Reply to reviewers of the manuscript: "Increase in precipitation scavenging contributes to long-term reductions of black carbon in the Arctic"

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We thank the reviewer for their positive and constructive comments. We have modified our manuscript based on their suggestions. We believe that the comments received by the reviewer have greatly improved both the text and the science. Please find our detailed reply below (given in blue colour), whilst changes in the text are given in italics and quotations. We hope that the following responses to your comments satisfy the reviewer. All page numbers and line numbers are given at the start of each reviewer comment and reflect the page and line numbers of the original submitted manuscript.

1 Reviewer 1

1.1 SUMMARY

Heslin-Rees and co-authors present one of the longest time series of aerosol absorption coefficient in the Arctic region and correlate the trend with the origin of airmasses, precipitation and biomass burning events. The topic of the manuscript is well within the scope of ACP and is of main interest to the aerosol scientific community. The presentation and structure are, however, critical to the explanation of methodology and communication of scientific founding and its discussion. The conclusions are linear and clear and can be used as a guideline to restructure the full manuscript. Overall, I suggest the authors reworking the structure of their text (reduce the length of the manuscript, merge the result and discussion section, reduce supplementary and remove all non-essential data or information), and resubmit the manuscript for a second round of review. In the current state, the scientific message is not delivered properly while the quality of the data treatment is hard to ass, which is a pity, considering the rarity of the dataset.

1.2 MAJOR COMMENTS

1.3 Method section

I suggest restructuring this section. Aerosol, trend analysis and back trajectory and mixed together, making the methodology section not coherent I thus suggest using subsections in this order: 1) absorption measurement and data treatment including subsections on technical description, data correction, data harmonisation and trend analysis, and scavenging ratio; 2) Aerosol origin and transport including subsections on transport model, clustering method, season definition and forest fire identification.

Yes, we agree. We have restructured the method section completely, using your suggestions. Various paragraphs have been rewritten and the order of the subsections has also been changed in most cases.

1.4 Results and Discussion section

First, the result section is full of not-needed subsections. The manuscript is already quite long with many sections and subsections, try to reduce their number by merging subsections together. The result section reads mostly as a list of results without the proper scientific context, which is confined to the discussion section. In the latter, many results already presented in Section 3 are repeated, making Section 4 unnecessarily long and redundant. I suggest complementing the results with the data interpretation and discussion in the results section. Section 4 could be, then, shortened to a minimum, or completely removed.

Using your suggestion, we have effectively merged the results and discussion sections into one section i.e. "Results and Discussion". This has allowed us to save pages and reduce the total word count. In addition, certain results and subsections such as the one on the scavenging ratio have been completely removed from the results and method sections respectively. Furthermore, we have of course gotten rid of all the pointless subsections.

We have attempted to provide some structure to the manuscript by dividing the now "Results & Discussion" section into 5 subsections: 1. Long-term trends in absorption coefficient (σ_{ap}) and single scattering albedo, this subsection describes the trends in the optical properties, 2. Sources, sinks and transport, this subsection features the concentration weighted trajectory mappings and the spatial trends, thus incorporating all the 3 factors discussed in this manuscript. 3. Transport, this section dedicates itself to understanding the impact of the changing transportation pathways to the Zeppelin Observatory, by describing the clusters and then the trends 4. Sinks and transport, this subsection focuses on the trend in the accumulated back trajectory precipitation (ATP), and then attempts to estimate the impact this trend has on the absorption coefficient, by understanding the site-and-receptor specific relationships between ATP and σ_{ap}, and finally 5. Sources and transport is a subsection that focuses on biomass-burning events as a potential source which could alter the magnitude of the trends.

1.5 Supplement

Supplementary material is important to explain specific technicalities or show interesting, but not essential results. In this specific work, the supplementary is 16 pages long with 17 figures. In some cases (harmonization of data), critical information is included in the supplementary but not in the main text. So, the reader is forced to constantly check the supplementary material in order to understand the content of the manuscript. This back-and-forth disrupts the reading. Please find a way, not trivial, to avoid iterative references to the supplementary.

We have moved the main harmonisation figure to the manuscript, however, we wanted to use the manuscript to focus more on the trends and how to explain them, and less on the actual methodology including the harmonisation. We understand that the harmonisation is an aspect, however, we did not want to over-emphasise this part potentially overshadowing the results. Various results from the supplement have been removed and attempts have been made to shorten it.

1.6 SPECIFIC COMMENTS

The title is a bit misleading. The manuscript present absorption data, not BC data or eBC data.

We agree that paper has to do with absorption coefficient measurements, hence we have changed BC to light-absorbing aerosol in places which refer to our measurements or our findings. However, there is still plenty of literature relevant to this study which focuses on either BC, eBC or EC, and hence BC is still used in certain cases where it references other research. Nonetheless, in regards to the title we have changed it to the following:

"Increase in precipitation scavenging contributes to long-term reductions of light-absorbing aerosol in the Arctic."

