Answers to reviewers

We wish to thank both reviewers for their excellent comments and suggestions that have greatly helped us to re-structure and improve the presentation of our campaign results in the form of this manuscript. We hope that the reviewers find the changes acceptable and the current version of the manuscript more appropriate for publication in AMT. We have modified the manuscript content significantly. We summarize below the major changes that were done and answer the detailed reviewers' comments below the summary.

Major changes, summary:

The modified manuscript focuses entirely on instruments capabilities for detecting low concentrations in a pristine field environment. All discussion on atmospheric relevance and implications of the measured aerosol concentrations, including the analysis of air mass origin and Arctic BC mixing state were removed. All figures, tables and instrument correlations were re-calculated using 100% of campaign data (i.e. not separately for periods 1, 2 or neglecting some periods). This affected slightly the correlation statistics described in subsection 3.4 (current 3.3). Results section structure was modified such that the former subsection 3.3 (on detection limits) was moved at the beginning of section 3 (current 3.1) and Allan variances were calculated and used to determine the lowest averaging times of the instruments. Subsection 3.1 is now followed by modified and compressed former sections 3.1 and 3.2 (campaign overview and observed absorption values). The former section 3.5 (Particle size and coating impact on measured absorption) was removed completely and a new short section 3.4 with a simplistic MAC-value calculation was added. The main conclusions of the modified manuscript are:

- filter-based methods are sensitive to detect absorption coefficients down to around 0.01 Mm-1 level (1-sigma) while the use of EMS method requires around 10-fold higher absorption coefficient values
- Arctic summer absorption values were most of the time between 0.06-0.1 Mm-1 in our study, which is well above the lowest detection limits of filter-based instruments, but too low for EMS methods
- Even at these low concentrations, the absorption values measured by different filter-based instruments show a good linear correlation, confirming both the accuracy and the precision seem to be adequate. Here the exception was the absorption measured by COSMOS instrument, where the pre-treatment of the sample can, as expected, modify the measured absorption. This led to a slope clearly below 1 between MAAP and COSMOS absorption.
- Mass-absorption cross section of 16 m2 g-1 was determined for MAAP, when compared to a residual BC mass measured by SP2. This is well in-line with the MAC values obtained in a comprehensive study in the Arctic by Ohata et al., 2020.

Following references were added: Werle et al., 1993; Ohata et al., 2020; Allan et al., 1966; Hagler et al., 2011; Springston and Sedlacek, 2007; Laing et al., 2020; Modini et al., 2021; Torseth et al., 2019; Bond et al., 2006; Jacobson 2001; Sinha et al., 2017.

Answers to Referee #1

The work of Asmi et al. presents optical properties of Arctic aerosol measured with a wide array of instruments. Considering the global intensive use of the considered instruments, understanding and quantifying the issues of each instrument in order to optimize its performances is an essential task. However, the present manuscript lays between a technical assessment of the performances of filter-based absorption photometers and a survey of arctic aerosol optical properties. Thus, the objectives of the manuscript are not very clear nor are the scientific and the technical conclusions. The dataset is of undoubtable value, the authors have, nonetheless, clarify their technical or scientific message. I do not recommend the publication of the manuscript in its present form. However, with the hope that my comments will be helpful to the authors, I suggest a major rethinking of the manuscript.

ANSWER: We thank reviewer for their time and effort dedicated towards our work. We largely share the reviewers' concerns and appreciate the very useful comments and suggestions made to improve the content and the structure of the manuscript. We have done our best to modify the text accordingly and hope that the reviewer finds the current form of the manuscript more appropriate for publication in the journal.

