

Replies to the Referee comments on “Seasonal variation of the size distributions of black carbon in the Arctic”

By John Backman, Sho Ohata, Yutaka Kondo, Jonas Svensson, Eija Asmi, Hitoshi Matsui, and Tatsuhiro Mori

We would like to thank all three Referees for their insightful comments and constructive comments on the manuscript. The authors' agree that these comments have substantially helped to make the manuscript better and improved the scientific impact. We have addressed the Referees' comments and suggestions to the greatest extent possible, while balancing the author's view on the focus on the manuscript, without diluting the main message. This delicate balance should make for a concise and impactful paper and provide a good starting point for future research on the topic of Arctic BC containing aerosols.

Major changes to the manuscript:

- Section 3.2 split into subsections of 3.2.1 Seasonal variation of the BC core size distribution, 3.2.2 Impact of in-cloud periods on the BC core size distribution, and 3.2.3 Seasonal variation of BC mixing state.
- New figure 8, showing: the mixing state of BC aerosols at Pallas, the amount of BC containing aerosol particles, shell/core relationships for the coating thickness of the BC aerosols.
- The BC analysis during cloudy conditions was significantly revised.
- The fraction of biomass burning BC to total BC was added to Figure 10 (was Figure 9) along with discussion about how this fraction will influence how we interpret the results.
- Section 3.4 revised with discussion added on the implications of modelled fraction of biomass burning (BB) and fossil fuel (FF) emissions which have different size distributions in the model.

The structure of this document is:

Referees' comments are underscored.

“Text added to the manuscript in quotes and dark red”

Replies to the Referees by the authors are in upright text.

Comments by Anonymous Referee #1, 12 May 2026

Comment: The most significant aspect missing from this manuscript is a detailed characterization of the rBC mixing state. The previous study at Pallas using SP2 shows that rBC particles in the Arctic are thickly coated ($D_p \text{ Shell}/D_p \text{ BC}=2$). Since the data from this work cover a full year, the authors are in a unique position to analyze the seasonal characteristics of the mixing state, including the particle diameter ratio (shell/core), coating thickness, and the number fraction of BC among total particles.

Reply: This was an important omission as pointed out by the Referee. At the time of the submission, we did not have the optical size distribution available from the SP2 and our first idea was to focus on the core size distribution changes. However, we do agree with the Referee that the mixing state is very essential for interpretation of data as well as being a very valuable parameter essential for future studies. Therefore, for the revised manuscript, we added analysis of the scattering signal and did the LEO-fittings to define single BC particle coating thicknesses. As a result of this new addition, we have updated the methods section, and added a new Figure 8 where interannual variation of the number fraction of BC containing particles and the shell-to-core diameter ratio are depicted. These new results show a very clear interannual variability in the fraction of the rBC containing particles, being elevated during the cold season. The shell-to-core diameter ratio varied in range 2.1 to 2.3, being the lowest in mid-winter and highest in mid-summer. These results were added in an additional subsection 3.2.3 after the discussion of the temporal variability of BC core size distributions:

“BC mixing state is a key property of the atmospheric BC affecting its aging processing and essentially, the climatic impacts. Atmospheric ageing of BC is the process by which fresh fractal-like BC particles gain a coating. That coating can change hydrophobic fresh soot into hydrophilic particles that can act as CCN, hence collapsing the fractal like structure and gain secondary aerosol mass through aqueous phase chemistry.

Figure 8a shows the number fraction of BC containing particles (F_{BC}). F_{BC} is the number of BC containing particles divided by the total number of particles as detected by the SP2. The average F_{BC} was 15.4%. As was pointed out by Matsui et al. (2018), a single mixing state is not a good approximation for BC in the Arctic as so few particles contain BC. F_{BC} show a clear seasonality, with the lowest values in summer, and highest in winter. The highest F_{BC} was observed in February (29.0%) and the lowest in June (3.2%). Furthermore,

the SP2 is not able to detect particles with an optical diameter below 175 nm so the F_{BC} reported here is to be considered an upper limit as the nucleation and Aitken modes are not detected by the instrument. Statistics of F_{BC} for different months are shown in Table 1.

Figure 8b shows the shell-to-core diameter ratio of particles in the optical size range of 175 – 225 nm. The detection limit of the scattering channel was 175 nm. The mean shell-to-core diameter ratio in the measured size range was 2.2 which is close to what Raatikainen et al. (2015) reported for January in 2015 (for the optical size range of 150 – 200 nm)."

We also added text on the coating thickness method in the Methods section 2.2.1 in the manuscript:

"The SP2 was calibrated using fullerene soot (Gysel et al., 2011; Laborde et al., 2012) for the incandescence channels, and polystyrene latex spheres (PSL) with a known size and refractive index for the scattering channels. The optical size of the BC containing particles was obtained using derived from the leading edge of the scattering signal (Gao et al., 2007). As in Raatikainen et al. (2015), for the ambient particles we assume that they are spherical and have the refractive index of ammonium sulphate. The method assumes compact BC particles with well-defined spherical shell-to-core structure." ...

The discussion section was also modified to reflect these new results and discuss their implications. Our interpretation is that the variability of the mixing state in BC mainly reflects the variability of aerosol sources, and the dominance of biogenic emissions in summertime whereas in winter the significance of anthropogenic sources increases. Further implications on aerosol radiative forcing or any conclusions regarding BC aging in the atmosphere would require significant extension of the modeling and data analysis, which we feel would be out of the scope of this article.

Comment: Variations in MMD could be overlooked by SP2 volume-equivalent diameter (VED) limitations. The study relies on SP2 measurements, which assume a core-shell spherical model. However, as demonstrated by Wu et al. (2023), this assumption leads to a significant underestimation of the actual diameter for fractal black carbon aggregates, especially for loose structures (underestimation up to 45%). If the fractal dimension (Df) of BC varies seasonally, the SP2's size inversion algorithm would misinterpret this as a size change.

