We would like to thank both reviewers for their time and consideration of this manuscript. Outlined below are our finalized, point-by-point responses to both reviewers. All changes & additions to the manuscript as a result of these comments have been explicitly stated herein.

RC1: Response to Comments

Major Comments:

- Structure of the paper: As reflected in some minor comments, wildfires influence is largely cited in §3.1 and 3.2, whereas the dust influence is more emphasized in §3.3. My impression they are the two main aerosol types apart from "standard ambient aerosols" and that they should be discussed at the same time in the three sections. Second, I wonder if the paper would be more coherent is structured in the following order: 1. the classification using optical properties (à description of measured aerosol types), 2. Climatology (all seasonal and diurnal cycles) and 3. Trend. Perhaps, the § 3.3.1 should be merged to some extent in present §1 on climatology since seasonality is described in both sections. Similarly, §3.3.2 on variability and also trends (see next comment) should also be used as introduction to §2 describing the QR trend analysis.

  We thank the reviewer for this feedback, and we agree that a slight restructuring would be helpful. We propose the following new structure, with some differences to the reviewer's proposed outline:
  3.1 Climatology of aerosol optical properties at SPL and BOS (merged sections 3.1 & 3.3.1)
  3.2 Aerosol classification using optical properties (merged sections 3.2 & 3.3.2)
  3.3 Trends
  We feel the climatology section should be first since it essentially presents 'baseline' conditions as well as a description of what these properties indicate and some observations on changes to previous analysis. That would provide context before looking at the direct relationships that can be used to identify aerosol types. Finally, the discussion of statistically significant changes in source intensity and type. This would also better segregate the discussions of observed changes vs statistically evaluated trends by keeping them in separate sections (in regards to the next comment on trend analysis).

- Trend analysis:
  - The study comprises a long-term trend analysis with the QR method but also often mentions trends from visual inspection of the time series (e.g. Fig. 7, S6, S8, S15, S16 and related descriptions) or by comparing two periods (Fig. 5, S2, S10, Table S7, and related descriptions). A clear distinction should be made between the statistical analysis and the visual inspection/period comparison. As already mentioned in the point concerning the structure of the paper, I think that the statistical analysis should be moved to the end to confirm the previous descriptions in §3.3.2.

    In addition to the structural changes indicated above, statements/terminology will be added to the aerosol classification section to better indicate that the section discusses observed / visual changes and not statistical trends. For example, the following revised sections with changes show in **bold**:

Lines 385 - 390: “The largest cluster of data falls into the BC dominated classification, with 39% and 52% of the total hourly data falling into this category at SPL and BOS, respectively. However, while the contribution of this category is relatively stable at BOS (Fig. S15), its contribution to the aerosol population at SPL **visually appears to show a marginal decrease** (Fig. S16). **This is consistent with the statistically significant decrease in aerosol at SPL, which is discussed in the next section on trend analysis**, and indicates that the background aerosol likely falls into this classification.”

Lines 236 - 244: “Long-term seasonal analysis of PM10 σsp at SPL **shows** that overall aerosol loading is decreasing, with the majority of quantiles showing decreasing trends in all seasons except the fall (Fig. 4a). **This confirms the observed changes discussed in previous sections** and is generally consistent with other trend analyses for aerosol scattering coefficient (Collaud Coen et al., 2020) and aerosol extinction coefficient (Hand et al., 2020) in the U.S. Differences were observed in the trends between seasons and over different data quantiles (Fig. 4 & S7). In all seasons, PM10 σsp values in and below the 70th quantile had significant but small changes (less than ± 2% yr-1, Table S5). At the higher percentiles, statistically **significant trends** were larger indicating that changes in PM10 σsp are being driven by extreme events (Fig. 4a). For the seasonal analysis of SAE, significant trends were generally decreasing and larger in the lower quantiles for all seasons except the summer (Fig. 4b). A decreasing trend in SAE indicates an increased contribution from coarse mode aerosol.”

- The statistical QR method is applied to the scattering coefficients and the SAE, but not on the absorption coefficients, on AAE, SSA and SSAAE. I’m wondering why since the two main aerosol categories (wildfires and dust) largely impact the absorption of light. I recommend to add these trends if possible.

  The scattering and SAE were chosen to give information on overall aerosol loading and size. While both the scattering and absorption are related to total loading, scattering dominates the total extinction at both sites throughout the year (Fig. 2, S2, 5c) and during wildfire and dust events both scattering and absorption increase (Fig. 1). Additionally, the scattering measurement has a lower overall uncertainty. For these reasons we used scattering trends to investigate the changes in aerosol loading.

  The QR trend analysis was not applied to the AAE and SSA for two reasons. First, because of the filtering constraints we placed on the calculated parameters (L137 - 146). With over 50% of the data removed for AAE and SSA, we felt less confident performing seasonal QR trend analysis on the data even though the overall statistics (e.g., median, 25th, and 75th percentile) of the variables were not changed significantly (Table S2). Second, straightforward QR analysis of AAE wouldn’t necessarily give useful information about changing sources due to the fact that an aerosol size indicator (SAE in this work) is needed to better contextualize AAE values. For example, a large AAE value (> 2) indicates dust if the particles are large but biomass burning aerosol if the particles are small. But you wouldn’t be able to contextualize

the AAE QR trends with the SAE QR trends since their percentiles may not coordinate (i.e. the 90th percentile values of SAE may not be when the 90th percentile of AAE values are being measured). QR trends in SSA would be similarly hard to contextualize. For this reason, we believe that the direct comparison of AAE to other variables in the systematic variability and aerosol classification analysis presented is more helpful for looking at source changes that can then be verified using QR trends in overall aerosol loading and size.

- L87: SPL measurements start in 1981. Why are 1981-2010 data not used in this study?

  L87: Apologies for the confusion. SPL was established as a monitoring site in 1981, however, measurements of in-situ aerosol optical properties did not begin until 2011. The original text has been changed to state: "SPL was established in 1981 and the facility is currently operated by the University of Utah. Measurements of in-situ aerosol optical properties began in 2011." Hopefully, this will clarify that the measurements used in this paper did not begin until 2011.

- QR is not a largely used method for long-term trend analysis in atmospheric sciences so that it could be somewhat better described. One important point is the potential prerequisites on the data for QR analysis.
  The methods section has been edited to give further detail on QR analysis, and to better explain how it is performed. QR does not have substantial prerequisites compared to other types of trends analysis. Similarly to other methods a sufficiently long time range is needed and data breakpoints need to be identified and resolved before the analysis is performed. A statement has been added to reflect this.

  Line 175: "Quantile regression (QR) applies asymmetric weighting at different quantiles and regresses the entire dataset at specific quantiles to estimate the rate of change across the entire distribution of data (Koenker, 2005; Koenker and Hallock, 2001). The regressions presented are not for a single quantile or subset of the data, but for the entire dataset at a specific quantile. For example, in the estimation of a 75th quantile regression a regression line is fit through the data so 75% of the residuals are negative (below the regression line) and then the weighted distances are minimized."

