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 are satisfactory. 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 2

1.1 General Comments

The authors investigated mechanisms for changes in the absorption coefficient (as a proxy for black carbon) at the Zeppelin Observatory, Svalbard over a twenty-year period. Out of three potential main mechanisms, they identified and quantified as the most dominant mechanisms a decline in the absorption coefficient based on precipitation and source emissions changes.

The paper is well in scope of ACP and presents an interesting, novel and long timeseries.

Overall, the manuscript is relatively well written, especially the introduction and the conclusion. The authors have convincing arguments for their hypothesis, although data is only presented as figures. However, there are a number of issues that are unclear and need to be addressed before the study should be published. The manuscript lacks a bit of clarity with regards to the methodology used, it is in parts unspecific and there is quite some redundancy in terms of information presented in the different chapters, as well as the Supplementary Material. Especially the conversion of the PSAP values from one wavelength to another with the assumption of an Absorbing Ångström Exponent of unity. That information is only presented in the Supplementary

Material, along with the remaining chapter on data harmonization, which is quite crucial to the study and should be in the method section of the main manuscript.

1.2 Main Manuscript

1.3 Specific comments (Numbers refer to Line numbers in the original manuscript PDFs)

Page 1, Lines 18-19:
How do these two sentences go together? Could the last sentence be clarified further?

We agree that it is a bit unclear what is meant here. We altered the text, by removing the last sentence. We wanted to express our finding that despite recent increases in the number of active fires increase, downwind of the Arctic, the long-term trends remained unaffected

The following line has been rewritten to express more clearly the meaning:
Previous statement: "Active fires have been shown to have a significant impact on the mean seasonal absorption coefficient, especially during northern hemispheric summer. However, no noticeable alteration in annual long-term trends can be observed."
New sentence: *"...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, Line 37:
"Significant" is not very specific. Can you put a number on that claim? FYI, chapters 7 onwards of the AMAP 2021 report have very recently been published. In chapter 8 they put the 2015 Total effective radiative forcing of Arctic BC at 0.96 ± 1.21 vs Global of 0.08 ± 1.08. Additionally, my personal preference would be to try to use this word in terms of "statistical significance" only.

We agree with the referee. We have included in the paragraph information from the Health AMAP Assessment 2021: Impacts of Short-lived Climate Forcers on Arctic Climate, Air Quality, and Human Health AMAP. The sentence now reads:
"... In the Arctic, the combined total radiative forcing from the interaction BC has with clouds, solar radiation and the surface albedo of the Earth, is significant, nonetheless, there is a great deal of uncertainty. The total forcing of BC for the Arctic is estimated to be 0.96 ± 1.21 Wm^{-2}, an estimation much greater than the global average BC forcing; this is a result of the impact deposited BC has on reducing albedo in snow and ice-covered regions (AMAP, 2021). ..."

Page 3, Line 73:

"why models overestimate observed BC (Sharma et al., 2013)" This is not universally true. Sharma et al. used only one specific model. If you look at Fig. 7.25 of AMAP 2021, you will see that the picture looks different for every model. And this is true for all SLCFs.

We agree and accept this comment and have altered the sentence to the following:

"...This lack of investigation could be one of the reasons why models fail to replicate observations of BC (AMAP, 2021)"

Page 3, Line 82:

"climatically relevant time period" I understand that 20 years of observational data of that kind is a big accomplishment, but in terms of climatically relevant data the (WMO, NASA, NOAA, etc) guidelines are usually 30 years.

We agree with the referee. 20 years (now 22 years) is not a sufficient amount of time to make the claim that this data set is of a length which is climatically relevant. This particular sentence has been removed and uses the phasing "almost climatic time scale" instead. The sentence now reads 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. ...

Page 4, Line 93:

What does "not so affected" mean? This is too unspecific.

We agree with the referee. Based on Platt et al. (2022) and (Dekhtyareva et al., 2018). The sentence now reads:

"... 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 4, Line 104:

"both slightly heated" Be more specific regarding the temperature of the whole-air inlet. This information is also not in Platt et al. 2022.

We agree with the referee. The sentence now reads:

"... The station itself and the whole-air inlet are both slightly heated to temperatures between 5-10°C to prevent freezing, meaning no additional drying of the aerosol is required as the relative humidity of sample air inside the station is kept below 30-40%. ..."

Page 4, Line 119:

"detection limits for a given sample rate." It is not clear to me what you mean with sample rate. Is this the flow? If so, could you give a LOD value for a typicall flow of 1 L/min? And what is the measurement frequency?

We agree with the referee. The word "rate" is changed to "flow". The sentence now reads:

"... The sample spot for the custom-made PSAP is much smaller than the commercial instrument (∼ 3.19 mm in diameter), which allows for lower detection limits for a given sample flow. ..."

Page 5, Line 139:

"regularly calibrated" What is the frequency of calibrations? and is the calibration process described somewhere?

The nephelometer is calibrated every month. The sentence now reads:

"... Nephelometers are calibrated once a month using CO_2 and particle-free air. ..."

Page 5, Line 148:

Could you give more details? Were the planes at different altitudes? Why 9 starting points?

More detail is added based on information from the HYSPLIT webpage/manual. The sentence now reads:

"... An ensemble of 27 back trajectories was initialised for every hour. The ensemble was generated by offsetting the meteorological fields in all possible combinations of x and y; a grid of three planes, consisting of nine starting points, where each plane is $\pm$ 0.1 sigma ($\sim$250 m) apart, was configured. ..."

Page 6, Line 166:

"as if the air parcel had travelled near the surface for the entire time period" Couldn't this effect your conclusions? What if the air parcel wasn't near the surface? What is the weighted average of parcels above or within the precipitate? Would in-cloud scavenging change your conclusions?

This sentence has been removed, instead a new paragraph has been written in the section about the trends in precipitation. We could have additionally focused on the relative humidity, and used the cloud base height (from ERA5), to get a better picture of whether the parcels are above or within the precipitate. However, we chose not to do this. The new paragraph attempts to tackle these issues and reads as follows:

"... Light-absorbing aerosol can be removed from the atmosphere via the process of wet deposition; there are two mechanisms by which these aerosol particles are removed: in-cloud scavenging and below-cloud scavenging (Pruppacher and Klett, 2010). In-cloud scavenging occurs when aerosol act as CCN, forming cloud droplets or when aerosol collide with preexisting droplets. Below-cloud scavenging describes the removal of ambient aerosol by falling hydrometers. Here, we focus on precipitation, and in particular, accumulated precipitation along the back trajectories, with the assumption that aerosol en route to ZEP, which have acted as CCN, are removed. For below-cloud scavenging, there is the chance that the impact of ATP is overestimated as aerosol particles can be above cloud height, however, ATP can also be underestimated if precipitation fails to reach the surface. This type of analysis places the focus on the removal of aerosol from the atmosphere, without emphasis on whether it is necessarily driven by in-cloud or below-cloud scavenging. Precipitation scavenging is assumed to act as a sink of aerosol."