MAJOR COMMENTS My biggest concern is represented by the overall "take home message", which is hard to grasp. The "Campaign overview" and "Absorbing aerosol characteristic" are not of scientific relevance, since similar results have been widely presented in previous and more comprehensive works. Hence, the characterization of aerosol properties and airmass origin, which does not have a clear impact on the instrumental comparison, adds only confusion. As an example, the distinction between period 1 and period 2 is not used in the more technical part. The 5 filter-based absorption photometers agree to a variable degree. The actual causes are, however, not clear or not investigated. As a matter of fact, Section 3.5 provides, citing the manuscript, "un-ambiguous evidence" on the impact of mixing and size on optical measurements. The SP2 is used to provide the degree of internal mixing of rBC particles. Why this is done with the lag time technique and not with the LEO fit. Although both are prone to large uncertainty, the first is only qualitative. If my understanding is right, the Mode1 Mode2 classes are based on a lag time distribution. It appears that the lag time analysis was applied to all BC particles. This might cause substantial bias in the fraction of thickly and thinly coated particles. By limiting the analysis to BC cores falling in the detection range of scattering detector, the fraction of thickly coated particles should degrees (see specific comments below). Hence, the very interesting and also surprising results shown in 3.5 might be wrong.

ANSWER: We do share reviewers concerns regarding the "take home message" and content of the manuscript. To respond to these concerns, we gave a thorough thinking on the ms structure and decided to completely re-structure the manuscript and sharpen the objectives. In the modified manuscript the objectives are to study the 1) absorption instrument's stability and detection limits with respect to the atmospheric concentrations commonly measured in pristine environments and 2) instruments accuracy when operated at the edge of their lower detection limits. In addition, we calculate 3) an Arctic-specific mass absorption cross-section (MAC) value based on SP2 as an rBC reference, and MAAP as an absorption reference. Any previous publication provides such a comprehensive instruments comparison in such a pristine field environment, yet, the very same instruments are often applied in these environments and data is compared (Scheisser et al., 2018; Tørseth et al., 2019; Ohata et al., 2020). To keep the "take home message" simple and sharp, we have removed all discussion on the climatic relevance of the results, and removed the sections on air mass analysis, aerosol mixing state and optical properties of atmospheric aerosol. We do agree with the reviewer that those have been more thoroughly characterized in several previous publications.

SPECIFIC COMMENTS: line of text; F: figure; S: section

Title: from the title the reader might expect a soot optical characterization on a large Arctic scale. I suggest to slightly modify the tittle specifying the location of measurements.

ANSWER: We accommodated the title with the new content and focus of the ms, taking into account this suggestion. The new title is “Absorption instruments inter-comparison campaign at the Arctic Pallas station”

L32: Worth citing the NILU report : https://www.amap.no/work-area/document/3058

ANSWER: Cited.

L49-51: I find the statement about MAC and eBC a bit out of place and might generate confusion. eBC should be rather mentioned in the filter-based instrument paragraph. Since the eBC and MAC are mostly used for the filter-based instruments, I suggest to mention them a bit earlier.

ANSWER: This sentence was moved in the end of the previous paragraph.

L78: It would be appropriate to shortly summarize the goal of the manuscript in this last paragraph, or clearly state that this work was performed within the framework of EMPIR BC, if this is the case.

ANSWER: The following text was added: “The goal was to test the stability, accuracy and detection capabilities of the commonly available absorption measurement methods focusing on the filter-based techniques, and to conclude on their applicability to pristine environments. To our knowledge, this is the most comprehensive absorption and BC mass measurement instrument parallel field comparison done in the Arctic.”

L95-100: description of goals and objectives does not belong to method sections, more to introduction.

ANSWER: Done.

L100-109: in this subchapter there are many abbreviations of the various instruments, which might become overwhelming and confusing to a non-expert reader. I suggest to move this plumbing description in a separate subchapter after the instrumental description.

ANSWER: These were moved to a new subsection “Sampling”

L145: I would not use the abbreviation MAC to describe the coefficient used internally by the AE31.

ANSWER: Agree. Term “MAC” was removed.

L110-126: only the SSA is really described. The MAC is briefly described elsewhere in the text. I suggest to rework a bit this part in order to provide a more systematic and inclusive description of all optical properties.

ANSWER: The text was re-checked and explanations on absorption coefficient and MAC were added.

L159-160: This is actually a very good point

ANSWER: I would assume to have some recommendation on this soon. Also the information given here adds to the knowledge.

L192: Continuous soot monitoring system...capital or non capital?

ANSWER: Non-capital, thanks for pointing this out.

L193-195: Many periods in this sentence, writing could be smoother and more enjoy-able. This is a constant feature of the paper. I suggest the authors to work a bit on it.