  Line 185: "Similarly to other trend analysis, long term observations are needed and breakpoints in the data caused by changes in measurement conditions (e.g. inlet and instrument changes) must be resolved. An effort was made to identify any breakpoints in the SPL data, though nothing significant was found. "

- I have also further questions regarding the uncertainty: You report in Tables S5 and S6 an error/uncertainty. Could you briefly explain how it is calculated? These uncertainties are quite low also for the largest slopes that correspond to the most extreme percentiles. Is the uncertainty of the determination of extreme percentiles (e.g. the 0.98 percentile) similar the one of the median? Is the uncertainty of the determination of the percentile considered to compute the uncertainty of the slope? Considering the scattering coefficient in summer, I would be pleased to see a plot of 0.98 percentiles with the slope. I'm wondering if the large slope is mostly defined by the very high 2020 and 2021 values. Finally, these uncertainties could perhaps also be reported on Fig. 4.

The standard error reported in Table S5 and S6 were calculated using the ‘nid’ method in the analysis package (nonlinear interaction decomposition), which computes a Huber sandwich estimate using a local estimate of the sparsity. This method was chosen given the heteroscedasticity of the dataset. Bootstrapping was also utilized and produced similar error estimates.

Quantile regression uses the full dataset to estimate the percentiles. The regression coefficients are estimated using all observations per season, therefore, this sample size is very large for asymptotic approximations. The current method uses the Huber Sandwich estimate to calculate the variance and uses local sparsity of the data around the percentile that we are fitting to help incorporate the uncertainty of the percentile. This information on the error calculations has been clarified in the trends method section:

Line 184: “All trends are reported with a standard error, calculated using nonlinear interaction decomposition. This method uses the Huber Sandwich estimate to calculate the variance and uses local sparsity of the data around the percentile being fitted, which helps incorporate the uncertainty of the percentile. Bootstrapping was also performed and produced similar error estimates.”

We show a similar plot to what the reviewer asks for in Fig. S7 (Fig. S16 in new files), though it shows all the data with the highest percentile slopes (provided again below). It's important to reiterate that QR regresses the entire dataset at specific quantiles and not just the data in the 98th percentile, so showing just the 98th percentile and the trend is not entirely appropriate, however, we do show the summertime 98th percentile values compared all summertime values so the reviewer can see what the upper percentile looks like over time. While we acknowledge that the 2020 & 2021 years were extreme and contribute to the upper percentile trends, there are three years of data after them that help reduce the bias those years may introduce.

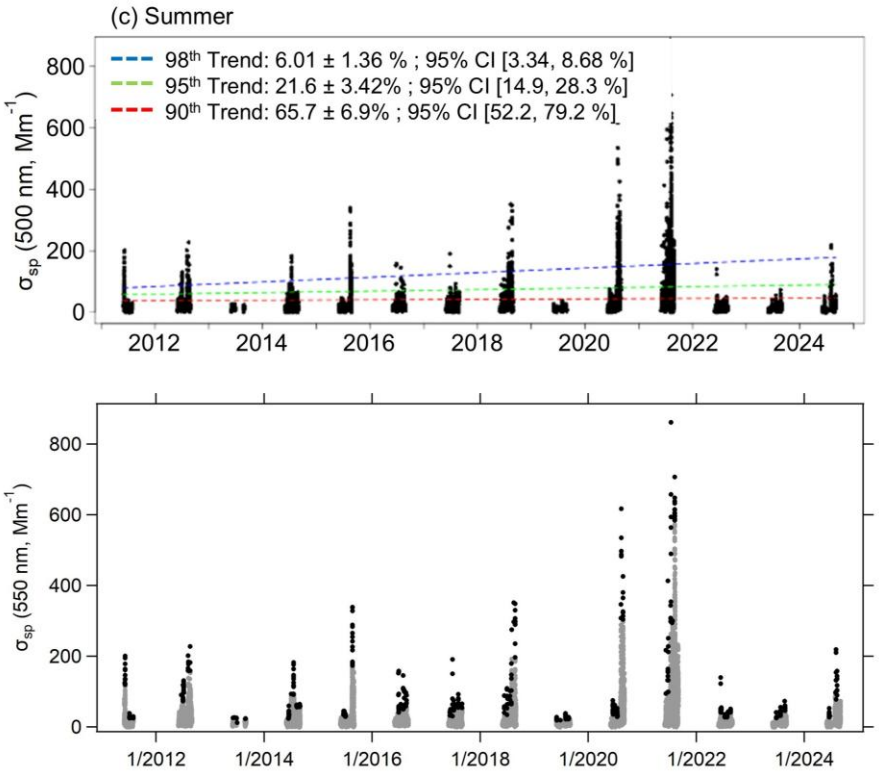

Figure RC1: Fig. S7a from the supplemental section along with a plot of hourly PM10 scattering data with 98th percentile values shown in black with all other data shown in grey.

We are happy to report the errors in Fig 4., the revised figure is shown:

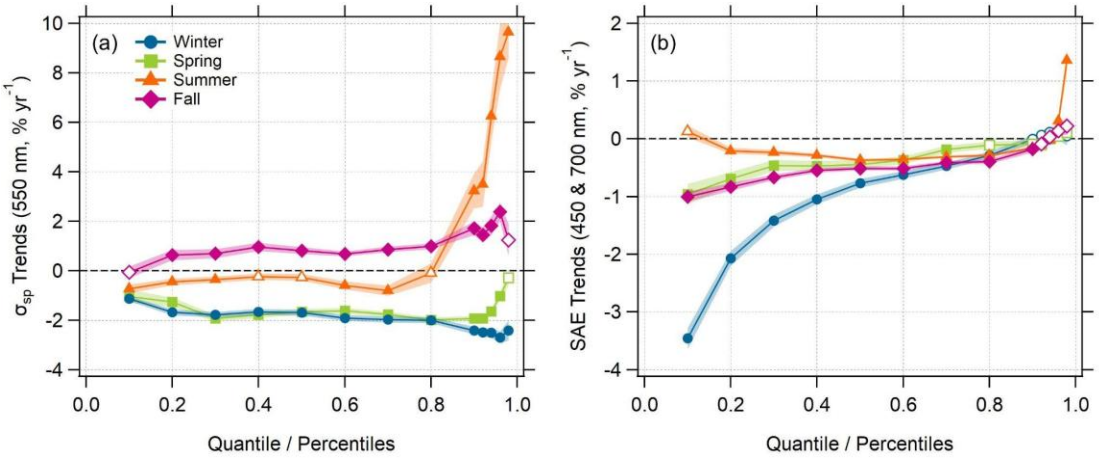

Fig. 4: Quantile regression trends for PM10 (a) σsp and (b) SAE at SPL over all seasons from 2011 to 2024. Winter = Dec – Feb, Spring = March - May, Summer = June - Aug, Fall = Sept - Nov. Open markers show trends that were not significant at (p-value ≥ 0.05). Numerical values and p-values for the quantile regression trends are provided in Table S5 & S6, along with the standard error which is shown as shading on the plot. As with other analyses presented, these trends are for dry sampling conditions (RH < 40%).