Page 6, Line 168-170:

Please elaborate on why you made this choice. Your title says "black carbon" after all.

We agree. We have since changed the title to better represent the manuscript.

Page 6, Lines 174-175:

I think it would be useful to be more specific and mention which property of "aerosol loading" you mean and give a range of the values as well as the LOD in your text here.

We agree with the referee here. We have added more details and specific numbers concerning the aerosol number and mass concentrations measured at Zeppelin Observatory. The sentence now reads: *"...Aerosol number and mass concentrations are* *generally very low (50 - 250 cm^{-3}, 0.2 - 0.8μgm^{-3}), as compared with continental sites (Tunved et al., 2013)..."*

The following sentence was also removed as it was deemed unnecessary:

... In general, it is necessary for instruments to be well-calibrated due to the high noise-to-signal ratio...

Page 6, Line 177:

What are the consequences of low signal to noise in summer? Is it zero values or higher uncertainty, i.e. error bars? Move this information here.

The correction factor for the PSAP measurements displayed a seasonality, potentially as a result of the seasonal cycle of SSA and its influence on the reliability of the correction scheme by Bond et al. (1999).

Page 3 of Supplement, Lines 67-68 and Figure S2. (refers to the comment made on Line 191, Page 7 of the manuscript):

Regarding FigS2: It is not clear what EBAS is. From which instrument does this data come? The description in the SM is insufficient. Additionally, the sentence on line 67 does not make much sense: "Despite, the EBAS data set and the data processed from the manual PSAP"

The data comes from the EBAS database infrastructure operated by Norwegian Institute for Air Research (NILU) (https://ebas.nilu.no/). The data taken from EBAS was from the same custom-built PSAP instrument and should in theory correspond 1:1 to the values processed in this manuscript, however, there were slightly different approaches to the handling of the PSAP data, and as such the correction factors are not 1:1. The caption for Fig. S2 in the supplement has been altered to explain this and now the caption reads as follows: ... *"EBAS refers to the database infrastructure operated by Norwegian Institute for Air Research (NILU) (https://ebas.nilu.no/). The data taken from EBAS was the absorption coefficient from the same custom-built PSAP instrument. It should in theory correspond identically to the values processed in this manuscript, however, there were slightly different approaches to the handling of the PSAP data, and as such the correction factors are not 1:1." ...*

Additionally, in the supplement. The following sentence was re-written:

... "Despite the fact that both the EBAS data set and the data processed here in this manuscript are taken from the same instrument (i.e. the custom-built PSAP), the values are not identical (i.e. a CF of 1 for all time points). The reason for the scatter, as mentioned above as well, can be related to small differences in any of the variables that enter the calculations. The aforementioned correction factors are applied in the correct order to various time periods to produce the harmonised full-time series."

Page 7, Line 198:

Regarding FigS4: I don't see AE31 data in that plot (unlike the claim in the figure caption). The old PSAP data is off the chart. The caption of the figure could be slightly improved as well. E.g. description of lines, etc.

The figure has not been explained well enough. The correction factor (CF) in Fig S4 corresponds to the ratio of the hourly absorption coefficient arithmetic means from the various instruments and data sets used in this manuscript (i.e. EBAS database, automatic and manual PSAP, and the MAAP) and the absorption coefficient from the Aethalometer (AE31), all adjusted to the wavelength 637 nm.

We have also renamed the y-axis such that it is clearer and it now reads: "Correction factor needed for Aethalometer (λ adjusted to 637 nm) to reinstate 1:1 line [-]". The R^2 value between the two sets of instrument measurements is also added to the plot.

Page 9, Lines 248-249:

How does the lower time resolution affect uncertainties around the scavenging ratio? And do you know how does the normalization with CO influence conclusions regarding sources of different combustion types? Smouldering, more common in solid fuel combustion, usually leads to more CO as compared to more efficient combustion. This would lead to a bigger correction from e.g. biomass burning. Close to a source a CO/CO2 ratio (modified combustion efficiency) could be used to infere the type of combustion. I don't think for your receptor site, long distance from a source, this would still be true.

This part of the manuscript was completely removed, and the scavenging ratio no longer features in the analysis. The decision was made given the length of the manuscript.

Page 9, Line 252:

"fourth ved from version of the GFED" Did you also use small fires? The potential number of small boreal fires could make a difference.

This is a quote from the README file for the fourth version of the GFED. It follows:

... "This fourth version of the Global Fire missions Database (GFED4s) provides monthly burned area, fire carbon (C) and dry matter (DM) emissions, and the contribution of different fire types to these emissions in order to calculate trace gas and aerosol emissions using emission factors. All these datasets are based on burned area boosted by small fire burned area, hence the "s" in the GFED4.1s name." ...

All the datasets that we used in this study included small fires, hence "GFED.1s" files were used. *..."The "s" denotes that these emissions are based on both the standard GFED4 burned area (Giglio et al., 2013) AND burned area derived from active fire information seen outside the burned area maps, see Randerson et al. (2012)."...*

It goes on to say: *"...The small fire fraction indicates what fraction of total emissions stemmed from the small fire burned area. GFED4 emissions can be calculated by subtracting this fraction from total emissions, but we recommend using GFED4s emissions. Note that GFED4 burned area cannot be calculated this was for various reasons, lease use the original GFED4 burned area datasets for this. ..."*

Emailed James Randerson. For questions regarding small fire burned area or the conversion of monthly to daily / 3-hourly emissions, but no response.

Page 10, Line 265:

"negative trend in σ_{ap}. The long-term trend, based on seasonal medians, is approximately -0.004 (-0.0063 to -0.0016) Mm-1yr-1" For one, I wonder how this changes with an updated AAE (see my comments for the Supplementary Material). Secondly, could you please give consistent significant numbers and decimals? Additionally, please mention explicitly what the values in brackets mean.

The whole plot has now been redesigned the brackets which corresponded to the confidence interval for the slope, are no longer displayed. Instead, we now present all the various trends and their time resolution, from seasonal to daily. We also include the pre-whitened trend using the 3pw method by Collaud Coen et al. (2020b). The updated AAE we believe would not alter the trend significantly. Using 0.8 - 0.9 as an AAE value would lead to an adjustment of the PSAP absorption coefficients of between 3 - 4%. Thus, if anything would potentially increase the magnitude of the trend.

Page 1, Line 268:

"correcting for autocorrelation" Mention how you correct for autocorrelation.