ANSWER: We hope to have improved the writing, here and in other sections too.

L206: not sure if capital letters for Black Carbon are needed.

ANSWER: True. This, and other similar mistakes were corrected.

L210: The work of Lim focussed on SP2 measurement in snow. I think there are better references: (Laborde et al., 2012).

ANSWER: Reference changed.

L211-213: as stated: “This technique is very sensitive but does not measure particle light absorption as such, and therefore, a direct comparison with other absorption measurement techniques is not straightforward.”. So, what the SP2 is used for in this work?

ANSWER: The original idea to use SP2 was to describe the atmospheric mixing state of the absorbing aerosol which affects the absorption enhancement. In the modified manuscript version, SP2 is used as a rBC mass reference to define the value of MAC. This is now clearly said in ms text.

L215: provide CAPS full name
ANSWER: Done.

L220-221: Despite very recent and in review, I suggest giving a look to (Modini et al.,2020).

ANSWER: Thank you, this was very interesting. The work by Modini et al. is now cited at the discussion of the CAPS accuracy, stability and error sources.

L262-270: these two subsections (2.4.10 and 2.5) are very short. I suggest combining them together with the plumbing description (L100-110) into a unique subsection: “Additional tools and methods”

ANSWER: In the modified ms some of these sections (air mass analysis, CPC) were completely removed. The remaining notes on AWS were connected with the suggested new section entitled “Sampling and environment”.

L273-277: these numbers are not very useful without any reference for comparison. Are these pristine, background, polluted conditions for Finnish Arctic? Considering the influence of different airmasses and, thus, different aerosol loads and properties, averaged values are definitely not of interest. I suggest removing this paragraph.

ANSWER: In the modified ms the absorption time series and the averaged values are compared to the instruments detection limits, and further to those typically measured in the Arctic. We feel that it is important to make the point that around the Arctic, such low concentrations do exist, justifying the need to understand the instruments capabilities to measure such low concentrations. In the end of section 3.2 we state: “The measured absorption coefficient values are in the lower end of that typically observed at Pallas site.

Lihavainen et al., 2015 long-term analysis showed that a sigma_ap in Pallas summer ranges between 0.1-1 Mm-1, where the lower values represent the clean Arctic air flow. Thus, the measured sigma_ap values during the campaign are well representative of the values measured around the Arctic during summer (Schmeisser et al., 2018)."

L278-279: the distinction of the two periods in not very clear. Especially considering the back trajectories shown in F3 (see related comment).

ANSWER: The two periods are no longer separated and data is no longer filtered.

L288-291: "The aerosol optical size related parameter", is confusing. Simply use the symbol or "Angstrom exponent". I would be careful to jump into conclusions: the absence of precipitation and thus wet scavenging (both from nucleation and impaction) might cause increase of number concentration and diameter decrease. Moreover optical diameter measurements are not available.

ANSWER: True. All this discussion was removed.

L292: typically observed...add reference and potentially a value.

ANSWER: This was removed. Our main reference to Pallas aerosol optical properties in ms is Lihavainen et al., 2015.

L296-298: What do you mean with "averageσAP630nm"? Average between all instrument? Unclear. Same at L319

ANSWER: Average sigma_AP is always an average value measured with one instrument. We tried to formulate this more clear and make appropriate references to Tables and Figures with numbers.

L307-309: lag-time description...move this to technical section. Is the lag applied to all rBC signal or a to a specific rBC diameter range? Although this measurement does appear to be only qualitative here and does not need supreme robustness in this case, applying a lower limit to rBC particles diameter (let's say above the detection limit of the scattering signal) will reduce the number of thickly coated BC cores (the weak incandescence signals with no coating (total particle size below 150-200 nm) will not be seen by scattering detector). Not compulsory, but worth trying. This might change your statement at L310.

ANSWER: The lag-time analysis was removed completely.

L321-322: from my understanding COSMOS directly provide a "eBC" with the constant COSMOS-MAC value. Here absorption coefficient is presented, which MAC was used. Worth specify in the respective technical section.

ANSWER: Done.

L320-334: These paragraphs so not provide relevant information; Or, at least, it is hard to understand what the authors want to show.