- Dust: With the used parameters, dust detection by negative SSA Ångström exponent is applicable and should allow to determine dust influence, at least for the remote station of SPL. It would allow a more detailed climatology with an estimate of the dust influence frequency, its seasonal cycle. This information can also help with the interpretation of the AAE-SAE figures. One question related to dust concerns the sharp increase in dust load in spring in BOS (Fig. S15) that is not visible in SPL. Do you have any explanation?

  The SSA Ångström exponent is a useful tool, however, given the dominant role of scattering at both sites throughout the year (Fig. S3 in paper & Fig. RC2a below) the SSAAE provides similar information / context as the other optical parameters already discussed. Analysis of this parameter could be added to the supplemental, but we do not feel that it would add substantially to the main text of this work. In the interest of completeness, the climatology of the SSAAE is provided (Fig. RC2b) along with a monthly comparison of SSAAE to σ_{sp} at both SPL and BOS (Fig. RC2c & RC2d). These figures show the clear influence of dust in the spring at both sites, marked by the negative SSAAE values. However, the information and identification of dust is nearly identical to the use of the SAE & BFR in the discussion of climatology (Fig. 5) and the comparison of SAE to σ_{sp} (Fig. 6) that is already provided. It's also worth noting that because the SSAAE depends on σ_{sp} and σ_{ap} at two wavelengths the impact of the filtering constraints applied to the calculated parameters in this work are significant, with ~65% of the hourly data points at SPL removed as a result. For these reasons, we think that if the SSAAE were to be added it should be in the supplemental material which we are happy to do upon request.

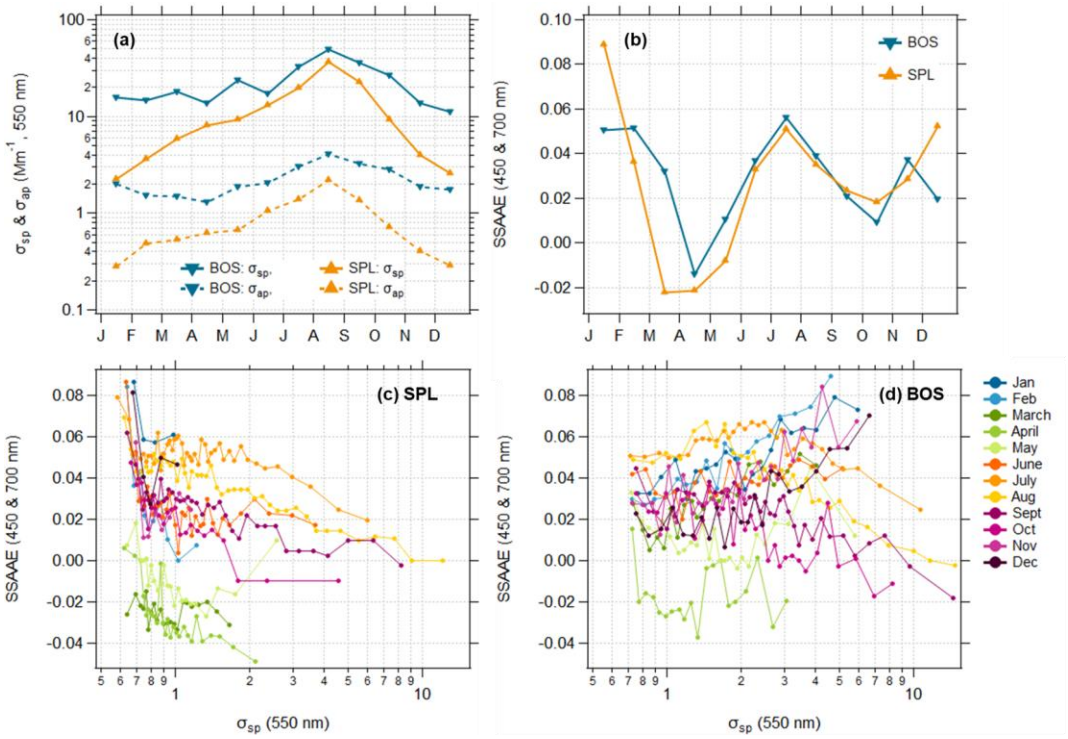

Fig RC2: (a) Seasonal cycles of median PM_{10} σ_{sp} and σ_{ap} (550 nm) for both SPL and BOS. (b) Monthly median PM_{10} SSAAE for 450 & 700 nm for both SPL and BOS. PM_{10} SSAAE (450 & 700 nm) binned by σ_{sp} (550 nm) for (a) SPL and (b) BOS. For all plots, each data point represents the median of 100 data points and traces are colored by month. Please note the log scale on panels a, c, & d.

Regarding the increase in spring dust loading at BOS (Fig. S15), there are several things to consider. First, it is important to note that while there is a sharp increase in the 'dust' classification at BOS it is an increase from 0% of measured hours to ~0.6% of measured hours in the spring (March - May), so not an increase to a significant fraction of the data. Additionally, without a longer measurement range we can't say that this is a trend or if we've just had a few big dust years since 2019 at BOS like in the 'dust' classification at SPL. Second, it's important to reiterate that these classifications have limitations especially as environmental conditions and source variability become more complex (L372 of text). For this reason, it's prudent to consider the 'dust' and 'mixed dust/BC/BrC' classifications together especially when considering sources at BOS - since the dust plumes sampled at the site are likely mixed with other aerosol types (L351, Fig. 6b of text). With all that said, at BOS the springtime contribution from the 'dust' and 'mixed dust/BC/BrC' classifications are both visibly increasing. And while there isn't a clear springtime increase in 'dust' at SPL there is an observable increase in the 'mixed dust/BC/BrC' contribution. This may be attributed to increasing dry conditions (Mankin et. al., 2021) and changes in land characteristics (Reynolds et. al., 2001) in the region that are likely leading to more local (i.e., from US sources) dust (Neff et. al., 2008). At this point, the increase in dust over the last 15 years is quite uncertain. For example, a recent study by Naple et al. (2025) demonstrated, using the MODIS record, that radiative forcing of dust on snow was decreasing slightly in the Colorado River Basin between 2011-2024. These patterns do not align with periods of drought in the Colorado River Basin, suggesting a complex relationship between aridity and dust (Naple et al., 2025). Similarly, Henen et al. (2023) did not find a statistically robust trend in dust within the Colorado Plateau, and describe the numerous controls on dust in this region.

Mankin, J.S., Simpson, I., Hoell, A., Fu, R., Lisonbee, J., Sheffield, A., Barrie, D. (2021) NOAA Drought Task Force Report on the 2020–2021 Southwestern U.S. Drought. NOAA Drought Task Force, MAPP, and NIDIS.

Reynolds, R., Belnap, J., Reheis, M., Lamothe, P., & Luiszer, F.: Aeolian dust in Colorado Plateau soils: Nutrient inputs and recent change in source, Proc. Natl. Acad. Sci. U.S.A. 98 (13) 7123-7127, https://doi.org/10.1073/pnas.121094298, 2001.