Reply: This is an important point and the work by Wu et al. have clearly demonstrated how a loose fractal structure can be misinterpreted as smaller particles. However, fractal BC aggregates, which are found in fresh emissions, collapse to spherical BC cores particles during atmospheric processing and cloud activation before reaching the

Arctic (Fierce et al. 2025, Zanatta et al. 2025). There are no major sources of local BC close to Pallas, and the greater majority of BC at the site is indeed aged BC. As discussed already in Raatikainen et al. 2015, and confirmed here (see Fig. 9), no significant amount of bare soot particles was detected at the site. Therefore, we consider the influence of seasonal variations in BC morphology on the retrieved BC-core size distributions to be limited, although it cannot be completely excluded.

In the revised manuscript, we acknowledge the fact that we assume compact soot particles that resemble the calibration material used. Wu et al. (2023) show how loose soot structures behave in the SP2 and how the instrument response changes with the level of compactness.

In section 2.2.1 we added the text: "[Recent work by Wu et al. \(2023\) demonstrated that highly fractal BC aggregates may lead to biases in SP2 size retrievals using Mie method. However, BC observed at Pallas predominantly represents long-range transported and atmospherically processed aerosol, which is expected to consist largely of compact or collapsed BC structures \(e.g. Raatikainen et al., 2015\), for which we estimate this source of error in sizing is not significant in our study.](#)"

Comment: The interpretation of MMD requires a more thorough physical explanation. The VED of the BC core is an intrinsic property that remains constant regardless of atmospheric aging or coating. Therefore, changes in MMD are not related to aging. Instead, they are influenced by variations in the characteristics of the emission sources. For example, this can include shifts in contributions from different combustion sources or differences in transport efficiency.

Reply: This is a good comment that requires some clarifications from our side. We have not meant to claim that MMD would be directly related to ageing and acknowledge, throughout the manuscript, that we discuss the BC core size.

In the revised manuscript, we have explicitly clarified that "[The BC core mass size distribution that is measured here is mainly a product of the characteristic BC sources and can only be modified during transport by coagulation between BC-containing particles and by size-dependent removal.](#)" This was added in the beginning of section 3.2.1. In addition, we have carefully checked the manuscript text discussing the BC size clarified where appropriate (changes marked in the new version of the manuscript) that we particularly discuss on the core size. In section 3.3 we additionally added that: "[Furthermore, it should be noted that these are core BC size distributions while cloud processing is dictated by the overall size and chemistry of the BC aerosol, where BC coating and mixing play a key role.](#)"

And in section 3.4 we now discuss how biomass burning and fossil fuel emissions in the model can change the comparison between measured and modelled BC core size distributions, and we explicitly mention what processes can alter the BC core size distributions in the model. Text added to section 3.4:

“The BC core size distribution is largely determined by the BC sources. BC emitted by biomass burning (BB) and fossil fuel (FF) sources have been shown to result in different size distribution, while BB BC emissions are larger in size than FF BC emissions (Ko et al., 2020), which is also the case for emissions in the model; i.e. BB BC are larger than FF BC. BC size distributions are altered during transport by coagulation between BC-containing particles and by size-dependent removal processes. The BC core size distribution at the receptor site therefore reflects both source-dependent emission size distributions and subsequent atmospheric processing, particularly coagulation and size-dependent removal. Matsui et al. (2018) demonstrated that the emitted BC size distribution strongly affect the BC lifetime and that mixing state affects radiative effects greatly.

The CAM5-ATRAS model takes these processes into account and therefore, the modelled MmD for Pallas is a combination of a mix of emissions and aerosol processing during transport.

In Fig. 910, the modelled mass fraction of BB (F_{BB}) is shown. F_{BB} is the fraction of BB BC mass (M_{BB}) to the sum of M_{BB} and FF BC mass (M_{FF}). In the absence of aerosol processes in the model, the modelled MmD would simply reflect the relative amount of BB BC between the MmD of pure BB and FF. As shown in Fig. 910, F_{BB} is almost zero (below 0.002) during winter months (December, January, February) and the modelled MmD is also low (below 170.7 nm). F_{BB} is high in July and August, with a maximum F_{BB} value of 0.178 in July. During summer months, the increase in the modelled MmD in Pallas is likely an indication of the increasing contribution from BB BC aerosols reaching Pallas. However, our analysis can not separate the individual contributions from different processes affecting the modelled BC core size.

Because the model simulations and measurements cover different periods, the comparison can only assess climatological variability and systematic bias rather than agreement for any specific month or year. Previous short-term campaigns support the hypothesis that BC core size is broadly stable in the Arctic surface layer (e.g. Raatikainen et al., 2015; Jurányi et al., 2023). Nevertheless, meteorological conditions and the location and strength of emission sources, particularly biomass-burning sources, may have differed between the 2009–2015 simulation period and the 2020 measurement period, thereby affecting their comparability. The magnitude of this effect on the model–measurement comparison of MmD in Fig. 10 cannot be quantified here. According to work by Hänninen et

al. (2026), October 2020 was influenced the most by BB aerosols, which is not seen in the measured MMD or MmD in Fig. 10, and only small amounts of BB aerosols reached Pallas in January 2020. The persistence of the model's low bias throughout the year suggests that it is unlikely to be explained solely by interannual or event-scale variability, which would further warrant research on this topic."