In the section where we discuss the trends, we reference the work of (Collaud Coen et al., 2020b). In (Collaud Coen et al., 2020b), a three prewhitening method (3PW) is described and recommended for the correction of autocorrelation for purposes such as the long-term trend analysis of aerosol light absorption coefficient measurements. Here, below is what is mentioned: *For daily averages, when the data is more likely to be plagued by autocorrelation, the data is "pre-whitened" using the three prewhitening method (3PW) (Collaud Coen et al., 2020b). In keeping with similar studies (e.g. Collaud Coen et al. (2020a)), daily averages were computed with the requirement that at least 25% of the day consists of valid data (i.e. ∼6 hrs). For more details about TS see Sen (1968); for the MK test see Hirsch et al. (1982); Gilbert (1987). The MK tests performed in this study made use of the Python package mannkendall v1.0.0 (DOI:10.5281/zenodo.4134435 Collaud Coen et al., 2020b)"*

For clarity, the following sentence in the results & discussion section has been added such that it now reads:

"Overall, the last 22 years of observations (2002 - 2023) have displayed a statistically significant (s.s.) decreasing trend in σ_{ap}. The long-term trend, based on seasonal medians, is approximately -0.004 $Mm^{-1}yr^{-1}$, whilst the trend based on daily medians corresponds to a s.s. -0.002 $Mm^{-1}yr^{-1}$, the pre-whitened trend (i.e. correcting for autocorrelation) is -0.0006 $Mm^{-1}yr^{-1}$ (see Fig. 1). In relative terms (i.e. the ratio of the trend over the average), the rate of decline in σ_{ap} corrected for autocorrelation corresponds to - 0.82 % yr^{-1}. "

Page 10, Line 270-271:

"The full-time series is probably not best explained using a single trend," Isn't that quite normal for a long time series and exactly the reason why climate normal timeseries should be 30 years or longer?

True, but plenty of studies have reported long-term trends in aerosol optical properties. Collaud Coen et al. (2020a) present trends using 9 years of data. You are correct that the fact that full-time series are not best explained using a single trend is why for example NOAA state 30 years as a climatic-relevant time period to understand trends. In this manuscript, we are trying to show the complexity of the trend at hand i.e. that it seems to display two trends with a local minimum between 2016 and 2017. Overall, we feel the trend, despite not being 30 years, is worth mentioning. Many trends in aerosol optical properties have been reported on with less data.

Page 10, Line 272-273:

"For the period after 2016, only the AHZ displays a positive s.s. (p=0.003) trend of 0.01 Mm-1yr-1 based on daily medians." This partly contradicts the previous sentences. Could you rephrase this?

We disagree, this must be a misunderstanding there is no contradiction presented here. Whilst there is an overall decreasing long-term trend in the aerosol absorption coefficient from 2002 - 2023, there is also an increasing tendency from 2016 - 2023. The shorter trend within the longer term is not a contraction, it simply echoes the comment raised above that a single trend cannot represent the whole time series. Overall we see, a decreasing trend however the recent increase is strong enough to compensate for the longer decreasing trend. It is important to note that the reliability of the trend is dependent on the slope and the length of the time series, and given that we report on a time series which is only 8 years, we are cautious to define it as a trend and instead opt for the word "tendency".

Page 13, Lines 315-316:

"then only perturbed based on the respective changes to the frequency of each cluster," I don't understand this explanation. Each cluster has a different frequency. At some point you must have summed up the contribution of the different clusters if you calculated the "resepective changes ... to each cluster". This info is later repeated in the discussion but doesn't make it clearer.

We agree. It is not the easiest to explain. Essentially, the same method as described by Hirdman et al. (2010) is utilised in this manuscript. The respective long-term trends for occurrences of each cluster are used to model the evolution of the absorption coefficient, such that NA increases at a rate of 3.4%yr^{-1}, G increases at 1.3%yr^{-1}, AO at -2.2%yr^{-1}, S at -5.6%yr^{-1} and E at 3.0%yr^{-1}. The initial absorption coefficient values are stated in the manuscript. The initial values are the arithmetic average of the first 3 years i.e. 2002 - 2004, and are as follows:

- $\sigma_{\mathrm{ap}}^{\overline{\mathrm{NA}}} = 0.21\,\mathrm{Mm}^{-1}$,
- $\sigma_{\mathrm{ap}}^{\overline{\mathrm{G}}} = 0.22\,\mathrm{Mm}^{-1}$,
- $\sigma_{\mathrm{ap}}^{\overline{\mathrm{AO}}} = 0.21\,\mathrm{Mm}^{-1}$,
- $\sigma_{\mathrm{ap}}^{\overline{\mathrm{S}}}, = 0.42\,\mathrm{Mm}^{-1}$,
- $\sigma_{\mathrm{ap}}^{\overline{\mathrm{E}}} = 0.69\,\mathrm{Mm}^{-1}$).

The changes in terms of the frequency occurrence for each cluster follow the linear trend (y = mx + c), where y is the normalised frequency occurrence and c is the initial frequency at the start of the data set. Using the equation of the trend line in Fig. 4 we can calculate the occurrence of each cluster at the end of the time series as follows:

- NA: 0.0034x22 + 0.0790 = 0.1538 at the end of the time series,
- G: 0.0013x22 + 0.1083 = 0.1369 at the end of the time series
- AO: -0.0022x22+0.2450 = 0.1966
- S: -0.0056x22+0.4154 = 0.2922
- E: 0.0030x22+0.1523 =0.2183

The changes to the normalised occurrences are presented in the new figure 5 a) that the clusters have changed in their occurrences for NA from 8% to 15%, G from 11% to 14%, AO from 25% to 20%, S from 42% to 29% and finally E from 15% to

22%.

We fixed the absorption coefficient for all the clusters (using the fixed values mentioned above, based on the mean of the first 3 years). What changes is the occurrence of each cluster, as defined by the trend in the occurrences. For example, using the trend in occurrence and the fixed starting value we can estimate the initial combined absorption coefficient = 0.0790*0.21 + 0.1083x0.22 + 0.2450x0.21 + 0.4154x0.42 + 0.1523x0.69 = 0.01659+0.023826+0.05145+0.174468+0.105087 = 0.371421 $\mathrm{Mm^{-1}}$. A very similar value to the initial value for the trend in the arithmetic means i.e. 0.3967 $\mathrm{Mm^{-1}}$. Using this implied method we can observe which clusters contribute the most, this is dependent on not only the average concentration but also the occurrence.