Neff, J. C., Ballantyne, A. P., Farmer, G. L., Mahowald, N. M., Conroy, J. L., Landry, C. C., Overpeck, J. T., Painter, T. H., Lawrence, C. R., and Reynolds, R. L.: Increasing eolian dust deposition in the western United States linked to human activity, Nature Geoscience, 1, 189–195, https://doi.org/10.1038/ngeo133, 2008.

Naple, P., Skiles, S. M., Lang, O. I., Rittger, K., Lenard, S. J. P., Burgess, A., & Painter, T. H.: Dust on snow radiative forcing and contribution to melt in the Colorado River Basin. Geophysical Research Letters, 52, e2024GL112757. https://doi.org/10.1029/2024GL112757, 2025.

Hennen, M., Chappell, A. and Webb, N.P.: Modelled direct causes of dust emission change (2001–2020) in southwestern USA and implications for management. *Aeolian Research, 60*, p.100852, https://doi.org/10.1016/j.aeolia.2022.100852, 2023.

Line / Minor Comments:

- L52-56: add a word on the potential use of REM observations ?
  L52-56: A note on remote sensing observations has been added and the paragraph from line 58 has been combined with the one from line 42 to give a complete description of aerosol measurement needs, and the roles of remote sensing and surface in-situ observations.

  "Determining the radiative properties of aerosol particles caused by dust and wildfires requires an understanding of the aerosol properties as well as atmospheric transport mechanisms (Davulienė et al., 2024), and meteorological conditions (Wilmot et al., 2021). However, due to the high spatiotemporal variability of aerosol particles and sensitivity to climate, land use change, land management policies, and human activity (Ford et al., 2018), the radiative impact of aerosol particles remains uncertain (IPCC, 2023; Jiang et al., 2020; Kok et al., 2023). Ground and satellite based remote sensing observations provide broad spatial information and can be used to investigate vertically resolved aerosol processes; however, they cannot provide fine details of conditions at specific locations. Further, variables such as single scattering albedo can only be retrieved during high loading events which may not be representative of the normal climatological conditions (Dubovik and King, 2000). Surface aerosol in-situ measurements provide aerosol information that is critical for quantifying aerosol climatology (temporal patterns, amount, and characteristics) at a specific location (Laj et al., 2020). These measurements can be used to estimate direct and indirect aerosol radiative forcing and to infer additional information such as aerosol type (Cappa et al., 2016; Schmeisser et al., 2017). Long term aerosol measurements can also allow diagnosis of trends and identification of the changes contributing to those trends."

- L61-64: Does the higher aerosol scattering and absorption coefficient patterns in the summertime relate only to increased wildfire? Does the high convective boundary layer (CBL) in summer associated with thermal wind system have no effect at SPL? Are dust events the only atmospheric aerosol leading to lower scattering Ångström exponent ?
  L61-64: High CBL in the summer has an effect at SPL, which is discussed in section 3.1 when the monthly changes in the diurnal cycles are evaluated. Overall scattering increases and the diurnal cycle is stronger, showing its effect. Dust is the dominant factor causing lower SAE - SPL is located in the central US distant from marine aerosol (e.g., sea salt) which are the other primary cause of low SAE atmospheric aerosol.

- L156: Please, explain what is the difference between BC and smoke: does smoke contains no BC?
  L156: The theoretical value of AAE for unmixed / unprocessed black carbon is 1, and that has been shown to be representative of aerosols from fossil fuel burning (Russell et al., 2010). Measured AAE values corresponding to light-absorbing OC (e.g. BrC) and biomass burning aerosol, which includes both BC and BrC, have been shown to be higher, as well as those for dust aerosols (Russell et al., 2010). So, while smoke/ biomass burning contains BC, the higher organic composition of smoke results in higher AAE values.

  Here, we used the term smoke to indicate biomass burning while also using the term BC to encompass fossil fuel burning which we acknowledge was not the best way to describe the variation

in AAE values. This section has been re-worded to be more accurate in the description of the aerosol types we are referencing: “The absorption Ångström exponent can yield information about particle composition (Russell et al., 2010) with values near 1 associated with black carbon and fossil fuel burning, while larger AAE values (≥2) are representative of biomass burning, light-absorbing organic carbon, and dust.”

- L167: Perhaps it is nice to mention that it corresponds to a 95% confidence level.
  L167: The sentence has been adjusted to state: “Trends using this method were considered significant when the p-value was < 0.05, corresponding to a 95% confidence level.”

- L176-178: the seasonal cycle with maxima in summer is easily explained and is similar to other mountain sites. BOS is however approximately at the same altitude as Boulder and Denver, where the higher CBL in summer should decrease the PM concentration if the PM sources are not higher in summer. The diurnal cycles (Fig. S5) are also similar at both sites apart in January. Could you please provide an explanation for BOS cycles.
- L188-190: yes, but stations near cities often have diurnal cycles bounded to vehicles’ emissions (morning and late afternoon) as well as BB due to heating in winter (visible in Nov-Jan in BOS). Why are the maxima in the middle of the day at BOS from spring to fall? If the summer maximum can relate to wildfires, there is no reason that wildfires have diurnal maximum.
  L176-178; L188-190: Concerning the diurnal cycle at BOS. BOS is located on a plateau that is directly east (~3 miles) of the Front Range region of the Southern Rocky Mountains. The topography of the site itself and its close proximity to complex mountain terrain means that the site is affected by thermally driven winds/ transport, which drive its diurnal cycle. However, its lower elevation and proximity to larger urban centers means that overall, it experiences higher concentrations of aerosols. Which is why the monthly/ seasonal trends show the urban influences but the diurnal cycle does not. So, while the site has anthropogenic influences it does not behave like a traditional ‘urban site’.

  The following addition has been added to section 2.1.2 to better describe the terrain around the site and better indicate that while it is close to urban regions it is not a traditional ‘urban site’: “BOS is located on a plateau on the eastern edge of the Rocky Mountain Front Range. The site is ~10 km north of the nearest population center of Boulder, Colorado (population ~105,900; Fig. 1) and 50 km northwest of Denver, Colorado (population ~715,000).”

  Additionally, a note has been made near line 195: “BOS exhibits a similar diurnal cycle given its location and proximity to complex terrain, though its cycle also shows influence from some anthropogenic emissions throughout the year. This is expected, as BOS resides within the boundary layer where most anthropogenic emissions take place.”

- L187: are upvalley wind also contributing to the SPL diurnal variability in summer or only upslope wind?
  L187: Likely both given the location and terrain around SPL. The term ‘upvalley’ has been added to acknowledge this.

- 2c: there is clearly a high burned area in Colorado explaining the 2018 and 2020 high PM load in summer. The 2021 scattering and absorption maxima are, however, not correlated with the burned acres. Please comment.
  Fig. 2c: While 2021 didn't have a clear maxima in terms of acres burned in Colorado, the Pacific Northwest region of Canada and the US experienced a severe heat dome that contributed to a significant wildfire season in that region. A note has been added to Line 202: "Often the magnitude of change could be visibly tied to wildfire metrics, such as the total acres burned locally and nationally as shown in Fig. 2c. A notable exception to this was in 2021, where the large increase in aerosol load was driven by transported smoke from the Pacific Northwest region of Canada and the United States, which experienced a severe heat wave that lead to a significant increase in wildfires (White et al., 2023), and smoke from central Canada (Bruce et. al., 2025)."