Comment: The manuscript averages the SP2 data to a 12-hour resolution for analyzing precipitation events. However, this approach may not adequately capture the rapid physical processes involved in wet deposition. Averaging over 12 hours can blur the sharp concentration gradients that occur before, during, and immediately after a scavenging event. As a result, this may lead to an underestimation of scavenging efficiency or even a complete failure to detect the event's impact.

Reply: This is an insightful comment, highlighting this process and the and the accompanying complexities. We can agree that our original conclusion was too strong given the limitations of our methodology. To complement our standpoint, we have done additional analysis but have also substantially changed our conclusions on this point to match the conclusions that can be taken with this analysis and data.

To address the specific questions of the Referee, we want to emphasize the unique position of the station Pallas. Pallas is frequently experiencing in-cloud episodes, while these clouds are mainly low-level polar stratus that could at times be influenced by the orographic features of the hill (e.g. Doulgeris et al., 2023). The LWC and cloud droplet numbers are strongly dependent on the air mass properties, roughly divided between Arctic and continental air masses features. Further information and clouds microphysical classification are described in Doulgeris et al., 2023. These additional explanations are now added to Section 3.2 to provide the reader with a background of the Pallas clouds and related studies. However, a deeper analysis of the clouds in Pallas remains to be done. This should take advantage also of the recently installed cloud radar and other available instruments. The station is equipped with both whole air and interstitial inlets and the SP2 was attached in the whole air inlet collecting both interstitial and cloud-activated aerosol. Also, this is now explicitly mentioned in the modified manuscript text.

As correctly highlighted by the Referee, the analysis presented in the manuscript is solely based on average rBC size distributions classified using visibility as a proxy for in-cloud conditions. While this provides a first-order comparison, it does not directly resolve the evolution of rBC during individual cloud-processing events. To evaluate this possibility, we conducted an additional exploratory event-based analysis where we identified 75 extended in-cloud periods during the study period (criteria: visibility <1000 m for >12 h), of which 45 contained sufficient SP2 data for further analysis. Then, we

compared mean rBC size distributions before, during and after the in-cloud periods. The results showed substantial event-to-event variability with no systematic change in rBC mass median diameter across the event ensemble, although individual events showed both increases and decreases in rBC mass and size. In summary, these 45 events resulted in:

	Increase [%]	Decrease [%]	Remains the same [%]
BC mass	33.3	60.0	6.7
BC MMD	17.8	31.1	51.1

Therefore, in >50% of the cases the MMD did not clearly change (applying 5% tolerance) and most often the BC mass decreased, while also in some cases increase in rBC mass was observed. The absence of a clear difference in the average size distributions should therefore clearly not be interpreted as evidence that cloud processing is unimportant. Rather, any cloud-processing signal may be obscured by several factors also indicated by the Referee, including changes in air mass origin during prolonged cloud episodes, entrainment processes, variability in cloud microphysical properties, and processing timescales that may be shorter than the averaging and classification approach applied here. Furthermore, cloud processing is expected to affect BC mixing state and coating thickness more directly than the refractory BC core size itself. Analyses of BC coatings and cloud microphysical observations are currently ongoing and will help address these questions in future work. In this manuscript, we have modified the text in several parts to better reflect the associated uncertainties, while also highlighting the potential of this dataset and its further value. We hope that this helps to answer the Referee's concern and avoid overinterpreting the observed differences between in-cloud and out-of-cloud mean distributions. The modified text on Fig. 7 in section 3.3 is now:

"Figure 7 shows the mean core BC size distributions for the three different conditions. It suggests that the ~~The mean~~ BC mass size distribution shape is not largely impacted by the presence of clouds nor rain, and while the mode remains around 200 nm. ~~suggesting that clouds have little to no impact on the shape of the core BC size distribution. However, a more detailed analysis of individual in-cloud episodes revealed substantial event-to-event variability: while BC mass decreased during most long-duration events (>12 h in-cloud) and the MMD of the size distribution remained unchanged in more than half of the events, several events also showed contrasting changes in both BC mass and the shape of the size distribution. It is evident that changes in air mass origin during prolonged in-cloud episodes, entrainment processes, variability in cloud microphysical properties or in processing timescales may all explain this observed event-to-event~~

variability. Furthermore, it should be noted that these are core BC size distributions while cloud processing is dictated by the overall size and chemistry of the BC containing particle aerosol, where BC coating and mixing play a key role. Combining this dataset with co-located measurements of aerosol chemistry, cloud microphysics, and air-mass history could help understand the roles of cloud processing, selective scavenging, and changing source regions in polar mixed-phase clouds. The present analysis supports a more limited conclusion: the annual mean BC core size distributions observed during in-cloud, rainy, and high-visibility conditions at Pallas do not differ substantially. These condition-specific BC distributions provide observational constraints for models, rather than direct evidence that the underlying cloud-processing pathways are the same."

Comment: The shipping traffic is quite dense along the Scandinavian coastline (see the attached pdf file, a snap from <https://globalmaritimetraffic.org/gmtds.html>). Does the seasonality of shipping traffic impact the variability of the observational results?

Reply: This is a very relevant and topical question. The attached map suggested by the Referee shows AIS tracks from all AIS equipped vessels. When limiting the selection to larger cargo ships and passenger ships, the activity is significantly reduced. Based on the online map, however, it is impossible to say how much these ship emissions would contribute to what we see at the Pallas (about 300km from the coastal line) measurement site. Such information would require extensive modelling of ship emissions other than just AIS data, including a database on engine emissions from differently sized ships, speed (engine load), fuel type, emission scrubbers etc in addition to chemical transport model. At FMI, we have very advanced modeling resources with both the The Ship Traffic Emission Assessment Model (STEAM) and our chemical transport model SILAM which would allow such more detailed characterization of the impacts of the Arctic shipping to aerosols at Pallas and this is certainly something that should be addressed in future.