So the final absorption coefficient is equal to 0.1538x0.21, 0.1369x0.22 + 0.1966x0.21 + 0.2922x0.42 + 0.2183x0.69 = 0.032298 + 0.030118 + 0.041286 + 0.122724 + 0.150627 = 0.377053 $\mathrm{Mm^{-1}}$. Thus, we make the argument here that just changes in the frequency of occurences is not a sufficient explanation for the trend.

In terms the amount/proportion of the absorption coefficient the changes are as follows for the individual clusters:

- NA: 0.01659 $\mathrm{Mm^{-1}}$ -> 0.032298 $\mathrm{Mm^{-1}}$
- G: 0.023826 $\mathrm{Mm^{-1}}$ -> 0.030118 $\mathrm{Mm^{-1}}$
- AO: 0.05145 $\mathrm{Mm^{-1}}$ -> 0.041286 $\mathrm{Mm^{-1}}$
- S: 0.174468 $\mathrm{Mm^{-1}}$ -> 0.122724 $\mathrm{Mm^{-1}}$
- E: 0.105087 $\mathrm{Mm^{-1}}$ -> 0.150627 $\mathrm{Mm^{-1}}$

From this analysis we can see that the contribution from NA has increased, E has increased too and G has also increased. AO'scontribution has decreased and so too has Siberia's.

We argue that the increase from the Eurasian cluster (i.e.polluted cluster), is a factor why the trend cannot be explained by the changing frequency occurrences, even though NA has increased as well.

Page 15, Line 349:

"9 years of data, 2002 - 2010," What is the reason that you could not use the full twenty year data?

Yes, this period is a bit arbitrary. The idea was to simply take a subsample of the data, so as to not base the "empirical" ATP-σ_{ap} on the full time series. The subsample was thought of as a "training" data set. In the latest revised manuscript, we have adjusted the methodology. Now, we take random subsets/fractions of the data and run the analysis 50 times for each set of random data points. This has made the methodology more robust and has allowed us not to base the ATP-σ_{ap} simply on the first 9 years when the absorption coefficient exhibits relatively high values during the Arctic Haze seasons.

Page 16, Line 350:

"back trajectories" Are these the same 10 day BTs used for your previous analysis? And is this the ERA5 precipitation data? I think it would be good if there would be a bit more information in this section or in the Methods section.

Yes, it is all the same back trajectories. This whole section has now been rewritten. Also, since the last submission, all the back trajectories have actually been rerun to ensure the same settings were used for all years. Hopefully, the text is now satisfactory. The text now reads as follows:

"To gauge the extent to which the aforementioned increasing trend in accumulated back trajectory precipitation (ATP) can influence σ_{ap}, the trend in σ_{ap} was recreated. The variable $\sigma_{\mathrm{ap,cal.}}$, denoting a calculated σ_{ap} as opposed to an observed σ_{ap}, was produced. $\sigma_{\mathrm{ap,cal.}}$ uses the dependency between σ_{ap} and ATP, to map the ATP variables onto $\sigma_{\mathrm{ap,cal.}}$ values. The dependency between σ_{ap} and ATP, reflects the influence of precipitation scavenging and was analysed for all three seasons (AHZ, SUM and SBU) and all five source regions (NA, G, AO, S and E) such that 15 empirical relationships were ascertained (see Fig. S17). Essentially, cluster and season-specific relationships for σ_{ap} and ATP were utilised to try and simulate the long-term trend in σ_{ap}; the relation between ATP and the median σ_{ap} was used on a seasonal basis to account for different precipitation regimes. For each cluster and for each season, every 1 or 2 mm of ATP was compared to its respective median σ_{ap} value; linear interpolation was used to extend the resolution beyond 1 or 2 mm. The relationship between ATP and σ_{ap} was developed using random samples of different fractions of the data set. The amount of data used (i.e. $\leq$ 50% of the data) is a compromise between having enough data points for a robust analysis of the relationship, and not using all of the data in the development of the parameterisation. For the majority of transportation clusters and seasons, σ_{ap} decreases as ATP increases, however, the extent of the decline in σ_{ap} as ATP increases varies depending on the cluster and season (see Fig. S17). It should be noted that a key assumption in these calculations is that σ_{ap} is solely dependent on ATP and that there are no additional factors."

Page 16, Line 351-352:

“For the majority of transportation clusters and seasons, there is an exponential decline in σ_{ap} as ATP increases.” I don’t know. This seems like a stretch. I can see a clear exponential decline in 4 of your subplots. Some might have initially an exponential decline but the y-scale such that it appears quasi-linear. In clusters 4 and 5 there is even an increase at higher ATP.

Yes, for certain seasons and clusters there is no such exponential curve, however, in the majority of relationships, there is a negative correlation between the absorption coefficient and the accumulated back trajectory precipitation (ATP). Attempts were made to fit an exponential curve to the medians in Figure S17 however, the majority of fits simply resemble a straight line. We believe that with enough data and for all seasons and clusters, the relationship is exponential. It can argued that an exponential curve can be fitted to all the plots and that it may not be clear given the magnitude of the observation, however, this misses the main point we want to convey which is that ATP adversely impacts the absorption coefficient, and the relationships depend on season and region. Hence we have rewritten the sentence and it now reads as follows:

For the majority of transportation clusters and seasons, σ_{ap} decreases as ATP increases, however, the extent of the decline in σ_{ap} as ATP increases varies depending on the cluster and season (see Fig. S17)

Page 16, Line 357:

“The hourly ATP averages are converted to their respective σ_{ap} values using the closest corresponding value for each σ_{apATP} relation” It is not cristal clear what you did here. The ATP data is hourly, but what time resolution does the σ_{ap} have?

Figure S17 displays the relationship between σ_{ap} and the accumulated back trajectory precipitation (ATP). The curve plotted in figure S17 is then used to create a mapping from ATP to σ_{ap}. This mapping is essentially the linear interpolation displayed in the example given in figure S17. The resolution of the ATP data set is hourly given the HYSPLIT model is set such an ensemble of back trajectories arrive every hour. The ATP hourly values are mapped onto hourly so-called calculated absorption coefficient values, using the aforementioned mapping.

Page 15 of the supplement, Line 358:

"see Fig. S17" The time resolution is not visible in this figure.

Figure S17 uses data which is of an hourly resolution. This information has been inserted into the caption of the figure such that it reads as follows: *"...The data for each subplot is composed of temporally collocated hourly averages of ATP and σ_{ap}."*

Page 12, Line 363:

"Section 11 in the supplement" That "section" is literally one sentence and not very informing.