  White, R. H., Anderson, S., Booth, J. F., Braich, G., Draeger, C., Fei, C., Harley, C. D. G., Henderson, S. B., Jakob, M., Lau, C.-A., Mareshet Admasu, L., Narinesingh, V., Rodell, C., Roocroft, E., Weinberger, K. R., and West, G.: The unprecedented Pacific Northwest heatwave of June 2021, Nature Communications, 14, 727, https://doi.org/10.1038/s41467-023-36289-3, 2023.

  Bruce, E. D., Folorunsho, A., Jaisawal, N., Gaw, E., and Li, Y.: Intra-Continental Transport of Western Wildfire Smoke Heightens Health Risks Across North America, International Journal of Environmental Research and Public Health, 22, https://doi.org/10.3390/ijerph22020226, 2025.

- L208-209 and Fig. S6: I do not agree with this conclusion. 2020 and 2021 have a much higher rate of hours over the 50 Mm-1 threshold, but it is not obvious (and should be statistically demonstrated) that the number of hours over the threshold is higher in 2022-2024 than 2011-2019.
  L208-209, Fig. S6: This is a fair critique, however, we don't think a direct comparison of the total number of periods between 2022-2024 vs 2011-2019 would be appropriate given the sheer difference in total data. However, we can compare the median number of instances per year between the periods (median over average given the magnitude of the variation). The median number of outlier periods in 2011- 2019 is 92 hrs y^{-1}, with 2022-2024 having a median of 101 hrs yr^{-1}. If we look at the whole period of 2011-2024 the median number of periods is 110 hrs yr^{-1}, and 97 hrs yr^{-1} if the 2020-2021 period is excluded. The median number of periods for 2011-2016, which is the range for the previous analysis we compare to in this work, is 90 hrs yr^{-1}. Instead of a broad statement of increasing instances, the conclusion has been changed to state:

  "In the SPL data, there is a large variation in the number of outlier periods per year with 2020 & 2021 having the highest rates of these periods. The median number of instances per year has increased since previous analysis (2011-2016; Japngie-Green et al., 2019) from 90 hrs yr^{-1} to 110 hrs yr^{-1} (97 hrs yr^{-1} if the extreme 2020-2021 period is excluded). While this does not indicate a significant trend, it does show that overall these periods have become more frequent. This change in frequency is not discernable in the shorter BOS time series."

- L250: when is the dust season in BOS and SPL? From L277 spring seems to be the dust season. Is there a trend of dust due to droughts? What are the sources of dust? Fig. 12 a and b have lower SAE and higher AAE with higher scattering in spring, that can correspond to dust events. Do you observe an inversion of the SSAAE during dust?

Line 250: Spring is the major dust season at both sites, which includes both local and transported dust. The influence of transported dust from Asia outflow is described by Augustine et al. (2008) for BOS. Hallar et al. (2011, 2015) describe the influence of springtime transport of dust from both local and remote sources to SPL. The influence of these dust sources is also shown by chemical data from the region (Hand et. al., 2024). An inversion of the SSAAE can be seen in the spring when dust is a prominent source (see provided figures from above, Fig. RC2).

A note of this has been added to Line 297 to comment on the dust sources:
"These changes are likely due to dust sources in the spring. Transported dust from Asian outflow is a prominent springtime dust source at both sites (Augustine et al., 2008; Hallar et al., 2015; Japngie-Green et al., 2019), though surface fine dust from local deserts and semi-arid regions in North America are also an important source of dust (Hallar et. al., 2011, 2015). The influence of these dust sources is also confirmed by chemical data from the region presented by Hand et al (2024a).

Concerning trends in dust due to droughts and other factors. Pu & Ginoux (2017) showed increases in dust in the southwestern U.S. in spring and the work attributed the frequency of dust in the western U.S. to reduced precipitation, changes in land characteristics, and increased surface wind speeds. Activities such as livestock grazing have led to surface changes in the western interior United States, which has contributed to increased dust and can increase the risk of dust during drought (Reynolds et. al., 2001). That said, it is difficult to clearly illustrate a trend from this single site. Generally, in the Western US "it is difficult to attribute the cause of changes in atmospheric dust, not least because there is no direct relation between the main factors controlling dust emission and the different factors controlling atmospheric dust dispersion and deposition" (Hennen et al., 2023). Within this region, there is not a direct relationship between aridity and dust loading (Naple et al., 2025). The diverse land use and management practices in the Western US change ecosystems that thus has the potential to both accelerate and reduce dust (Hennen et al., 2023).

Hallar, A. G., Chirokova, G., McCubbin, I. B., Painter, T. H., Wiedinmyer, C., and Dodson, C.: Atmospheric Bioaerosols Transported Via Dust Storms in Western United States, Geophys. Res. Lett., 38, L17801, doi:10.1029/2011GL048166, 2011.

Hand, J. L., Prenni, A. J., Raffuse, S. M., Hyslop, N. P., Malm, W. C., & Schichtel, B. A.: Spatial and seasonal variability of remote and urban speciated fine particulate matter in the United States. *Journal of Geophysical Research: Atmospheres*, 129, e2024JD042579, https://doi.org/10.1029/2024JD042579, 2024.

Pu, B., Ginoux, P.: Projection of American dustiness in the late 21st century due to climate change. Sci Rep 7, 5553, https://doi.org/10.1038/s41598-017-05431-9, 2017.

Reynolds, R., Belnap, J., Reheis, M., Lamothe, P., & Luiszer, F.: Aeolian dust in Colorado Plateau soils: Nutrient inputs and recent change in source, Proc. Natl. Acad. Sci. U.S.A. 98 (13) 7123-7127, https://doi.org/10.1073/pnas.121094298, 2001.

Hennen, M., Chappell, A. and Webb, N.P.: Modelled direct causes of dust emission change (2001–2020) in southwestern USA and implications for management. *Aeolian Research*, *60*, p.100852, https://doi.org/10.1016/j.aeolia.2022.100852, 2023.

Naple, P., Skiles, S. M., Lang, O. I., Rittger, K., Lenard, S. J. P., Burgess, A., & Painter, T. H.: Dust on snow radiative forcing and contribution to melt in the Colorado River Basin. Geophysical Research Letters, 52, e2024GL112757. https://doi.org/10.1029/2024GL112757, 2025.

- 4b: do the lower SAE quantiles correspond to negative SAE?
  Fig 4b: No, negative SAE values were included but were incredibly sparse. After applying scattering thresholds for calculated parameters (L142) ~0.3% of the SAE hourly data points were negative over the entire 13 year data range at SPL. The winter had the majority of these periods but still only 0.7% of hourly periods out of the entire record had negative values. Additionally, none of the wintertime 10th percentile cut off values were negatives so those trends should not be written off as just being attributable to negative SAE values.