However, unfortunately we feel again that this would be well beyond the scope of this manuscript. The work by Geels et al. (2021), which included the STEAM model, estimated that "Shipping contributes to 1 %-15 % of the $PM_{2.5}$ levels in the Nordic region, with the highest contribution along the shipping routes and in coastal areas". Pallas is not a coastal site so the influence should be at the lower end of the estimate, but without further analysis this is impossible to estimate.

While this manuscript is not focusing on resolving the sources of the observed BC, we have not added this discussion there. It would deserve a specific article with dedicated modeling resources to understand how different sources contribute to the observed BC abundance and size in Pallas.

Comments by Anonymous Referee #2, 15 May 2026

Comment: Model-observation temporal mismatch and interpretation

The CAM5-ATRAS simulations cover 2009-2015, while the measurements are from November 2019 to December 2020. This ~5-year gap is acknowledged but the implications are not adequately discussed. Arctic BC concentrations and source regions have been changing (e.g., declining European emissions, increasing Siberian wildfire activity).

How might interannual variability in source contributions (especially from wildfires, which were pronounced in 2019-2020) affect the comparison? Could the 20% low bias in modelled MmD partly reflect different emission source mixtures rather than model structural issues?

Reply: This is a very good point and was also raised by the first Referee. The revised manuscript now discusses this in length. Based on this insightful comment, we now acknowledge that the source regions might be different during the period modeled and when the measurements were conducted, as was kindly pointed out. In the revised manuscript, we acknowledge the fact that the measurements and the modelled time period do not overlap and that we compare the results in terms of variability and bias and caution that the measurement year 2020 might differ from the multi-year behavior of the modelled year 2009 – 2015. The changes made to the manuscript in section 3.4 is shown in full above in the reply to anonymous Referee #1.

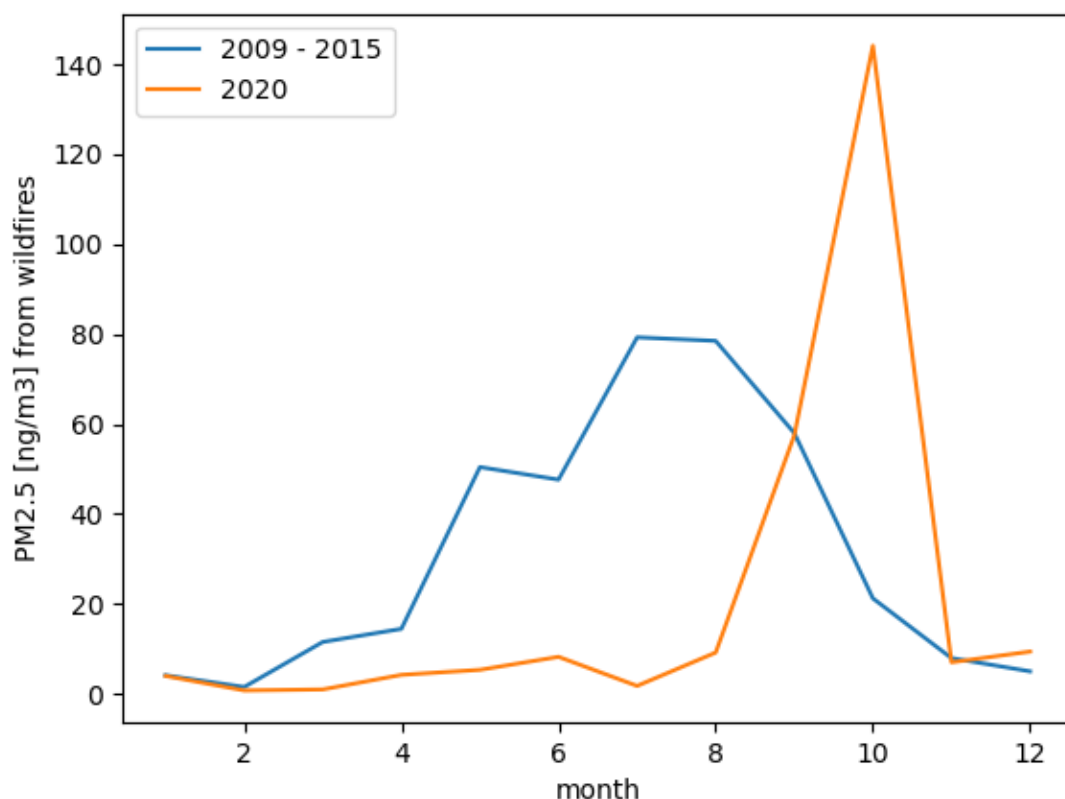
Comment: Perhaps adding a sensitivity analysis using back-trajectories for the measurement period to characterize dominant source regions, then compare with the model's climatological source representation.

Reply: This is yet another great suggestion from the Referee and we understand that our unique dataset enables different type of event analysis and process studies that will certainly be the focus of our future work. However, the goal of this paper is not to go into detailed event analyses but provide an overview of the general behavior of the Arctic BC core sizes, for the first time for the whole year. Based on another Referee's suggestion, we have now additionally extended our analysis to BC mixing state variability. We feel that further extension of the scope would dilute the main message of this paper too much. In the revised manuscript we have added discussion on different mixtures of sources and updated Fig 9 (new Fig 10) with the BB fraction of total BC mass as a general overview. Wildfires (BB aerosols) most frequently arrive at Pallas during summer and this is directly reflected in the modeled BC core diameter

showing a clear increase (Fig. 10). This information is important for understanding the MmD variability in the model since BB and FF emissions have a fixed BC aerosol size distribution in the model and are only changed due to wet removal or coagulation. From our updated Fig. 10, it is evident that BB BC during summer increases the MmD as one would expect based on the emissions in the model.

