checked and corrected all wavelengths. For the nephelometer different wavelength pairs were used depending on which comparison was performed. For the Ångström matrix we used the wavelengths proposed by Cappa et al. 2016 but to compare to other sites we sometimes needed to recalculate the scattering coefficients at these other wavelengths.

Page 5, line 130–132: several sentences are overly long; consider splitting for readability.

Sentences about SAE and AAE have been split for better flow and readability.

Page 6, line 155: abbreviations for FLEXPART sectors (DOM, TRA, SHP, etc.) should be introduced at first occurrence in the main text, not only in figure captions.

The abbreviations are already introduced in the main text in section 2.5, which is before the first figure uses the abbreviations.

Page 7, line 182–185: ensure spacing between numbers and units is consistent (e.g., "450 nm" instead of "450nm").

We have checked the paper to have consistent spaces between numbers and units.

Page 8, line 210: check figure cross-references; some appear out of order (e.g., S15–S18 referenced before S12–S14).

Indeed the references to some of the supplementary figures were missing. We have corrected this now.

Page 9, line 235–240: equation formatting should be standardized to match journal style (subscripts, λ-notation, exponents).

The equation formatting has been standardized to use subscripts to denote wavelength dependence for derived quantities (such as SSA_λ) while keeping standard notation for wavelength-dependent optical coefficients (such as σ(λ)).

Page 10, line 280–285: typographical inconsistencies in references to case-study periods; ensure identical naming across text, figures and captions.

We have corrected this.