- S9: months with the greatest SSA correspond to the largest wildfires activities (July and August). How do you explain this? If BB are produced in Colorado (e.g. clearly visible in 2020), is the explanation of aged smoke aerosol (L287) correct? Usually, the mentioned gaseous coating is found to produce a lensing effect that increases the absorption of the light. The interpretation of AAE seasonal cycle (L297) with a mixture of anthropogenic BB and BC in summer is also difficult to bound to the SSA seasonal cycle.
  Fig S9 / L287: This is a great question. It's important to reiterate that SSA is the relative contribution of scattering to total extinction. So even though absorption increases during wildfire periods, it doesn't necessarily become the dominant contributor to extinction. Biomass burning (BB) from wildfires has been shown to be more scattering overall, with higher measured mass scattering efficiencies (MSE) compared to mass absorption efficiencies (MAE) leading to higher SSA values. This was shown during the Fire Laboratory at Missoula Experiments (Levin et. al., 2010; Mack et. al., 2010), and is consistent with measured MSE and SSA for BB sources under ambient conditions (Laing et. al., 2016). It's also been shown that SSA can increase as the age/ transport time of BB plumes increase. Using flights with repeated transects of BB plumes, Kleinman et. at. (2020) showed increases in both SSA and MSE for aged wildfire smoke. Selimovic et. al (2019) also observed increases in SSA as a function of time over a prolonged period of biomass burning. With these results we do expect the SSA to be high when the sites are measuring BB sources, even with the increase in BB / BC in the summer as indicated by the AAE.

  Regarding the lensing effect that can increase absorption of BC particles. While we acknowledge that this is an important process affecting absorbing aerosols, the overall effect of this is poorly constrained and there are many conflicting studies on the magnitude of this enhancement (e.g. Cappa et al., 2012; Liu et al., 2015). As shown above, when sampling BB sources as a whole the scattering still tends to dominate the total signal. Without a dedicated long-term BC measurement there is no way for this work to comment on that effect.

Levin, E. J. T., McMeeking, G. R., Carrico, C., Mack, L., Kreidenweis, S. M., Wold, C. E., Moosmuller, H., Arnott, W. P., Hao, W. M., Collett, J. L., and Malm, W. C.:Biomass burning smoke aerosol properties measured during Fire Laboratory at Missoula Experiments (FLAME), J. Geophys. Res.-Atmos.,115, doi:10.1029/2009JD013601, 2010.

Laing, J. R., Jaffe, D. A., and Hee, J. R.: Physical and optical properties of aged biomass burning aerosol from wildfires in Siberia and the Western USA at the Mt. Bachelor Observatory, Atmos. Chem. Phys., 16, 15185–15197, https://doi.org/10.5194/acp-16-15185-2016, 2016.

Mack, L. A., Levin, E. J. T., Kreidenweis, S. M., Obrist, D., Moosmüller, H., Lewis, K. A., Arnott, W. P., McMeeking, G. R., Sullivan, A. P., Wold, C. E., Hao, W.-M., Collett Jr., J. L., and Malm, W. C.: Optical closure experiments for biomass smoke aerosols, Atmos. Chem. Phys., 10, 9017–9026, https://doi.org/10.5194/acp-10-9017-2010, 2010.
Kleinman, L. I., Sedlacek III, A. J., Adachi, K., Buseck, P. R., Collier, S., Dubey, M. K., Hodshire, A. L., Lewis, E., Onasch, T. B., Pierce, J. R., Shilling, J., Springston, S. R., Wang, J., Zhang, Q., Zhou, S., and Yokelson, R. J.: Rapid evolution of aerosol particles and their optical properties downwind of wildfires in the western US, Atmos. Chem. Phys., 20, 13319–13341, https://doi.org/10.5194/acp-20-13319-2020, 2020.

Selimovic, V., Yokelson, R. J., McMeeking, G. R., and Coefield, S.: In situ measurements of trace gases, PM, and aerosol optical properties during the 2017 NW US wildfire smoke event, Atmos. Chem. Phys., 19, 3905–3926, https://doi.org/10.5194/acp-19-3905-2019, 2019.

Cappa, C. D., Onasch, T. B., Massoli, P., Worsnop, D. R., Bates, T. S., Cross, E. S., Davidovits, P., Hakala, J., Hayden, K. L., Jobson, B. T., Kolesar, K. R., Lack, D. A., Lerner, B. M., Li, S.-M., Mellon, D., Nuaaman, I., Olfert, J. S., Petäjä, T., Quinn, P. K., Song, C., Subramanian, R., Williams, E. J., and Zaveri, R. A.: Radiative Absorption Enhancements Due to the Mixing State of Atmospheric Black Carbon, Science, 337, 1078–1081, https://doi.org/10.1126/science.1223447, 2012.

Liu, S., Aiken, A. C., Gorkowski, K., Dubey, M. K., Cappa, C. D., Williams, L. R., Herndon, S. C., Massoli, P., Fortner, E. C., Chhabra, P. S., Brooks, W. A., Onasch, T. B., Jayne, J. T., Worsnop, D. R., China, S., Sharma, N., Mazzoleni, C., Xu, L., Ng, N. L., Liu, D., Allan, J. D., Lee, J. D., Fleming, Z. L., Mohr, C., Zotter, P., Szidat, S., and Prévôt, A. S. H.: Enhanced light absorption by mixed source black and brown carbon particles in UK winter, Nature Communications, 6, 8435, https://doi.org/10.1038/ncomms9435, 2015.

- L310: a QR analysis of AAE could help investigating the increase of AAE with time.
  L310: Possibly, however, the interpretation of the AAE trend would need to be done with care and a straightforward QR analysis wouldn't necessarily give useful information about changing sources. See the response to this comment from above.

- L338: remove ‘,’ after two
  L338: Done

- L391-392: the shift towards large particles can be caused by either an increase in dust contribution or a decrease in ambient aerosol contributions. Please comment.
  L391-392: That is true, and in other sections we made explicit comments on that. For example, in line 256: “This could be the result of a decrease in the intensity of dust events, or a decrease in background aerosol overall…”
  And in line 331: “...the AAE increase in springtime [relative to previous literature] could be the result of either an increase in dust events, overall decrease in background aerosol, or some combination of the two.”

  To make it clear in this section that it could be either we have changed the text to read:
  “Clear seasonal differences exist in aerosol classification at both sites (Fig. S17, Table S7). In the springtime at SPL the dominant aerosol classification changes to mixed dust/ BC/ BrC with 43 ± 18 % falling in this group. Contribution of dust and mixed dust/ BC/ BrC also appear to be increasing in the spring and winter at SPL. However, the number of data points in the dust only class is small (on average ≤ 1 % of data in all seasons, Table S7). At BOS, the BC dominated class is still largest in the spring, but the mixed dust/ BC/ BrC class is the second largest (Table S7) and contribution from dust at BOS is clearly increasing (Fig. S15). Looking specifically at extreme events in the spring, Fig. 7a and 7b show data where the daily average σsp value was greater than or equal to the 90th percentile of the month; the points are colored by year. At SPL the AAE of springtime events is increasing (Fig. 7a), which could indicate a trend towards less mixed (i.e. a decrease in other ambient aerosol) or more extreme dust events. BOS shows a clear shift in the class of springtime extreme events towards dust and mixed dust/ BC/ BrC categories (Fig. 7b). Quantile regression on SAE at SPL also indicates a shift towards large particles in the spring, again consistent with either an increase in dust frequency and intensity or a decrease in contribution to background aerosol loading.”