Based on our analysis for the Referee's comments, in 2020, there was a low amount of BB aerosols reaching Pallas, compared to the years 2009 – 2015. We cannot run the CAM5-ATRAS model again, for the measurement period. But in the revised manuscript, we acknowledge the possibility that source regions are different in 2020 than during 2009 – 2015 in general.

We have tried to estimate the difference of BB aerosols for Pallas for 2009 – 2015 and 2020 using the chemical transport model SILAM. That model data is available at <https://etsin.fairdata.fi/dataset/bde9f0f9-ec30-474e-9e75-657366b15e77>



The figure above shows modelled PM_{2.5} levels from biomass burning (BB) using the SILAM chemical transport model for Pallas. The figure shows the average BB for each

month during the time period that we have data from the CAM5-ATRAS model (2009 - 2015) and for the year 2020 (from where we have measurement data but not CAM5-ATRAS model results).

We did not want to add yet another model, with all the description about their differences (emissions, chemistry, deposition, advection, transformation etc.) to the revised manuscript, but use the data here to show how the years differ, which is now also discussed in the revised manuscript.

Text added to section 3.4:

"The BC core size distribution is largely determined by the BC sources. BC emitted by biomass burning (BB) and fossil fuel (FF) sources have been shown to result in different size distribution, while BB BC emissions are larger in size than FF BC emissions (Ko et al., 2020), which is also the case for emissions in the model; i.e. BB BC are larger than FF BC. BC size distributions are altered during transport by coagulation between BC-containing particles and by size-dependent removal processes. The BC core size distribution at the receptor site therefore reflects both source-dependent emission size distributions and subsequent atmospheric processing, particularly coagulation and size-dependent removal. Matsui et al. (2018) demonstrated that the emitted BC size distribution strongly affect the BC lifetime and that mixing state affects radiative effects greatly.

The CAM5-ATRAS model takes these processes into account and therefore, the modelled MmD for Pallas is a combination of a mix of emissions and aerosol processing during transport.

In Fig. 910, the modelled mass fraction of BB (F_{BB}) is shown. F_{BB} is the fraction of BB BC mass (M_{BB}) to the sum of M_{BB} and FF BC mass (M_{FF}). In the absence of aerosol processes in the model, the modelled MmD would simply reflect the relative amount of BB BC between the MmD of pure BB and FF. As shown in Fig. 910, F_{BB} is almost zero (below 0.002) during winter months (December, January, February) and the modelled MmD is also low (below 170.7 nm). F_{BB} is high in July and August, with a maximum F_{BB} value of 0.178 in July. During summer months, the increase in the modelled MmD in Pallas is likely an indication of the increasing contribution from BB BC aerosols reaching Pallas. However, our analysis can not separate the individual contributions from different processes affecting the modelled BC core size.

Because the model simulations and measurements cover different periods, the comparison can only assess climatological variability and systematic bias rather than agreement for any specific month or year. Previous short-term campaigns support the

hypothesis that BC core size is broadly stable in the Arctic surface layer (e.g. Raatikainen et al., 2015; Jurányi et al., 2023). Nevertheless, meteorological conditions and the location and strength of emission sources, particularly biomass-burning sources, may have differed between the 2009–2015 simulation period and the 2020 measurement period, thereby affecting their comparability. The magnitude of this effect on the model–measurement comparison of MmD in Fig. 10 cannot be quantified here. According to the work by Hänninen et al. (2026), October 2020 was influenced the most by BB aerosols, which is not seen in the measured MMD or MmD in Fig. 10, nor in Figs 5 – 6; only small amounts of BB aerosols reached Pallas in January 2020. The persistence of the model’s low bias throughout the year suggests that it is unlikely to be explained solely by interannual or event-scale variability, which would further warrant research on this topic.”

Comment: January outlier and statistical treatment

January is identified as an outlier with MMD = 235 nm (Table 1), and is excluded from some analyses. However, the justification ("measurement and modelled periods do not overlap") seems insufficient. If January represents real Arctic conditions (possibly from different transport patterns), excluding it may bias the characterization of BC size variability.

What meteorological/transport conditions prevailed in January 2020 that produced larger BC cores? Was there enhanced long-range transport from specific source regions (e.g., Siberian industrial areas)? Could the larger January MMD reflect measurement artifacts (e.g., unusual coating characteristics affecting SP2 detection)?

A detailed analysis of the January period (air mass history, BC concentration, other aerosol species if available) would be helpful before deciding to exclude it.

Reply: Yes, this is an interesting question. There are no indications in the instruments housekeeping data that would suggest that the instrument was not working in January. All periods that suggest that it was not working within its parameters have already been removed from the data. Sufficient laser power is the most important parameter to follow to ensure that BC particles are detected after the shell has evaporated.

In the revised manuscript, we have not excluded January as an outlier but rather acknowledge that it stands out from the rest of the months. We reason that, as reasoned by the Referee, that since the time periods do not overlap, it is likely that the source areas and meteorological conditions are different during the measurement period and the modelled period which could result in larger BC cores reaching the site than what was modelled for 2009 – 2015 on average. Furthermore, in the revised manuscript we show that biomass burning BC is most common during summer as

modelled by CAM5-ATRAS (years modelled 2009 – 2015) and is contributing to the larger MmD in during summer months when BB BC aerosols are modelled to reach the site. We have added discussion on the possible differences between the measured and modelled years in terms of meteorological conditions (in general) and the location and source strength of namely BB emissions that change from day to day in the model. Section 3.4 and Fig. 10 were updated to discuss these possibilities in more detail, as stated in the replies above, and hence not repeated here.