Page 1, Line 3:

specify the region

We agree that we were not clear and it is important to state the region we are focusing on (i.e. the Arctic).

We altered the text, such that it reads as follows:

"Light-absorbing aerosol exert a warming effect on the Arctic, a region..."

Page 1, Lines 15-16:

These sentences are a bit unclear, what the authors mean with "explaining approximately a quarter of the overall trend" and "scavenging ratio" ?

We agree that it is a bit unclear what is meant here. We altered the text, such that it reads as follows:

"We argue that the increase in precipitation, as experienced by air masses arriving at the station, can explain a quarter of the

long-term reduction in the light absorption coefficient, stemming from precipitation acting as a sink of aerosol"

Page 1, Lines 17-19:

I would move the forest fire discussion up in the text, together with the "source identification" part.

We agree and the sentence has been moved up to line 13 of the abstract. The sentence has also been altered and parts deleted. It now reads as follows:

"The proportion of air masses en route to Zeppelin, which have been influenced by active fires has undergone a noticeable increase starting in 2015. However, this increase has not impacted the long-term trends in the concentration of light-absorbing aerosol, and it is concluded not to have caused the recent increase in light-absorbing aerosol."

Page 2, Lines 25-29:

please provide some references for the climatic impacts of BC.

We agree these statements need references. For the statement, "Despite the relatively short lifetime of aerosol particles compared to other climate forces" we chose Liu and Matsui (2021), for the statement, "they are still able to exert a major influence on climate" we have now referenced Bond et al. (2013), for the statement "Furthermore, the deposition of light-absorbing aerosol on snow and ice increases the impact light-absorbing aerosols can have in all semi-persistent snow areas, such as polar environments, by reducing the albedo and enhancing the melting of snow and ice." we have used Hansen and Nazarenko (2004) and Skiles et al. (2018)

Page 2, Lines 35-36:

absorption is poorly explained here, but I also think it does not belong to introduction but rather methodology.

We agree and have removed the following sentence starting on line 35:

"BC can be measured by utilising a specific property, the ability to attenuate light, at a given wavelength (λ). The light absorption coefficient (σ_{ap}) describes the amount of attenuation, per given length unit."

Page 3, Lines 55-74:

mix various topics (measuring challenge, long term dataset, cleaner combustion, emission reduction, aerosol sinks) in a relatively confused order. It is hard for the reader to follow the discussion and understand the message of this subsection.

We agree, this paragraph is poorly written and the sentences do not connect, hence we have rewritten it as follows:

"Long-term measurement sites, such as Zeppelin Observatory, mean that changes in key meteorological and aerosol parameters can be examined over an almost climatic time scale. Maintaining measurements in the Arctic poses numerous challenges, including being able to establish techniques sensitive enough to measure at low aerosol loadings (i.e. $\sim$ tens of ng/m^{-3}) (Sinha et al., 2017; Eleftheriadis et al., 2009; AMAP, 2021); however despite the challenges, the long-term time series that ZEP provides serve as important assets in understanding the transformations taking place in the Arctic. Studying the changes to aerosol optical parameters over decades sheds light on the anthropogenic influences on the Arctic from regions further south (Shaw, 1982; Stohl et al., 2007); numerous studies have reported on significant declines in sulphate aerosol loadings (Acosta Navarro

et al., 2016) and BC concentrations measured from the Arctic (Sharma et al., 2013; Collaud Coen et al., 2020; Bodhaine and Dutton, 1993; Schmale et al., 2022). This has led many to suggest that the decrease in long-range transported pollutants to the Arctic is the result of the advent of cleaner combustion techniques and the reductions in emissions from Europe and the former Soviet Union (FSU). It is important to note that during the same period, BC global emissions continued to increase due to increased emissions mainly from Asia (Klimont et al., 2017; Schmale et al., 2022). Nonetheless, few studies examine in detail other possible factors for the observed aerosol optical trends. The role that aerosol sinks could play when it comes to trends has received little attention (Garrett et al., 2011), despite the well-documented role scavenging plays on aerosol seasonality in the Arctic (Garrett et al., 2011). This lack of investigation could be one of the reasons why models fail to replicate observations of BC (AMAP, 2021). Furthermore, scavenging could also explain why there is a pronounced difference between trends in atmospheric and BC ice core measurements; ice cores from Svalbard glaciers present rapid increases in BC (operationally defined as elemental carbon) between 1970 and 2004, which contracts the generally decreasing atmospheric BC concentrations since 1989 (Ruppel et al., 2017, 2014)."