- 7: figure caption should be AAE vs SAE. Could you please also mention either on the figures or on the y-axis that a-b are for March-May and c-d for June-September ?
  Fig. 7: The caption has been corrected and the month ranges have been added to the figure.

- Table S5: quantile =0.1 in Spring: 0.0.3 to correct
  Thank you, this has been corrected to -1.0 ± 0.3.

- S8: what is the difference between bottom and top (namely between a/b and c/d)?
  Fig S8: Apologies for not including this distinction originally. Panels (a) & (b) show data for the entire Rocky Mountain Area – including Colorado, Kansas, Nebraska, South Dakota, and Wyoming – while (c) & (d) show data only for Colorado. This has been added to the caption.

RC2: Response to Comments

Major comments:

- The authors use relationships between optical properties to try and identify aerosol types and composition. They could take advantage of long-term composition data near these sites from the IMPROVE and CSN networks. These data have been reported in Hand et al (2024, https://doi.org/10.1029/2024JD042579) and may help place the results of this paper into the context of what has been reported for the aerosol composition in this same region and to discuss whether their results appear consistent with measured composition.
  This is a great suggestion. Given that none of the IMPROVE or CSN sites are directly collocated spatially with the SPL and BOS measurement sites and that the temporal resolution is different (hourly and continuous for the SPL and BOS measurements vs 24h every 3 days for IMPROVE), we chose not to pull the data from those networks into this analysis. But the reviewer is correct that Hand et al. (2024) is a great study that could be used to contextualize this optical analysis.

  That said, Hand et. al. (2024a) shows for the 2019-2022 time period the Central Rockies (where SPL is located) are dominated by dust in the spring months while organics dominate the summer and early fall (June-October), which they partly attribute to wildfires (their Fig. 6). In contrast, the Colorado Front Range (where BOS is located) is less impacted by dust in the spring and organics are highest in August and September. Hand et al. (2024b) look at mean trends in individual aerosol types over 2000-2021. They show a positive trend in summer and fall organic mass concentrations in Colorado that they attribute in part to wildfires (their Fig. 9) although the overall mean trend for the SW region is negative for all seasons (their Fig. 10). They show no or negative mean trends in dust for Colorado and the SW region for winter and spring (their Figures 13 and 14). This is generally consistent with our quantile regression figure that only shows trends at the extremes, but not for the median. The trends cannot be directly compared because different years were considered and different methods used.

  The following references have been added to the text:
  Line 285: “This is consistent with positive trends in organic mass concentration in Colorado that have been previously attributed to wildfires (Hand et al., 2024b)”

  Line 315: “The influence of these dust sources is also confirmed by chemical data from the region presented by Hand et al. (2024). ”

  Hand, J. L., Prenni, A. J., Raffuse, S. M., Hyslop, N. P., Malm, W. C., & Schichtel, B. A.: Spatial and seasonal variability of remote and urban speciated fine particulate matter in the United States. *Journal of Geophysical Research: Atmospheres*, 129, e2024JD042579, https://doi.org/10.1029/2024JD042579, 2024a.

  Hand, J. L., Prenni, A. J., & Schichtel, B. A.: Trends in seasonal mean speciated aerosol composition in remote areas of the United States from 2000 through 2021. *Journal of Geophysical Research: Atmospheres*, 129, e2023JD039902.

https://doi.org/10.1029/2023JD039902, 2024b.

- Have the authors compared size-related optical properties to each other? For example, the ratio of scattering a 1 um to 10 um compared to SAE? Or BFR

  We did briefly evaluate the differences between PM1 vs PM10. The scattering and absorption sub-micron fractions were discussed in lines 176 - 180, and their monthly medians were shown in Fig S2. We also show the sub-micron values for SAE and BFR in Fig. S9 & S10. These values and figures are all referenced throughout the discussion of the main text.

  However, if the reviewer is commenting on a comparison between sub-micron fraction and overall SAE/ BFR then no, that was not initially done in this work. The analysis could be added to the supplemental, but we do not think it would add substantially to this work. In the interest of completeness, we did the same comparisons presented in Fig. 6 & S12 - binning the SAE and BFR by the Rsp monthly with each point representing 100 hourly data points (see figures below). For SAE at both sites, the lowest values / increased particle size tend towards the lowest Rsp values / decreased sub-micron contribution. The BFR to Rsp relationship is similar, showing that when there is a shift to larger particles (lower BFR) the Rsp / sub-micron contribution also decreases though the relationship is not as strong given that BFR is more sensitive to changes in accumulation mode aerosols. The top row of Fig RC3 is consistent with previous work done by Li & Ginoux (2025), who saw similar relationships in the SAE and sub-micron fraction for 8 NFAN sites (labeled AE and FMF in their Fig. 3), and Delene & Ogren (2002), who showed similar Rsp to SAE relationships for other surface in-situ sites.

Li, X., & Ginoux, P.: An empirical parameterization to separate coarse and fine mode aerosol optical depth over land. Geophysical Research Letters, 52, e2024GL114397. https://doi.org/10.1029/2024GL114397, 2025.

Delene, D. J., & Ogren, J.A.: Variability of Aerosol Optical Properties at Four North American Surface Monitoring Sites. J. Atmos. Sci., 59, 1135–1150, https://doi.org/10.1175/1520-0469(2002)059<1135:VOAOPA>2.0.CO;2, 2002.