Section 11 attempts to describe the seasonality of the absorption coefficient, the precipitation along the back trajectory, and the emission inventories (transported to ZEP). The idea is to show that the emissions do not display the same seasonality as the absorption coefficient. The figure was improved and a better description was added such that the caption reads as follows: *"Seasonality plots for the absorption coefficient, σ_{ap}, (black), the accumulated back trajectory precipitation (ATP) (blue) and the accumulated emission of BC on the back trajectories, based on emission inventories from ECLIPSE (yellow). The absorption coefficient and ATP show monthly median values with the error bars displaying the 25$^{\mathrm{th}}$ and 75$^{\mathrm{th}}$ percentiles."*

Page 16, Line 364:

"calculated seasonal medians of σ_{ap} is approximately -0.001 Mm-1yr-1" What is the uncertainty in this number? This trend is so tiny, that it might only be clearly visible on a log scale.

Yes, the trend in the σ_{ap} is tiny, and even more so once the time series is corrected for autocorrelation. However, it should be noted that the trend is over 22 years of data and $-0.82\%\mathrm{yr}^{-1}$, leads to a significant decline overall. For the trend analysis based on the calculated absorption coefficient the trends are also tiny, however, still represent approximately a quarter of the long-term trend in the in situ observations for the absorption coefficient.

Page 16, Line 368-369:

"The back trajectories arriving at ZEP have experienced an increasing influence from forest fires. The number of active forest fires each back trajectory traversed over has increased since 2002, with the most notable shift occurring after 2015" Can you back this up with data or a reference?

Figure 7 has now been completely removed and replaced with a table. The figure did a poor job of displaying the trend in the number of active fires influencing the back trajectories.

Page 17, Line 373:

“not shown here” Why not? You come back to it in the Discussion and it seems relevant to your argument.

For this, we changed the sentence such that it reflects the literature and case studies that have come before. We wrote the following: *"... There are several examples of high σ_{ap} concentration episodes, observed at Arctic sites, caused by emissions from fires (e.g. Stohl et al., 2007), hence it is well-known that large biomass burning (BB) events are connected to increased σ_{ap} values.*

Page 17, Line “Figure 7.”

What happened in 2013? The year without a summer.

It is collocated data. There is a gap in the absorption coefficient data, hence there is now a gap in this data set.

Page 17, Line “Figure 7.”

“Note that the Arctic Haze season (AHZ) of 2006 is significantly smaller compared with the MODIS fire count” Is this data now only MODIS, or also from other data? And did you remove exreme biomass burning events or not? It’s not clear to me.

Figure 7 has now been completely removed and replaced with a table. The figure did a poor job of displaying the trend in the number of active fires influencing the back trajectories. The idea with figure 7 was to demonstrate that the number of fires that the back trajectories traversed over has increased, and that consequentially the accumulated BC emissions influencing the 22 years of air mass history has also increased.

Page 17, Line Figure 7.

“Note that the y-axis is displayed on a log scale” Even on a log scale, the trend is barely visible.

Page 17, Line 377-378:

“Removing the data points which correspond to the most extreme number of active forest fires (i.e. defined as the 99th percentile of a running 15-day average of the BC emissions from GFED), reduces the seasonal means, however, has a negligible effect on the seasonal medians.” Without showing us the data there is little meaning in these words. Does the figure show data where most extremes have been removed or not? How much are the means reduced? How much is negligible?

This statement now has a corresponding figure. See Fig. S16 in the supplement. A threshold for every “Day Of the Year” (DOY) was created using all the data from 2002 - 2023. The threshold corresponds to the rolling 99$^{\mathrm{th}}$ percentile for the BC emissions from the GFED. Any value above this defined threshold for the individual DOYs is deemed to be an extreme value. The figure essentially removes the date times that correspond to these extreme values. Figure S16 shows the impact removing these extreme data points can have on the arithmetic seasonal means. There is minimal, if no impact, on the seasonal medians due to them being more robust to extreme values.

Due to lack of space, we only simply refer to the trend in the data set with the extreme B.B. values removed and no do show a plot, as there is little to show.

Page 18, Line 387:

“increasing trend during the Arctic Haze season of 0.001 Mm-1yr-1” Again, this reads like a contradiction. You have discovered a counter trend in a longer trend. If you would look at even shorter periods, you would find trends in every direction you like. But when is a trend really a trend?

We agree that you cannot just arbitrarily choose to start and stop trends, and then decide there is a counter-trend within longer trends. However, we still believe that is important to address the possible causes of the increasing values for the absorption coefficient after 2016-2017. We simply split the time series in two based on the fact that there is a turning point, this seems to us to be a fair justification for looking at the data after 2016 separately.

Page 18, Line 389-390:

“we can conclude that the amount of scattering in relation to the absorption has increased” I’ve now read this several times, but am not entirely sure what you are trying to communicate.

We have removed this sentence. It simply refers to the long-term trend in the single scattering albedo (SSA). SSA increases from 0.9 to almost unity in this time series. We added the following sentence, which we hope makes more sense:

"During this period of time, the increase in the SSA is a combination of a long-term increase in σ_{sp} *(Heslin-Rees et al., 2020) in conjunction with a decrease in* σ_{ap} *(as presented here)."*

Page 18, Line 390 “Fig. S3”

All I can see in this Fig is a decrease and then stagnation in σ_{ap}.

This sentence has been changed and the reference to Fig. S3 is removed. The sentence now appears as follows: *"... The single scattering albedo has experienced an increase in the last 21 years from 0.9 to close to 1 (see Fig. S9 in the supplement). During this period of time, SSA has increased because of a long-term increase in* σ_{sp} *in conjunction with a decrease in* σ_{ap}*. ..."*

Page 19, Line 415-416:

“The increase in this region southeast of Europe could also be the result of increases in contributions from sources further south” It is not clear what you mean here. You are still speaking about the emission inventory. The increase of emissions in one area does not influence another area. Emissions are “grid-bound”.

We are not discussing emission inventories here. We agree that the emission inventory trends are grid-bound and that emissions in one area do not affect emissions in another area. However, here we are referring to the CWT plots, hence, it’s referring to a combination of in-situ observations and back trajectories. Emissions from regions further south can be misclassified as coming from regions slighter upwind, given the limitations of the length of the back trajectory model (i.e. 10 days). This sentence has been changed and the reference to Fig. 2 is made clearer. The sentence now appears as follows:

"... In regards, to the CWT trends in Figure 2, another possible explanation for the increase displayed in the region southeast of Europe could be changes to the contributions from sources further south e.g. emissions the Indo-Gangetic plane over Central Asia and into the high Arctic (Backman et al., 2014). ..."

Page 19, Line 426:

“has been explained” Please point us to peer-reviewed publications where this is the case.