ANSWER: We agree that this was repetitive with previous section. The point was to provide information on the atmospheric sigma_ap values measured by different instruments and their deviation. This is now completely re-structured and combined with previous sections.

L337: in the equation there is a ">". Is this correct or it should be " * "?

ANSWER: This equation is removed. The symbol was correct, though.

L361-365: Why? Is the low sensitivity the sole explanation to the bad correlation between EMS and MAAP ? This is quite interesting since CAPS and nephelometer should not suffer from filter matrix-effect.

ANSWER: This is a good question, and we are happy that you find it as an interesting observation to point out. It is one of the main conclusions of the manuscript.
Indeed, these instrumetns should not suffer from matrix-effects but rather the components noise and drift and truncation errors can explain the relatively high lowest detection limit of the techniques. Important is, that at high SSA values such as here (0.97) the absorption is defined from a subtraction between two big number which can amplify the errors (Modini et al., 2021).

L379-380: What do you mean with “a clear tendency”. I suggest plotting the AE31 and AE33 results in figure 8. Same for figure 9.
L393-395: the increase of absorbing organic carbon could be seen with the absorption angstrom exponent.

ANSWER: The last section was removed.

S3.1-3.2: Since the main focus is absorption I suggest merging these two sections
ANSWER: Agree, and done.

S3.4: is the analysis done on the full campaign or on a selected period?
ANSWER: Full campaign.

F1: this figure is partially needed to understand the scientific message of the paper. IC5would move it in the supplementary.
ANSWER: Done.

F3: I find the legend and caption a bit confusing. These are the 2 considered periods: Period 1 (June 19 – July 7) and Period 2 (July 7 – July 17). The legend does not reflect this partitioning
F5: what the meaning of bins is?
F6: Define the difference between panels
F7 the colour scale does not provide useful additional information. Scattering coefficient is not even mentioned in the text. I wonder if SSA might provide a more info.
F8 the x-axis label is a bit confusing, I suggest to use a more understandable label“Fraction of thickly coated rBC”

ANSWER: Most figures were either removed or completely modified.

REFERENCE Laborde, M., Schnaiter, M., Linke, C., Saathoff, H., Naumann, K.-H.,Möhler, O., Berlenz, S., Wagner, U., Taylor, J. W., Liu, D., Flynn, M., Allan, J. D.,Coe, H., Heimerl, K., Dahlkötter, F., Weinzierl, B., Wollny, A. G., Zanatta, M., Cozic,J., Laj, P., Hitzenberger, R., Schwarz, J. P. and Gysel, M.: Single Particle Soot Pho-tometer intercomparison at the AIDA chamber, Atmos Meas Tech, 5(12), 3077–3097,doi:10.5194/amt-5-3077-2012, 2012.
Modini, R. L., Corbin, J. C., Brem, B. T., Irwin,M., Bertò, M., Pileci, R. E., Fetfatzis, P., Eleftheriadis, K., Henzing, B., Moerman, M.M., Liu, F., Müller, T. and Gysel-Beer, M.: Detailed characterization of the CAPS single scattering albedo monitor (CAPS PMssa) as a field-deployable instrument for measuring aerosol light absorption with the extinction-minus-scattering method, Atmospheric Meas. Tech. Discuss., 1–56, doi:https://doi.org/10.5194/amt-2020-292, 2020.

ANSWER: References added.