Page 3, Lines 75-87:

it is very important to define the goals of this work in the introduction. However, objectives and goals being are repeated several times. I suggest reducing the first part and focus on the bullet list, which is short and easy to understand.

Completely agree. The paragraph has been changed to the following:

"In this study, in situ measurements of atmospheric light-absorbing aerosol particles are used to explore how concentrations have changed, from 2002 to 2023. Concentrations of light-absorbing particles in the Arctic depend on a range of factors including emissions, meteorological conditions and atmospheric dynamics that promote certain transportation pathways, and also the effectiveness of various removal processes in the atmosphere, hence multiple factors within the context of sources, sinks, and transport will be analysed.

The main research questions that this work aims to tackle are as follows: (i) What are the long-term trends in the absorption coefficient (σ_{ap}) and how has the single scattering albedo (SSA) changed at the Zeppelin Observatory? (ii) What are the key factors controlling these trends (e.g. changes in scavenging, transport pathways and/or sources)? (iii) What is the influence of precipitation en route to the Zeppelin Observatory (iv) Are the calculated long-term trends in σ_{ap} influenced by extreme events (e.g. biomass burning events and high amounts of accumulated precipitation)?, (vi) Which source regions contribute most to observed σ_{ap} at Zeppelin Observatory, and how have their respective contributions changed over time?."

Page 4, Lines 94-95:

please the grammar of the phrase.

We agree. This part has been changed to the following:

"... Measurements are considered to be unaffected by emissions from the research village, due to temperature inversions below

500 m and the infrequency of northern wind flows; as a result, the observatory can be regarded as representative of regional background Arctic conditions (Dekhtyareva et al., 2018). ..."

Page 5, Lines 136-137:

please remove these statements. Focus on your study and not the history of Zeppelin, which is impressive but not of primary interest here.

We agree with the referee. The following sentences are now removed:

The TSI nephelometer has been used since May 1999. The Ecotech nephelometer was installed in April 2018 and has been operated continuously since (see Platt et al. (2022) for further details). and *Both nephelometers utilise the same principle to perform continuous measurements of the light scattering of particles;*

Page 6, Lines 161-162:

remove sentences, not usefull.

We disagree with the referee here. The collocation between the HYSPLIT model output and ERA5 data allows for a continuous set of data. Prior to this, the HYSPLIT output was composed of runs which were fed with FNL and GDAS meteorological fields, this meant that the time series generated was not continuous as there was a noticeable "jump" in the accumulated precipitation variable. The use of FNL meteorological fields led to an underestimation of the accumulated precipitation as compared with the use of GDAS. The following clarifying sentence is ended to the end:

"... As a result, the precipitation variable is a product of the spatial coordinates of the HYSPLIT model runs using FNL (i.e. 2002 - 2004) and GDAS (i.e. 2005 - present) and the surface precipitation data from ERA5. ... "

Page 10, Section 3.1:

You have only one subsection in section 3.1. no need for subsection here

We agree with the referee here and have removed the needless subsection

Page 10, Figure 1:

Although the text is clear, Figure 1 is hard to read and the trend described in the text cannot be observed in Figure 1. I suggest reducing the vertical scale range (even if it cuts the errors bars) and, maybe, split in three panels each of them showing the seasonal temporal series. There is a bit too much red, so that is hard to identify what you refers to.

We agree with the referee. We have modified the figure in regard to the comments of the referee. See the new figure, which now represents the trends for each season, in addition to the overall long-term trend. The subplots have been split into two as well, to be able to display the values that are not presented in the plot below. Also, the pre-whitened trend (3pw method) has been added along with the daily and seasonal trends.

Page 11, Line 275:

information on SSA is superficial here. I would remove this sentence.

We agree with the referee. We have removed this information

Page 11, Section 3.2:

No need for subsection here

We agree with the referee here and have removed the needless subsection

Page 17, Section 3.5:

No need for subsection here

We agree with the referee here and have removed the needless subsection

I stop here the specific comments, since the manuscript need a deep revision of its structure.

References

Acosta Navarro, J. C., Varma, V., Riipinen, I., Seland, Ø., Kirkevåg, A., Struthers, H., Iversen, T., Hansson, H.-C., and Ekman, A. M.: Amplification of Arctic warming by past air pollution reductions in Europe, Nature Geoscience, 9, 277–281, 2016.

AMAP: Impacts of short-lived climate forcers on Arctic climate, air quality, and human health, Arctic Monitoring and Assessment Programme (AMAP), 2021.

Bodhaine, B. A. and Dutton, E. G.: A long-term decrease in Arctic haze at Barrow, Alaska, Geophysical Research Letters, 20, 947–950, 1993.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., et al.: Bounding the role of black carbon in the climate system: A scientific assessment, Journal of geophysical research: Atmospheres, 118, 5380–5552, 2013.