A reference was added to this sentence. We have toned down the messaging and simply written that Schmale et al. (2022) suggests that this could be an explanation for the lack of a biomass-burning signature on the long-term trends. The sentence now appears as follows:

"... Schmale et al. (2022) suggested that another possible explanation for the lack of a BB signature on the long-term σ_{ap}/eBC trends could be related to the height at which wildfire plumes are injected; BC from BB events could be emitted into the atmosphere further aloft, and thus arrive to ZEP at higher altitudes, while still slowly filling the Arctic domain with biomass burning aerosol...."

Page 19, Line 427:

“further aloft” I don’t think this is an entirely valid point. Do you mean chimneys? There are a lot of different types of wildfires, and they are not all crown fires, unless we get massive fires like currently in Canada. There is also a lot of burning shrub and peat, very close to the ground.

We refer to it in terms of the injection height of the fires. It is referred to in the following paragraph only as a possible explanation.

Page 19, Line 427-428:

“In this study, it was shown that the potential influence from BB events has increased (see Figs. 7)” Again, I think you don not have very strong evidence. It is a hypothesis of yours. Although a reasonable one. But your evidence does not fully prove your claim.

We disagree. The number of fires downwind of ZEP with the possibility of being transported up to ZEP has significantly increased since 2015. We have replaced Figure 7 with a table to make this point more clearly.

Page 19, Line 433:

“report the trends using seasonal medians” Why does the axis of Fig 7 say “mean”?

There is a misunderstanding here, as we have not properly explained it here. The sentence “Here, though, we report the trends using seasonal medians and as a result, extreme events such as BB events do not possess the ability to alter the magnitude or sign significantly.” is referring to the long-term trends in the seasonal median σ_{ap}. We use the mean GFED emissions of BC to ascertain the extreme biomass burning events. We then use these events and remove these periods from the σ_{ap} data set.

Page 20-21, Line 482-489:

I specifically liked this part. It shows a fair and balanced view of all the involved issues.

Okay great. We made sure to keep it in

Page 21, Line 496:

"decline" Looks more like stagnation to me.

Whether or not it is a decline is heavily dependent on the length of the time series. Given that there are not so many years to analyse this stagnation, the robustness of the trend analysis which follows is not as strong. .

1.4 Technical corrections

Page 1, Line 2:

Add "online" or "in-situ" to filter-based

We agree and have added the word sayonline

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

"... Here, BC is studied indirectly using online filter-based methods to ascertain aerosol light absorption parameters. ... "

Page 1, Line 8:

Does the haze season increase in time or magnitude?

We agree the statement is ambiguous, it has been made clearer and reads as follows:

" ...the last 8 years, 2016 - 2023, showed a slight increase in the magnitude of the light absorption coefficient for the Arctic haze season period..."

Page 2, Line 22:

has undergone - is undergoing

We agree and have made the corrections as follows: *" ... The Arctic region is undergoing increased rates of warming compared to the global average ... "*

Page 2, Line 44:

"major source of BC" You might want to cite (Stohl et al., 2013) here

We agree and have made the following correction. It now reads as follows:

"... Russian gas flaring is considered another potential major source of BC, however, direct measurement of its respective contribution is lacking (Winiger et al., 2019; Stohl et al., 2013) ...

Page 3, Line 57:
There is a question mark and your two Sinha ea 2017 references are the same paper. It is a nice study but only covers one site. Here, I think AMAP 2021 would be a good (additional) reference.

Apologies, we wanted to reference Eleftheriadis et al. (2009). Thank you for the recommendation of AMAP (2021) as well. It now reads as follows: *"... The techniques required need to be particularly sensitive, due to the low aerosol loadings. (i.e. approximately in the tens of ng/m-3) (Sinha et al., 2017a; Eleftheriadis et al., 2009; AMAP, 2021). ..."*

Page 3, Line 73-74:
"pronounced difference between trends in atmospheric and BC ice core measurements" I think this is a very valid point and it would make it more clear if you could specify the time horizon you are referring to.

We have added more details to this statement. It now reads as follows:
"... This lack of investigation into the role scavenging can have on aerosol concentrations could be one of the reasons why models fail to replicate observations of BC (AMAP, 2021). 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 6, Line 173:
mention that Eq 2 is in the SM

Done. It now reads as follows:
σ_{ap} *measurements from instruments with different operational wavelengths were adjusted to* λ*=637 nm (see eq. 2 in the supplement).*

Page 7, Line 206:
"number of active numbers" I assume this should be active fires? Yep, this has been changed such that it now makes sense and reads as follows:
"... number of active fires... "

Page 10, Line 270:
"3PW" I think it would be easier to understand/read if you just write it out. You only use this abbreviation once, besides it's introduction.

We agree. We have included the full term (not abbreviated) above in the sentence on Line 268.

Page 11, Line 276: Regarding FigS8

“decrease in σ_{ap}.” A decrease (or any trend) of this value is really difficult to see in the figure. Could you add trend lines?

The long-term trend in the absorption coefficient is already presented. The long-term trend in the scattering coefficient is detailed in other studies Heslin-Rees et al. (2020). We just wanted to show that SSA is increasing. The point of figure S8 is not to display the decreasing trend in the absorption coefficient.

Page 11, Line 284:

“, the trend is calculated for each and every grid cell on an annual resolution” Are all these trends s.s. ?

We have marked the grid cells that represent a statistically significant trend by presenting their trend. The non-s.s. grid cells just display a grey box, as opposed to blue or red signifying their sign.

Page 15, Line 335:

“quite some” Please be more specific.

We agree. We have added more explanation to the extreme value analysis we carried out.

Page 15, Line 338:

“Short-term” Define what this means.

The mention of the scavenging ratio has been removed in order to shorten the text. However, “short-term” refereed to 2 weeks.

Page 16, Line 353:

“decline” doesn’t hurt to write that you mean decline in σ_{ap}, to be more clear.

Agreed. This sentence has been altered and now reads: *"... For the majority of transportation clusters and seasons, σ_{ap} decreases as ATP increases, however, the extent of the decline in σ_{ap} as ATP increases varies depending on the cluster and season (see Fig. S17). ..."*

Page 16, Line 355:

“exhibit the lowest amounts of wet removal strengths;”) Shouldn’t this be the highest amount of wet removal strength?

We agree. We have removed this sentence as it does not add much. We are concerned about the length of the paper.

Page 16, Line 360:

"The proportion of the trend is approximately the same" That is not very specific information. Almost useless.

This sentence has been removed. It was considered unnecessary. The sentence refers to the fact that the analysis mentioned above can be performed either by calculating the trend on a monthly or seasonal resolution. Thus, we get the same result independent of the temporal resolution chosen. However, since then we have added the 3pw method which accounts for auto-correlation.

Page 16, Line 363:

"Fig. 7" change to Fig 6

Yep. Very true. We have now replaced it with a new figure and labelled it correctly.

Page 17, Line 375:

"it is clear" Not really it isn't.