One event at the end of January could be from BB aerosols in the Balkans reaching Pallas with elevated concentrations of BC. Another event in the middle of January, without elevated BC concentrations also showed somewhat larger BC cores but we cannot determine the origin of these BC containing aerosols since we don't know the chemistry of the particles, but we have added discussion about different source areas.

Comment: Cloud processing analysis limitations

The conclusion that clouds have "little to no impact" on BC size distribution (Section 3.2, Fig. 7) is potentially too strong given the methodology. The classification uses visibility as a proxy for in-cloud conditions, which is appropriate but coarse. More importantly, you compare mean distributions rather than examining whether BC size changes during cloud events.

What is the typical cloud liquid water content and droplet number concentration during these in-cloud periods? Have you examined whether BC size distributions differ between the beginning and end of extended in-cloud periods (≥ 6 hours)? A time-lagged analysis might reveal processing effects. How does the cloud-base altitude at Pallas compare to the station elevation of 565 m? Are you sampling cloud interstitial aerosol, droplet residuals, or both?

A more nuanced discussion acknowledging that cloud processing effects might be masked by: (a) continuous entrainment of fresh air masses, (b) rapid processing timescales relative to your 12-h averaging, or (c) the fact that BC cores themselves do not grow (only coatings do) is needed.

Reply: The Referee is correct that the classification is coarse given the methodology we present. In the revised manuscript, we address that the scavenging of BC aerosols during rain and in-cloud events is more intricate than what we present here and state the limitations raised by both Referee #1 and #2. See our reply and additions to the revised manuscript by Referee #1 where we address the same points as was raised here, and hence not repeated.

Comment: Spatial comparison across sites

The conclusion that BC size distributions are "very similar" across Pallas, Alert, and Ny-Ålesund is a key finding. However, the reported MMDs differ by ~15-20% (Pallas annual: 194 nm; Alert winter/spring: 216 nm; Ny-Ålesund winter/spring: 228 nm).

Have you tested whether these differences are statistically significant given the different measurement periods and sample sizes? The difference between Pallas (194 nm) and Ny-Ålesund (228 nm) is ~17%, which seems substantial for constraining models.

Statistical significance tests (e.g., bootstrapped confidence intervals for MMD differences) should be added and possible reasons for remaining differences (e.g., proximity to open ocean at Ny-Ålesund, different transport pathways to Alert vs. Pallas) could be discussed.

Reply: This is an important point and is now addressed in the revised manuscript. Bootstrapping the data points that we have from Ny Ålesund (MMD) we have calculated the confidence interval of the mean MMD on the 1 hour data. These statistics are also presented in the revised manuscript, along with discussion about the statistical significance between the two sites. It would not have been possible to bootstrap the statistical significance with the data that we have from Alert as we simply have too few data points (1), just the campaign average.

Added text to section "3.3 BC size distribution spatial variability in the Arctic":

"Due to the different sample sizes, the bootstrap method was used to estimate the confidence interval for the mean MMD for Ny-Ålesund. The Ny-Ålesund data, comprising of 227 h, the 95% confidence interval for the mean MMD for Ny-Ålesund is estimated to be between 224.9 and 231.9 nm. The resampling and sample size was 10^4 . The same bootstrap was performed on the Pallas data, with 1 h time resolution and resulted in a 95% confidence interval of 197.0 and 198.5 nm for the mean MMD. The difference is statistically significant. For Alert, we only have the mean distribution of the whole campaign, so bootstrapping is not possible.

The size distributions at Ny-Ålesund, Alert, and Pallas were measured during different years, but the MMD is within 18% of each other; i.e. the mean MMD of Ny-Ålesund is 18% greater than at Pallas. In the Arctic, M_{BC} typically varies by an order of magnitude, or more; e.g. Fig. 2 and Law and Stohl (2007), but the MMD and σ_m remains comparatively stable."

Comment: Missing mass correction details

The bi-modal log-normal fitting to correct for BC mass above SP2's 537 nm detection limit is important but described briefly.

Why is a bi-modal fit preferred over a single log-normal mode? Is the second mode at 475 nm physically meaningful or an artifact of fitting to the truncated distribution? What fraction of total M_{BC} comes from particles >537 nm? This should be reported. How sensitive is the corrected M_{BC} to the choice of fitting range and initial parameters?

Reply: The Referee is correct here and we hope to better justify our choices. The bi-modal fit was selected because it provided a better empirical representation of the measured mass-size distribution near the upper end of the SP2 detection range than a single log-normal fit. Similar high-diameter shoulders or rBC mass contributions have been reported in ambient BC size distributions, although they are commonly represented using a single log-normal extrapolation rather than explicitly fitted as a second mode (e.g. Raatikainen et al. 2015, Zanatta et al. 2023, Pileci et al. 2021, Ohata et al. 2021). We therefore do not interpret the fitted mode at 475 nm as evidence of a distinct physical particle population. Instead, the second log-normal component is used as an empirical representation of the observed shoulder and to estimate the small fraction of rBC mass beyond the upper detection limit. The average total mass calculated from the bi-modal distribution was 15.67 ng m^{-3} which is 1.9% higher compared to the average mass based on unimodal fit (15.37 ng m^{-3}). Our method, interpretation and the associated uncertainty are now stated explicitly in the revised manuscript.

Added text to section 3.1:

"The SP2 upper detection range is limited to 537 nm, as shown in Fig. 4; therefore, the entire BC size distribution is not captured by the SP2 which often shows a shoulder at the upper end of the detection range (Ohata et al. 2021, Pileci et al. 2021, Raatikainen et al. 2015, Zanatta et al. 2023). This shoulder can comprise up to 6.5% of the mass if using one log-normal fit to the BC mass distributions for urban and rural sites (Pileci et al. 2021)"

We further acknowledge that

"The bigger mode is an empirical representation of the observed shoulder on the mass size distribution which is not correctly captured by a single log-normal fit."

