

Dear Dr Charles Brock,

I am on behalf of all the authors of this manuscript very grateful for your excellent and prompt editing work that has truly helped to improve this manuscript. Also, I would like to express our thankfulness to both excellent reviewers who have done so great work. I appreciate this very much.

We truly hope that you find the manuscript improved, from the original and the previous version. Please, do not hesitate to contact me for any requests of modification or additional corrections, advises or technicalities.

Below you may find our answers to Reviewer #2 questions and concerns. The manuscript has been modified accordingly and marked in the pdf.

Kind regards,

Eija Asmi

Answers to Referee #2:

The authors have made some attempt to reframe the manuscript toward focusing on intercomparing the absorption instruments, which is an improvement. However, the overall depth of analysis for this paper remains very shallow, and I'm left wondering what the main findings / recommendations of this work are for the scientific community as promised by the last line of the abstract: "provide some useful guidelines for instruments selection and uncertainty analysis". While the revision appears to be moving in the right direction, the manuscript is not yet at a level of scientific quality where I could recommend it for publication in AMT. Consequently, I recommend that the manuscript be returned to the authors for another major revision/rewrite that focuses on improving its scientific contribution and depth of analysis, conclusions, and recommendations.

REPLY: We thank the reviewer for acknowledging the work done for improving the manuscript. We are happy to hear the direction has been correct. We intend to continue to improve the presentation of our results, with the goal to reach a good quality manuscript with clear conclusions and a scientific message.

General comments:

1) What is the new development, significant advance, or novel aspect of in situ aerosol measurements being communicated by this manuscript? Why are the results meaningful and important?

REPLY: While several previous intercomparisons of absorption measurement instruments in both in field and lab have been done, their reported results highlight the instrument performances at medium- to high-concentration ranges and are nearly neglecting the measurements done at around or just above the zero concentrations (e.g. Mölders and Edwin, 2018). We found only couple of previous studies that have contributed to this major knowledge gap. The following text was added to introduction:

“The co-located PSAP and CLAP instruments showed a good agreement in real-world Arctic inter-comparison (Ogren et al., 2017), but the study did not include Aethalometer or MAAP instruments, which are slightly different techniques. The co-located aethalometers in the Arctic showed relatively more discrepancies, which were discussed by Mölders and Edwin, 2018. However, to the best of our knowledge, comprehensive studies of co-located parallel filter-based instruments inter-comparisons in pristine field environments are lacking and that is one reason for the poorly quantified Arctic absorption baseline. “

We focus here to this previously neglected concentration range. We show using both new and previous results that at very pristine environments these low concentration levels are frequently observed, and form part of the important concentration baseline. According to the instrument manufacturers, the devices can measure down to very low concentrations. Here we present evidence that this is indeed quite the case, and estimate the lowest detection limit and accuracy of the absorption measurements.

Mölders, N. and Edwin, S. (2018) Review of Black Carbon in the Arctic—Origin, Measurement Methods, and Observations. *Open Journal of Air Pollution*, **7**, 181-213. doi: [10.4236/ojap.2018.72010](https://doi.org/10.4236/ojap.2018.72010).

Ogren, J.A., Wendell, J., Andrews, E. and Sheridan, P.J. (2017) Continuous Light Absorption Photometer for Long-Term Studies. *Atmospheric Measurement Techniques*, **10**, 4805-4818. <https://doi.org/10.5194/amt-10-4805-2017>

2) Based on the instrument measurement intercomparison presented in Section 3.3 and in Figure 4, it is clear that there are significant differences between the absorption measurement techniques. In particular, the AE31 significantly underperforms the AE33, while even the AE33 and COSMOS appear to have non-zero intercepts. While the PSAP-MAAP comparison looks pretty good, there are also some notable departures from the 1:1 line where the PSAP is seeing very low or negative values. What is going on with the instruments and air masses during these periods? This intercomparison is the only real meat of this study, and there needs to be some additional discussion about what is being observed here and why (not just simply campaign-average slopes and  $R^2$  values). In addition to adding more discussion to Section 3.3, I'd like to see recommendations for how to use these results to make better, future measurements of aerosol absorption, scattering, and extinction at low-aerosol, Arctic sites like Pallas.

REPLY: We thank reviewer for this excellent comment. Generally we feel that the filter-based absorption instruments agree surprisingly well in these demanding conditions, but as pointed out by reviewer, there are also several interesting discrepancies shown in Figures 2 and 4 that we examined more carefully. Most interestingly, at extremely high SSA values the PSAP scattering-corrected data appears to be “over-corrected” while at lower SSA values, the scattering-correction appears to be necessary to correct the data to a level of MAAP. In the manuscript the AE31 and the AE33 data were not corrected with a scattering-correction function due to the practical reason that it doesn't exist for AE33 and also, due to the reason that a common practice in GAW is to use Cref value instead. This difference finally shows in our data analysis giving support for the necessity of the scattering correction as a general rule, when possible, however, keeping in mind the possibly increasing noise. We added now more discussion on these matters in the text. In addition, we wrote our recommendations clearly in conclusions, including preference to select filter-based instruments in low-concentration environments, include sufficient averaging of data to reach the instruments detection limit, use of co-located instruments as transfer standards to estimate data precision, have co-located scattering measurements if not provided by the absorption instrument and to be

specifically cautious in data analysis at  $>0.99$  SSA values, which still occur in the Arctic, as discussed in section “Representativeness of the measured absorption values”.