We have replaced the figure with a table. The table now shows that the averages have increased.

Page 17, Line 375:

"potentially influential" On what? Be more specific (BMS).

We use the word "potentially" because we are unsure whether the emissions from the active fires will be measured at ZEP (i.e. there may be fires downwind, but do the emissions reach ZEP). Possible reasons could be that the emissions are too high in altitude by the time they arrive, the BC is scavenged and hence never makes it to ZEP, or alternatively, the HYSPLIT model is simply incorrect in its prediction of the source region. There may of course be other explanations.

We have changed the statement to the following:

"The air masses arriving at ZEP have experienced an increasing influence from active forest fires during the study period (see Table 1). The average number of active forest fires each back trajectory traversed over has increased since 2002, with the most notable shift occurring after 2015. The same can be said for biomass burning BC emissions, based on the GFED. Stathopoulos et al. (2021) observed from their analysis of aerosol light-absorbing carbon from 2001-2015, that emission rates from open fires increased by factors of between 3.4 - 3.6 for the warm period (May-October) for the years 2006 - 2010 and 2011 - 2015 relative to 2001 - 2005."

Page 17, Line 379:

"no impact on the long-term trend" Of what? BMS

The point we wanted to address here is that the influence of biomass burning events, despite their increase downwind of ZEP, has not impacted the long-term trends. We have since added more explanation to this point.

Page 18, Line 383:

"distinctly lower values" Please remind us again from where to where the values went. It's really hard to read any numbers of the chart of figure 1, i.e. where was the LMS value in 2003 vs 2022?

This part of the text has been rewritten and the discussion is now part of the results section. This part is now part of the following paragraph:
"... Overall, the last 22 years of observations (2002 - 2023) have displayed a statistically significant decreasing trend in σ_{ap}*. The long-term trend, based on seasonal medians, is approximately -0.004* $Mm^{-1}yr^{-1}$*, whilst the trend based on daily medians corresponds to a s.s. -0.002* $Mm^{-1}yr^{-1}$*, the pre-whitened trend (i.e. correcting for autocorrelation) is -0.0006* $Mm^{-1}yr^{-1}$ *(see Fig. 1). ..."*

Page 18, Line 385:
“anthropogenic influence” I guess you could call a decrease in haze and other pollution, partly due to in increase in wet scavenging, anthropogenic influence, since we influence everything. I just think it reads funny

We acknowledge this and now we have rewritten this section and added it to the results section. This sentence is now part of the following, where we have basically exchanged “anthropogenic” with “the influence of long-range airpollution”:
"... The fact that all seasons display negative trends is suggestive of the fact that the Arctic Haze is not the only season which experiences the influence of long-range air pollution. ..."

Page 19, Line 426:
“a signature” what exactly do you mean with this? BMS

Signature refers to the influence of biomass burning events on the long-term trends. The sentence now appears as follows:
"... Schmale et al. (2022) suggested that another possible explanation for the lack of a BB signature on the long-term σ_{ap}*/eBC trends could be related to the height at which wildfire plumes are injected; ..."*

Page 19, Line 431:

“just 3% of the data” By time or CWT or what measure? BMS Good point this is not clear However, it means that 3% of the hourly data points have been removed.

Page 19, Line 434:
“when” did you mean to write "if" or "why"?

Again, because the discussion section is added to the results section. Most of the paragraph was rewritten. We decided not to include this sentence, given the need to reduce the number of pages. The point we wanted to make is in which cases do you end with high absorption coefficients at ZEP after high emissions of BC from active fires downwind, and in which cases is this, not the case? Whether we use “when”, “if” or “why” I think we achieve similar ideas.

Page 19, Line 436:
“potential” prossibility?

We use the word possibility because it must be stressed that the increased influence from forest fires does not include changes in the other processes mainly removal processes. The number of biomass burning events and the emissions that they entail could have increased in the time, but whether the sources and the transport are relevant for ZEP is another question.

Page 21, Line 497:
“most recent years” BMS

This sentence was rewritten and reference to the scavenging ratio has been removed.

Page 21, Line 504: “there has been a shift in the sign of the trend.” When? BMS

"... It is interesting to note that reduction in σ_{ap} has not been consistent, and in fact we report an AHZ minimum of $\sim 0.02\,\mathrm{Mm^{-1}}$ in σ_{ap} between 2016-2017. Thus to aid in the interpretation we can divide the time series into two periods: 2002 - 2016 and 2016 - 2023. The more recent period, 2016 - 2023, is characterised by an increasing tendency during the Arctic Haze season of $0.004\,\mathrm{Mm^{-1}yr^{-1}}$. This shift in sign and the general reduction in the rate of decrease has been referred to as a “stagnation” (Schmale et al., 2022). ..."

Page 21, Line 505: “collocated” How do you collocate a property that has a fixed place with something that is moving?

The aerosol properties were temporally collocated with the arrival time of the back trajectories. The text has therefore been altered and reads as follows: *"... The aerosol properties were temporally collocated with the arrival time of the back trajectories to understand which atmospheric parameters were potential factors in influencing averages..."*

1.5 Supplementary Information

1.6 General Comments

The figure captions are really insufficient and many figures are poorly described.

We agree. We have added to the captions and the explanations of the figures.

1.7 Specific comments

Page 2, Line 35: “For periods in which nephelometer measurements were invalid or not present,” (p. 2) Show this in Fig S1

The periods where the DMPS data was used has been added to Fig. S1.

Page 3 Line 49: “σ_{ap} is acquired from EBAS” (p. 3) based on which instruments?

An additional sentence has been added to explain where the data from EBAS came from. The data from EBAS is based off of the same instrument i.e. the manual PSAP. The next sentence reads as follows:

The σ_{ap} from EBAS is from the manual PSAP, however, slightly different post-processing has been performed.

Page 3, Lines 56-57: “Only values greater than 0 Mm-1 were considered valid. This is despite the stated detection limit by the manufacturers, for a 30-minute time resolution, being approximately <0.13 Mm-1” (p. 3) Why would you do this? At least explain your rational.

Good question! Using $0\,\mathrm{Mm}^{-1}$ as the minimum value is always a compromise between having enough data and being reliable. We point the reviwer’s attention to the value $0.012\,\mathrm{Mm}^{-1}$ as opposed to $0.13\,\mathrm{Mm}^{-1}$ for the MAAP detection limit (Asmi et al., 2021). Hence the reduction with this value is fairly good. We could have used $0.012\,\mathrm{Mm}^{-1}$ and applied it to the full data set (after re-sampling to hourly means) including the PSAP data, however, this would have removed useful and reliable PSAP data. Instead, we decided to impose a value of $0\,\mathrm{Mm}^{-1}$ which meant including some potentially unreliable MAAP data but including all the PSAP data.