And later we give the statistics compared to the classic unimodal fit:

"The total mass of the bi-modal size distribution is 15.67 ng m^{-3} and 6.4% of that mass is from BC that has a mass equivalent diameter greater than 537 nm. The total mass of the bi-modal fit is 1.9% greater than the total mass of a unimodal fit to the data."

Minor:

Comment: Line 150 (page 6): The wavelength range is given as 350-800 nm. Specify the center wavelength of the broadband channel (typically ~650 nm for SP2).

Reply: The following text was added to the manuscript: "The peak sensitivity of the broadband channel is at 650 nm (Laborde et al. 2012, Lamb 2019)."

Comment: Equation (1): There appears to be a typo in the exponent denominator.

Reply: In the numerator, yes. The typo was corrected.

Comment: Table 1: The column headers have a formatting issue – " σ_m " appears twice (should be σ_m and σ_n). Also, the bottom rows show both "MMD model" and "MMD meas" but the main MMD column already shows measured MMD. Please clarify.

Reply: The column header was checked and we could not identify these problems. Perhaps there was some formatting issue with the PDF on the Referee's version which we don't see.

Comment: Figure captions: Figure 8 caption refers to " $R_{\text{eff}}(D)$ " but the figure shows $R_m(D)$ and $R_n(D)$ as in the main text.

Reply: The caption reads the same as the figure. We checked this for all the figures and could not find the " $R_{\text{eff}}(D)$ " mentioned.

Clarifications needed in text

Comment: Line 245-250 (page 9): "For the unimodal size and mass distribution" – but you then use bi-modal fitting. Clarify that the unimodal fit is for the detectable range only, while bi-modal is for full distribution.

Reply: The Referee is correct that the wording was confusing when we say that it is for the size range from 10 – 1000 nm. All fits are on the detectable range and the mass fits comes from Eq. 1 (M_{BC}) and has the size range of 0 to infinity. The fitting parameters are thus only M_{BC} , MMD, and σ_m . This is also true for the bi-modal size distribution as the total mass is the sum of the two distributions ($M_{\text{BC,fit}_1} + M_{\text{BC,fit}_2}$) that results from

the bi-modal fitting. We removed the size range from the text as this is just the x-axis limits of the figure and was confusing in the text.

Comment: Section 3.4, line 355-360: The comparison uses MmD (mass mean diameter) from model vs. measured MmD. But Table 1 reports MMD (median) and a separate "MMD meas" column. Ensure consistent terminology throughout.

Reply: Thank you for this comment. We have removed any reference to MMD and keep the comparison between MmD in model and meas only. Table 1 and Figure 9 (now Fig 10) should be more consistent with each other in the revised manuscript.

Comment: Line 410 (page 14): "decreased with altitude at a rate of -5.3 nm km^{-1} – this is an interesting vertical gradient. Does your ground site at 565 m (which is above the surface but not free troposphere) align with extrapolations from aircraft profiles?

Reply: Thank you for this comment. Indeed, our observations at ground level seem to be approximately consistent with the results obtained at higher altitudes when applying this gradient. However, the gradient is calculated for MmD and the variability in flight altitudes and in how the results are reported between various studies does not enable very detailed comparison based on extrapolation.

Comments by Anonymous Referee #3, 31 May 2026

Minor comments:

Comment: Lines 45-46: The text states that when aged BC is cloud activated, the fractal-like structure of fresh BC will collapse. Mikhailov et al. (2006) showed that this restructuring occurs when the relative humidity is increased to saturation, rather than during true cloud activation. Please consider rephrasing this sentence. For example: "When coated BC is exposed to water-saturated conditions, its open, fractal-like structure collapses into a compact geometry (Mikhailov et al., 2006)."

Reply: Changed as suggested.

Comment: Lines 88-90: Please rephrase this sentence to improve the grammar and clarity.

Reply: The sentence "Although the focus was on evaluating three different methods for measuring BC, the authors also reported in their seasonal averages for size distributions for four seasons based on log-normal fits to a narrow size range. "

was modified to read:

"Although the study focused on evaluating three methods for measuring BC, the authors also reported seasonal mean size distributions for all four seasons"

Comment: Lines 137–139: Please check this sentence for punctuation errors.

Reply: Thank you, the punctuation typos were corrected.

Comment: Lines 195 and 197: Define symbols CMD and σ_c in the text.

Reply: Explanations were added

Comment: Line 199: The authors state that the SP2 field measurements at Pallas were conducted between November 2019 and December 2020. However, Section 2.3 states that the CAM5-ATRAS model simulations cover the period 2009–2015. While the text later justifies this timeline gap by showing that the observed BC mass median diameter (195 nm) is similar to 2011 observations (194 nm from Raatikainen et al., 2015) proving the size distribution shape is highly constant over the years, this connection should be made clearer earlier on. Please include a brief discussion in the methods section explaining how a model simulation ending in 2015 can be reliably used to validate observations from 2020, given the expected interannual variability in Arctic transport pathways and biomass burning emissions.

Reply: Thank you for this suggestion. The mismatch between the measurement and modelling periods was also raised by the other Referees, and we have now addressed the issue more thoroughly. Although we were unable to repeat the modelling exercise for a period overlapping with the SP2 measurements, we examined the year 2020 with respect to specific emission and transport episodes, including possible biomass-burning influence in January.

We also note that our results are similar to those reported by Raatikainen et al. (2015) for winter 2012. This agreement provides additional support for our conclusion that the BC core mass-size distribution shows relatively limited temporal variability and suggests that the 2020 observations may be broadly representative beyond the measurement year. However, we now explicitly acknowledge that differences in meteorology and source contributions between the measurement and simulation periods may affect the comparison. Accordingly, the comparison is interpreted in terms of climatological variability and systematic model bias rather than direct year-specific agreement.