Answers to Referee #2

This manuscript describes measurements of aerosol concentrations and optical properties (absorption, scattering, extinction) measured during a month-long period at the Pallas ground site in northern Finland. Filter-based aerosol absorption instruments include two models of aethelometers, a MAAP, a PSAP, and COSMOS. Extinction and scattering coefficients are obtained from CAPS and nephelometer instruments, respectively. Information on black carbon mass and coating thickness is obtained from an SP2, while particle number concentrations are measured by a CPC. In total, this is a comprehensive aerosol measurement suite! The primary focus of the study is to examine the instrument consistency during two time periods – Period 1 is characterized by relatively low aerosol scattering coefficients, while Period 2 sees higher particle scattering coefficients. Aerosol absorption coefficients and number concentrations are similar across both periods, after removing cloud/fog artifacts when ambient visibility was greatly reduced. Backtrajectories are included to provide context for air mass history, which indicate consistently northeasterly winds during Period 2, while the wind directions during Period 1 are much more variable. Despite some degree of variability between the two time periods, the overall aerosol absorption, scattering, and number concentrations are quite low, which would be expected for the Pallas region (far from continental pollution aerosol sources). This challenges many of the instrument comparisons because the measurements are close to the lower limits of detection, and it appears that the data are highly averaged to reduce noise (which is appropriate and fine). Overall, the manuscript is well written, enjoyable to read, and nicely describes the Pallas scientific instruments and the observations obtained during this time period; although, the scientific importance of these observations is not clear from the paper. The depth of analysis is pretty shallow as only a short period of time is being examined and the mean concentrations shown in Table 2 tend to be close to zero (with large standard deviations relative to the magnitude of the mean), which makes it hard to draw conclusions regarding instrument agreement/disagreement. I'm also unsure if AMT is the appropriate journal for this manuscript. All instruments are commercially available and have previously been described in the literature, and there don't seem to be novel conclusions related to their performance and/or operation that would be informative to the broader scientific community. Since I do not think that the present paper would be acceptable for publication without bringing in additional data and completely rewriting/reframing its scope, I recommend that it be rejected for publication in AMT.

ANSWER: We appreciate your kind words and an excellent summary on our work. We take the message that the manuscript in its previous form is rather confusing and presents no clear value for the community, for which we have completely re-written the results section. Many of your excellent suggestions have been considered in the modified manuscript, and for example the Periods 1 and 2 are no longer separated, and the focus is not on aerosol climatic relevance, but rather on instruments ability to measure in such pristine environments, for which this campaign data serves as a unique data set and can show a broader value. We hope that you find our changes adequate.

Specific Comments:

1) As stated above, the depth of analysis in this paper is low. Simply comparing the data from multiple, filter-based aerosol optical property measurements for a month and reporting summary statistics that largely overlap with zero provides little value regarding the instrument operation or the remote measurement site characteristics. One way to increase the depth of analysis might be to reframe the paper to describe the annual aerosol climatology relevant to the Pallas site. This might include monthly and seasonal data from satellites and/or models that provide some long-term context for the site measurements. It would also be great if more in situ data could be included beyond a single month; although, I recognize that that might be prohibitive. Having more in situ data might allow for more dynamic range in the aerosol abundance and optical properties. Another approach to increase the depth of analysis might be to examine the Pallas aerosol instrument suite response to laboratory-generated aerosol

under very controlled conditions to understand their interconsistency and accuracy. These results could then be compared to the ambient measurements to understand/evaluate the instrument performance. These are just a couple of ideas for the authors' consideration in a potential future manuscript.

ANSWER: Thank you for your excellent suggestion on how to modify the focus of the ms. Finally, we took the decision to present the campaign results in a slightly different context, but with adding no data. We hope that you find this acceptable. One reason is that there are several previous long-term studies made at Pallas and other Arctic sites, but very view studies have focused on the instruments inter-comparisons in pristine field conditions. Secondly, we aim at reporting a laboratory characterization on these very same instruments. However, the results do not fit in this paper and it would change the scope completely, giving very little meaning to present the atmospheric inter-comparison, which is here the main focus. There are previous laboratory studies, too.

2) The sensitivities and lower detection limits given in the abstract (and elsewhere) need to include the averaging interval. I'm assuming that 0.05 Mm-1 as achieved by averaging for > 1 hr. What are the overall instrument accuracies?

ANSWER: Yes, these were added. Defining the accuracy is a question of a reference. Here, we used MAAP as an "absorption reference" and SP2 as a "mass reference", however understanding that these can not provide real references as such. The PSAP provided the best comparison to MAAP.

3) On Line 24, what is meant by the statement, "additional activation of secondary particle formation mechanisms"?

ANSWER: Removed.

4) Strike first sentence on Line 29 as not relevant. The second sentence is referencing a filter-based measurement method, which I'm assuming is of aerosol absorption yes? Also please correct typos on Line 29 (begun -> began) and Line 31 (graphic ->graphitic).