Collaud Coen, M., Andrews, E., Alastuey, A., Arsov, T. P., Backman, J., Brem, B. T., Bukowiecki, N., Couret, C., Eleftheriadis, K., Flentje, H., et al.: Multidecadal trend analysis of in situ aerosol radiative properties around the world, Atmospheric Chemistry and Physics, 20, 8867–8908, 2020.

Dekhtyareva, A., Holmén, K., Maturilli, M., Hermansen, O., and Graversen, R.: Effect of seasonal mesoscale and microscale meteorological conditions in Ny-Ålesund on results of monitoring of long-range transported pollution, Polar research, 37, 1508 196, 2018.

Eleftheriadis, K., Vratolis, S., and Nyeki, S.: Aerosol black carbon in the European Arctic: measurements at Zeppelin station, Ny-Ålesund, Svalbard from 1998–2007, Geophysical Research Letters, 36, 2009.

Garrett, T. J., Brattström, S., Sharma, S., Worthy, D. E., and Novelli, P.: The role of scavenging in the seasonal transport of black carbon and sulfate to the Arctic, Geophysical Research Letters, 38, 2011.

Hansen, J. and Nazarenko, L.: Soot climate forcing via snow and ice albedos, Proceedings of the national academy of sciences, 101, 423–428, 2004.

Klimont, Z., Kupiainen, K., Heyes, C., Purohit, P., Cofala, J., Rafaj, P., Borken-Kleefeld, J., and Schöpp, W.: Global anthropogenic emissions of particulate matter including black carbon, Atmospheric Chemistry and Physics, 17, 8681–8723, 2017.

Liu, M. and Matsui, H.: Improved simulations of global black carbon distributions by modifying wet scavenging processes in convective and mixed-phase clouds, Journal of Geophysical Research: Atmospheres, 126, e2020JD033 890, 2021.

Platt, S. M., Hov, Ø., Berg, T., Breivik, K., Eckhardt, S., Eleftheriadis, K., Evangeliou, N., Fiebig, M., Fisher, R., Hansen, G., et al.: Atmospheric composition in the European Arctic and 30 years of the Zeppelin Observatory, Ny-Ålesund, Atmospheric Chemistry and Physics, 22, 3321–3369, 2022.

Ruppel, M., Isaksson, E., Ström, J., Beaudon, E., Svensson, J., Pedersen, C., and Korhola, A.: Increase in elemental carbon values between 1970 and 2004 observed in a 300-year ice core from Holtedahlfonna (Svalbard), Atmospheric Chemistry and Physics, 14, 11 447–11 460, 2014.

Ruppel, M. M., Soares, J., Gallet, J.-C., Isaksson, E., Martma, T., Svensson, J., Kohler, J., Pedersen, C. A., Manninen, S., Korhola, A., et al.: Do contemporary (1980–2015) emissions determine the elemental carbon deposition trend at Holtedahlfonna glacier, Svalbard?, Atmospheric Chemistry and Physics, 17, 12 779–12 795, 2017.

Schmale, J., Sharma, S., Decesari, S., Pernov, J., Massling, A., Hansson, H.-C., Von Salzen, K., Skov, H., Andrews, E., Quinn, P. K., et al.: Pan-Arctic seasonal cycles and long-term trends of aerosol properties from 10 observatories, Atmospheric Chemistry and Physics, 22, 3067–3096, 2022.

Sharma, S., Ishizawa, M., Chan, D., Lavoué, D., Andrews, E., Eleftheriadis, K., and Maksyutov, S.: 16-year simulation of Arctic black carbon: Transport, source contribution, and sensitivity analysis on deposition, Journal of Geophysical Research: Atmospheres, 118, 943–964, 2013.

Shaw, G. E.: Evidence for a central Eurasian source area of Arctic haze in Alaska, Nature, 299, 815–818, 1982.

Sinha, P., Kondo, Y., Koike, M., Ogren, J., Jefferson, A., Barrett, T., Sheesley, R., Ohata, S., Moteki, N., Coe, H., et al.: Evaluation of ground-based black carbon measurements by filter-based photometers at two Arctic sites, J. Geophys. Res., 122, 3544–3572, 2017.

Skiles, S. M., Flanner, M., Cook, J. M., Dumont, M., and Painter, T. H.: Radiative forcing by light-absorbing particles in snow, Nature Climate Change, 8, 964–971, 2018.

Stohl, A., Berg, T., Burkhart, J., FjÆaa, A., Forster, C., Herber, A., Hov, Ø., Lunder, C., McMillan, W., Oltmans, S., et al.: Arctic smoke–record high air pollution levels in the European Arctic due to agricultural fires in Eastern Europe in spring 2006, Atmospheric Chemistry and Physics, 7, 511–534, 2007.