Please see our response to Referee #2 for further details and for a description of the corresponding revisions made to the manuscript.

Comment: Lines 262-263: A secondary mass mode peaking at 475 nm is rather unusual, as it typically implies highly episodic events. Furthermore, this peak does not correspond to a distinct physical mode in the raw data, but rather appears to accommodate a slight leveling-off of the data points near the SP2 detection boundary (537 nm). Given its proximity to the edge cutoff and its absolute invariance across seasonal datasets, this second mode can be a mathematical artifact of the bimodal fitting algorithm rather than a distinct physical population of super-large black carbon cores. This could also explain the small drop in r^2 observed in the 12-hour corrected data. Please clarify the physical origin of this second mass mode or explicitly address it as a numerical fitting limitation.

Reply: See the replies to Referee #2 where we address this in full and provide additional statistics to the revised manuscript on the second mode. This approach is consistent with previous research about shoulders on BC mass distributions. Shoulders are not unusual. People often use a unimodal fit which does not capture this shoulder at the upper end of the detection limit as shown by Pileci et al. (2021). Distinct modes are not always visible. In the revised manuscript, we acknowledge that this second mode is empirical and fits the data much better than a unimodal fit. We added the fitting parameters to the revised manuscript, which were 100 – 350 nm for the smaller mode, and 350 to 540 for the bigger mode. Changes to the manuscript:

“Here, log-normal fits were done using the method described by (Hussein et al., 2005), which we modified to work with BC mass distributions; one mode between 100 and 350 nm, and another between 350 and 540 nm.-”

Comment: Lines 331–342: The claim that Arctic BC size distributions are homogeneous is oversimplified. Comparing the Pallas winter average (209 nm) to Ny-Ålesund (228 nm) reveals a 19 nm shift, which actually corresponds to a ~30% increase in core mass per particle due to D^3 scaling. Please soften this homogeneity claim and briefly discuss how these systematic offsets likely reflect distinct regional transport pathways or aging timescales.

Reply: This was an important point. The homogeneity claim was softened throughout the manuscript. However, when looking at MMD, σ_m and emissions of BC into a model, from a modelling perspective, there is no D^3 scaling. Changing the MMD, given that M_{BC} and σ_m stays the same, the mass stays the same. But for radiative effects, this 19 nm shift might still be significant and warrants future research into the topic.

Indeed, there are interesting events to look at more carefully, but the results do not support temporal variability would be significant e.g. for global model validation exercises.

Comment: Line 353: Typo on punctuation.

Reply: Corrected

Comment: Figure 9 and Caption: The text description in the caption contradicts the plot colors and legend. The caption refers to a "yellow line" and a "red line," but the plot actually displays an orange line (MmD PAL) and a green line (MmD PAL model). Please rewrite this caption to accurately match the colors and variables shown in the figure.

Reply: Corrected

Comment: Lines 411–412: The phrasing here oversimplifies the cited text: "In summary, the Aircraft campaigns from 2009 – 2017 in spring and summer, for all altitudes, the MMD was 187 ± 16 nm." In Jurányi et al. (2023), the 187 ± 16 nm value is explicitly specified as the spring average, while their summer observations were lower (around 170 nm). Please rephrase this sentence to separate the spring and summer values so it accurately reflects the cited study.

Reply: Thank you, this is a very important correction. We modified the text accordingly.

Comment: Lines 420–423: The observation that background BC core sizes decrease with altitude is a compelling feature of the vertical profile. To provide a physical explanation for this trend, please consider attributing this gradient to size-dependent wet scavenging during long-range transport (e.g., Moteki et al., 2012; Ohata et al., 2016).

Reply: Thank you for this helpful suggestion. We agree that size-dependent wet scavenging provides a plausible physical explanation for the observed decrease in background BC core size with altitude. Previous aircraft observations have shown that larger BC-containing particles are preferentially removed during cloud processing and upward transport, leaving a smaller BC (core) size distribution in the free troposphere (Moteki et al., 2012; Kondo et al., 2016). We have added this interpretation to the revised manuscript. However, because the compiled vertical profile combines observations from different regions, seasons, and transport conditions, we present size-dependent wet scavenging as a likely contributing mechanism rather than attributing the gradient exclusively to this process.

Added text: "This trend is consistent with size-dependent wet scavenging of BC-containing particles during transport (Moteki et al., 2012; Kondo et al., 2016) which could have contributed to this result. However, it should be noted that the compiled vertical profile combines observations from different regions, seasons, and transport conditions increasing the uncertainty."

Comment: Line 446: Typo. Change "do not wary substantially" to "do not vary substantially".

Reply: Corrected.

Comment: Lines 456-461: Please soften this conclusion to acknowledge that systematic regional mass variations remain significant for climate models.

Reply: We added text here: "While our analysis focused on long-term mean BC core size distributions, we also observed substantial short-term and event-to-event variability that warrants further investigation. In particular, BC core size distributions responded differently among individual in-cloud episodes, but these contrasting responses largely cancelled out when averaged over the full measurement period. Thus, the apparent stability of the long-term mean should not be interpreted as an absence of short-term variability. Such variability may be important for process-level understanding and model evaluation, especially on regional scale, even if its net effect on annual or seasonal mean BC core size distributions is limited."

Comment: Lines 465-466: As noted previously, this specific value represents only the spring average from their study. Please correct the description here to keep the seasonal context accurate.

Reply: Corrected

Comment: Line 470-471: The verb is missing.

Reply: Thank you. Corrected.

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