ANSWER: These were corrected as suggested.

5) On Line 96, it is noted that the Arctic aerosol absorption measurements are "demanding". Is this related to the low concentrations due to a lack of local combustion sources? If so, it might be worthwhile to clarify that, while also noting that such, low-aerosol conditions are also not unique to the Arctic.

ANSWER: This part was re-formulated to refer to pristine environments where techniques suffer from signal-to-noise challenges.

6) Is the mass absorption coefficient (MAC) defined on Line 145 the same as the MAC used on Line 200? On Line 146 it is called a "wavelength dependent specific attenuation".

ANSWER: MAC was removed from line 145, because it is not exactly the same. Thank you for notice.

7) What value(s) of the MAC were applied as mentioned on Line 201?

ANSWER: COSMOS MAC was 8.73 m2 g-1. This was added.

8) On Lines 248 (and elsewhere), I find the extinction-minus-scattering (EMS 1 2) terminology confusing because this is neither an instrument nor a complex method. Rather, it's just a

difference between two values. I suggest that it would be worth denoting as σCAPSex−σTSI and σCAPSssa−σAUR4 to help avoid confusion and to be consistent with the notation in Figure 1.

ANSWER: We are aware that this name has been in use only for a short time, partly because the technique as such is still relatively little applied. However, some previous work (e.g. Modini et al., 2021) use the name “EMS” and we would therefore wish to keep it as it is, to clarify that the method is the same as in their ms.

9) On Line 257, it is noted that differences in CAPS instrument response may be due in part to "discrepancies in inlet tubing sizes and flow rates", which seems unlikely to me. Can this sentence be clarified to explain how these differences would affect the aerosol measurements?

ANSWER: We agree that the disagreement between the sampling line lengths and tubes was a negligible source of error and is unable to explain much of the difference. As we mention that the nephelometer sampling settings were slightly different, this could have a minor impact. This was re-phased now more clearly in the text.

10) On Lines 274-275, Table 2, and elsewhere, I note that the magnitudes of arithmetic standard deviations often exceed the magnitudes of the arithmetic means, which gives implies non-physical, negative values. Is this because of instrument noise that results in a negative baseline or is this because the observational data are not normally distributed about the mean. At least in the case of the CPC data reported in Table 2, it would seem that the latter explanation is correct, in which case it would be appropriate to report the summary statistics as either a geometric mean */ one geometric standard deviation or as median and percentiles.

ANSWER: You are correct that in some cases data are clearly not normally distributed and the std values lead to unphysical interpretations. We have now preferred the use of median and quartiles values, rather than averages and std in most of the ms tables and figures.

11) Rather than using the qualitative SP2 lag times, can the data be reanalyzed to provide quantitative coating thicknesses? I think that this is important and would help to increase the paper depth of analysis.

ANSWER: In the modified ms version, the aerosol mixing state is no longer discussed since the interpretation of this data (section 3.5) was too uncertain and speculative with such low concentrations and short campaign time. We hope that you understand this decision.

12) What support is there for the statement made on Lines 310-11 that the more thickly coated aerosols have longer atmospheric ages?

ANSWER: Referring to our previous answer, this section was removed from the modified ms.

13) The final conclusions on Lines 429-434 are not really connected to the data that are presented in this manuscript. What site- and aerosol-specific filter artifacts were presented and overcome? How were good measurement practices and careful data post-processing demonstrated in this study that differ from conventional techniques? To put it bluntly, what is new or novel from the present paper in terms of measurement techniques?

ANSWER: We removed this sentence and aimed at sharpening the conclusions according to the new ms structure, also to answer better “what is new and novel in this paper”.

14) There seem to be too many significant figures presented in Table 3, which imply the absorption coefficient measurements can be made with a precision of 0.001 Mm-1. I realize

that the’re a lot of averaging going on here to help tamp down the noise, but I suspect that at least the last digit is probably not significant.

ANSWER: This is probably very much true. We know that these 1h-average values are measured with instruments with a of lower detection limit on the order of ~0.01 – 0.1 Mm-1. What is the accuracy, is currently unknown since we do not have a reference method for aerosol absorption.