75-76: “assuming an Absorbing Ångström Exponent (ÅAE) of 1 (i.e. pure EC) (see Eq.2). Examining the ÅAE from the Aethalometer data this was considered a fair assumption.” (p. 3) (Liu et al., 2018) have shown that an AAE of 1 is not an appropriate assumption. You should either back up your claim that "Examining the ÅAE from the Aethalometer data this was considered a fair assumption." by showing us the data and statistics, or consider using size distribution data (SP2) to find the geometric mean diameter (GMD) and an appropriate AAE, e.g. analog to (Tunved et al., 2021)

Good question! We looked into the AE31 data. We calculated the Ångström Absorption Exponent (AAE) using all wavelengths. When we consider all aethalometer wavelengths there is a need for a loading correction which may increase slightly the AAE. We used the AAE from 2005 to 2020 to look at the changes. The AAE was always lower than 1 and by our calculations around 0.8. The trend itself is interesting, but given variability and uncertainties, it will not affect your wavelength harmonisation to any great extent. Using the estimated Ångström exponent of ca 0.8-0.85 instead of the commonly used factor 1, will increase the absorption coefficient (at $\lambda = 637\,\mathrm{nm}$) by 3 to 4%, after the the conversion from 525 nm (i.e. PSAP wavelength).

FigureS4 caption: “Aethalometer (A31) data” (p. 6) Where is this data?

There is a misunderstanding here, and the caption did not explain well how the Aethalometer (AE31) data was utilised. The data from the instruments is compared to the Aethalometer (AE31), the CF stands for the correction factor - it is a comparison between the two data sets. Because of the confusion we have relabelled the yaxis

92: “zeroing mode” (p. 7) Can you explain this for people not familiar with the TSI?

Apologies for using technical language and not explaining it. The text in the supplement now reads as follows: *"The problems with the TSI were likely caused by the instrument being stuck in the zeroing mode, which is when the light scattered by the carrier gas, the instrument walls and the background noise in the detector is measured."*

FigureS7 caption: “at least 5 year’s worth of long-term data is required to perform any reliable trend analysis” (p. 8) Says who? Can you give a reference and could you also give the absolute trend?

I agree. There was no reason for this limit. For example, Collaud Coen et al. (2020a), only considers “stations with at least 10 years of measurements” in order to compute long-term trends for absorption and scattering coefficients. In the revised manuscript we have softened this wording and instead of talking about “trends” for such short time periods i.e. just 8 years, we now speak of a tendency.

“Figure S14.” (p. 12) caption missing

Yep, very true. Furthermore, the trend line has been removed from the figure as it was rather needless.

1.8 Technical corrections:

Page 3, Line 51 in the supplement: “1000 l” (p. 3) In this font the "1" looks like the "l". I suggest to use capital L, always.

Fair. We agree and have changed “l” to “L”.

Page 3, 88: “need to: reference as to why the TSI is better)” (p. 7) Please enlighten us.

We simply used TSI as the inference instrument due to its increased sensitivity, compared to the Ecotech. Also, the TSI has been used much more extensively at ZEP. However, there is no strong reasoning behind such a decision. We just needed to choose one of the nephelometers. The sentence now reads as follows: *"... For harmonisation, the TSI was used as the reference instrument. Using an instrument-specific correction factor, the σ_{sp} values of the Ecotech were adjusted to match the TSI. ..."*

Page 7, Line 91 in the supplement: “additional problems” (p. 7) Additional to what exactly?

We agree. The sentence is not so clear. The additional problem with the nephelometer was that the TSI was believed to be stuck in the zeroing mode (measuring carrier gas, as opposed to ambient air). The sentence now reads as follows: *"...It should be noted that there were additional problems with the TSI from 2016 onwards. The problems with the TSI were likely caused by the instrument being stuck in the zeroing mode, which is when the light scattered by the carrier gas, the instrument* *walls and the background noise in the detector is measured. The problem was dealt with by taking only the measurements where the instrument was considered not to be in the zeroing operation mode. This procedure was able to be performed and the data were cleaned as it was found that these normal modes of operation had a recurring pattern."*

Page 8, Figure S6 caption: “red” (p. 8) blue?

Yes, well spotted. We have corrected this mistake

Page 8, Figure S6 caption: “The number of active fires at any given time has been calculated and coloured according to the number of fires present in that grid” (p. 8) This is not explained very clearly, the color bar label is also unclear.

Yes, we agree and have added more explanation to the caption. It now reads as follows:

"... Example plot showing how the active count along the back trajectories is calculated: a back trajectory arriving at Zeppelin Observatory 01:00:00 on the 19th of July 2002 is displayed (blue). The active forest fires, for that period in which the back trajectory is present, are displayed as orange triangles. The number of active fires within each grid cell is counted. The colours of the grid cells correspond to the density of active fires; this is displayed by the colour bar. All active fire data is taken from NASA satellite products, and in particular the MODIS Satellite (https://firms.modaps.eosdis.nasa.gov/download/). The number of active fires that each back trajectory traverses over whilst in the mixed-layer is counted. The sum for an ensemble of back trajectories for a particular arrival time is added together. In this example, for this given timestamp, grids were traversed containing a total of 384 active fires. ..."

Page 13 Line 140 “short-term perturbations in” (p. 13) what is short-term

Parts to do with, and any mention of, the scavenging ratio have been removed from the manuscript. However, previously 2 weeks was what was meant by “short-term perturbations”, in keeping with Garrett et al. (2011).:

Page 13, Figure S15 caption: “S” (p. 13) undefined

Again, the scavenging ratio parts have been removed from the manuscript. However, here the S denotes $\Delta\sigma_{\rm sp}/\Delta{\rm CO}$. We understand that this is not clear in the text:

Page 14, Figure S16 caption: “short-term” (p. 14) unlcear what this means

Again, the scavenging ratio parts have been removed from the manuscript. However, here short-term is referring to the life-time of aerosol particles, and roughly 2 weeks. This part is now deleted from the text.

Page 14, Line 159: “despite” (p. 14) Despite?

This whole paragraph is removed and it simply reads as follows: *"...For each cluster and season, the relationship between $\sigma_{\rm ap}$ and ATP is examined...."*

1.9 Recommendation

I recommend publication after the herein suggested major revisions to this manuscript.

1.10 References

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2 Additional changes

Page 5, Lines 140-142:

The following sentences have been removed. It was deemed unnecessary to the message of the manuscript:

"... The contribution of light scattering by air molecules is automatically corrected by regular zero measurements of particle-free air, about every hour. Corrections to the measurements are necessary to account for the light source and angular non-idealities. ...

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