3) A lot is made of the extinction minus scattering (EMS) absorption estimates, which one would've expected to perform poorly under low aerosol loadings and high scattering:extinction ratios (i.e., where one is differencing two relatively large numbers to calculate a much smaller absorption coefficient). True to these expectations, the differencing technique does perform poorly. I don't see this as a real meaningful or important finding and suggest that Figure 3 is not necessary.

REPLY: We have reduced discussion on EMS-technique and corresponding results as compared to the previous manuscript version. We still find it relevant to keep a small amount of results in the discussion, especially considering that EMS is a good candidate for traceable absorption reference method, it needs to be clearly acknowledged in the literature why it can not be used directly in the field, or at all concentration ranges that are met in the field, and is thus limited in range.

4) I understand the authors' hesitance to bring in new data and expand the scope of the study. Since there are previous long-term studies made at Pallas, it would be helpful to synthesize the results from those studies to contextualize the results being presented here. For example, what are the typical absorption, extinction, and scattering coefficients reported by these prior Pallas studies for June-July and throughout the year? How does the 2019 summer compare to "typical" conditions? Are there periods where aerosol absorption is higher than what was seen in 2019?

REPLY: We added a new subsection on this, putting these new findings into the context of Pallas (Arctic) “typical” absorption levels.

Specific Comments:

Line 125: What is ATN? What are the units of 60 ATN?

REPLY: ATN compares the light intensity after passing through a filter with aerosols to that of the intensity while passing through a clean filter. The definition is given e.g. on Backman et al. 2017, Eq. (1). Reference was added.

Line 142: What is insufficient about the manufacturer correction? This is not clear from this manuscript.

REPLY: The sentence is now corrected as: “...is too low for most atmospheric aerosols (Laing et al., 2020)”.

Line 171-172: Please add some additional discussion about why the absorption Angstrom exponent would be uncertain for aerosol with SSAs close to unity. Is this because the absorption signals are low or are there scattering interferences that are not well accounted for? This uncertainty is likely to be especially important for the Pallas site.

REPLY: This comment makes reference to Backman et al., 2014 paper, where scatter in AAE is shown to increase with increasing SSA for Virkkula et al. 2010 correction (in particular). This was clarified in modified manuscript text: “However, it should be noted that at high SSA values the corrected  $a_{AP}$  becomes uncertain and should be interpreted with caution, as shown in Backman et al., 2014 Figure 7: intercomparison of correction algorithms.

Line 225: I don't understand why it is noted here that EMS is traceable to SI units. This is just differencing the extinction and scattering measurements discussed in previous sections.

REPLY: This is mentioned for the project connection to metrology.

Lines 226: It's true that differencing two measurements avoids filter-based artifacts, but there can also be other sources of uncertainty that are quite large. In particular, when the aerosol SSA  $\sim 1$ , the EMS technique becomes very uncertain as it involves difference two numbers of similarly large magnitude relative to the absorption.

REPLY: Exactly.

Line 239: How do the different geometry correction factors explain why the CAPSex and CAPSssa derived EMS calibrations would be different? Is this disagreement caused by the CAPS monitors or by the nephelometers?

REPLY: This was clearly confusingly written. The geometry correction factors were proposed to explain the differences observed in CAPS. There were very minor differences in nephelometers too, explained by other factors mentioned in text. The text was slightly improved.

Lines 239-241: I don't think that the instrument flow rates and/or inlet settings would have any impact on the correction factors. Hopefully, these calibrations were done with conductive tubing and at steady state conditions, and, ideally, with most of the actual ambient sampling setup. Are these sentences suggesting that there were significant particle transport losses that bias the calibration procedures?

REPLY: Agree. The calibrations were done with conducted tubing with similar length and setting, using an aerosol with Ångström exponent similar to outdoor air (in range 1.5-1.7). Text was slightly modified.

Line 286-287: I don't think it can be stated that there is even qualitative agreement. This is particularly the case in July where the EMS inferred values are trending up even as the direct measurements show no obvious trend.

REPLY: Good point. The agreement is weak, and in modified Fig. 2 it is obvious that EMS-derived absorption follows more closely the scattering and extinction time series. Text was slightly modified to clarify this.

Line 288: Do you think that the high absorption values implied by EMS of  $\sim 3 \text{ Mm}^{-1}$  are real or caused by taking the difference of two, large, noisy numbers?

REPLY: This is most likely the explanation, and was added in manuscript text.

Lines 312-313: I think it's fine to choose the MAAP as a common reference for comparison, but I'm confused about the rationale presented in the manuscript that there is "low demand of any artifact related post-correction in MAAP". What does this mean?

REPLY: Corrected as "MAAP is known to be essentially an artifact-free technique to measure aerosol absorption on a filter and has been widely utilized as a practical field reference method also in the past."

Lines 354-357: Please report averaging intervals for these limits of detection.

REPLY: Done.

Line 357: What support is there for the statement that EMS methods are applicable above 0.1 Mm<sup>-1</sup>? Indeed, I would think that the applicability of these methods would be dependent on both the instrument signal-to-noise as well as the aerosol SSA.

REPLY: This is true. We modified the sentence to state that our evidence only shows that the method is not applicable at below 0.1Mm<sup>-1</sup> levels and that due to the nature of the method (taking a difference of two large numbers), it is supposedly also highly dependent on the aerosol SSA.

Figure 2: Please make these panels full page width and add dashed lines to the lower panel that show the -0.2 to 0.6 Mm<sup>-1</sup> range. Also, please include additional panels showing the time series for the scattering and extinction coefficient measurements as well as the SP2 BC timeseries.

REPLY: These were added in Figure 2.