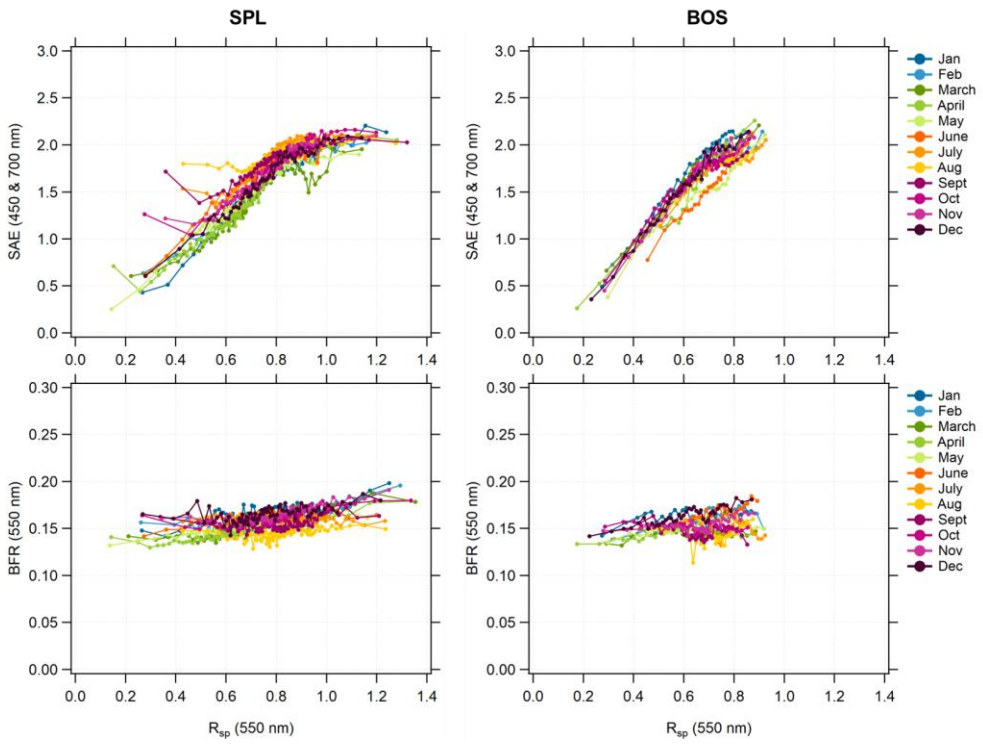

Fig RC3: PM_{10} SAE (450 & 700 nm) binned by R_{sp} (550 nm) for (a) SPL and (b) BOS. PM_{10} BFR (550 nm) binned by the BFR (550 nm) for (c) SPL and (d) BOS. For all plots, each data point represents the median of 100 data points. Traces are colored by month.

- "Data" are commonly referred to in the plural within scientific literature, but there are many instances in this paper where it is referred to as the singular. Consider using the plural consistently.

  Thank you for the comment, a conscious effort has been made to remove and correct the instances of this in the text. If there are any notable instances in the revised text please let us know.

- When referring to trends, it is helpful to refer to a trend as "negative" (or "positive") rather than "decreasing" (or "increasing") because the trend itself is a rate. For example, for decreasing light scattering, the trend is negative. Consider replacing these instances in the text.

  We have changed the terminology in the section on trends to use negative / positive instead of increasing / decreasing.

- Organizational comment: It might be useful to reorganize the paper so that the trend analysis comes at the end. In this way the reader can contextualize the patterns observed in the data to how they have changed over time.

  We thank the reviewer for this feedback. Reviewer #1 also had this comment and we agree that a slight restructuring would be helpful. We propose the following new structure:

3.1 Climatology of aerosol optical properties at SPL and BOS (merged sections 3.1 & 3.3.1)
3.2 Aerosol classification using optical properties (merged sections 3.2 & 3.3.2)
3.3 Trends

The new text will be re-structured with this outline.

- Figures: It would help to increase the font on the axis labels. It's difficult to see the difference between the subscripts for "sp" and "ap" for scattering and absorption, respectively.
  We can absolutely increase the axis and legend labels to improve visibility, thank you for the comment.

Line / Minor Comments:

- Line 75: The Storm Peak Laboratory and Table Mountain Field Site acronyms have been previously introduced and so could be used here.
  Line 75: We elected to use the full name in this instance since it's the first reference to the sites in the methods section.

- Line 79: Define GAW
  Done

- Line 104: Aerodynamic diameter is usually greater than physical diameter (Dp ~ Dae*SQRT((Chi/rho_p)), where Dp is physical diameter, Dae is aerodynamic diameter, Chi is shape factor, and rho_p is particle density.
  Line 104: This was not meant to denote physical vs aerodynamic diameter. This was a statement of manufacturer specification vs aerodynamic cut diameter for sampling in our system. An additional citation has been added to better reference performance testing, and the text has been edited to read:

  "A switched impactor system provides size-segregated measurements using Berner-type multijet cascade impactors for 10 μm and 1 μm size cuts, which have aerodynamic cut diameters of ~7 μm and ~0.7 μm under our sampling system (Berner et al., 1979; Hillamo and Kauppinen, 1991)."

- Line 111: Acronyms for light scattering and backscattering have been introduced and can be substituted here.
  Line 111: The mention of these has been changed to "σ_{sp} and σ_{bsp}".

- Line 115: What are the uncertainties in scattering and absorption?
  Line 115: Generally total uncertainty in scattering from the TSI 3760 Nephelometer is ~9.2%, and for absorption from the CLAP is ~20% for a 1-hr averaging time for typical continental conditions. The following statements and references have been added to the instrumentation section to indicate this:

  Line 120: "The overall uncertainty in the nephelometer measurements has previously been evaluated to be ~9.2% under average conditions at continental sites (Sherman et al., 2015). Error was calculated for the instrument and measurement conditions at SPL and BOS and were found to be generally comparable under both extremely clean (~10% error) and heavy aerosol loading (~9.2% error) conditions (Table S2)."

Line 135: “Uncertainty in the CLAP σap measurement is ~20%, though it increases substantially under extremely clean conditions to ~40% (Section S2; Ogren et al., 2017; Sherman et al., 2015).”

- Line 138: Change “wavelength” to “wavelengths”
  Done

- Line 139: Was this filtering also applied to analyses of light scattering and absorption, or only for calculated properties? Uncertainties in these calculated values could be calculated by propagating uncertainties in scattering and absorption. It would be helpful to provide these uncertainties, especially given the map of aerosol types shown later.
  This filtering was only applied to the calculated parameters. Following previous established methods for error propagation (Sherman et. al., 2015), we have calculated the typical fractional uncertainties for hourly averaged data when PM_{10} σ_{sp} = 50 Mm^{-1} and when σ_{sp} = 1 Mm^{-1}. These are provided below in Table RC1. We chose those two values to represent average uncertainties for extreme/outlier events and for very clean conditions that we used as the filtering cut offs. Note these are the averages for SPL and BOS - the uncertainties are slightly different for both sites as they depend on things like measurement temperature and pressure, and the correlations between various optical parameters. More details are in the supplemental materials of Sherman et al. (2015).

  We have added the following statements to the text to indicate this:
  Line 152: “Below those values the calculated parameters are more heavily influenced by instrument noise and are less reliable. Average fractional uncertainties for SSA, BFR, SAE, and AAE at these σ_{sp} and σ_{ap} constraints are 3.5%, 8.2%, 6.3% and 32.2% respectively.”
  Fig 7 caption: “Average fractional uncertainties for AAE and SAE at these levels are approximately 10% and 2.9% respectively.”

  We have added Table RC1 to the supplemental under Section S2: Description of auxiliary data treatment and calculations.

  Table RC1. Uncertainties of measured and calculated as a function of loading

	σ_{sp}	σ_{ap}	SAE	BFR	SSA	AAE	Rsp	Rap
σ_{sp} = 50 Mm^{-1}	9.2%	20%	2.9%	2.3%	1.0%	10%	2.8%	5.9%
σ_{sp} = 1 Mm^{-1}	10%	40%	6.3%	8.2%	3.5%	32.2%	3.1%	30%

Sherman, J. P., Sheridan, P. J., Ogren, J. A., Andrews, E., Hageman, D., Schmeisser, L., Jefferson, A., and Sharma, S.: A multi-year study of lower tropospheric aerosol variability and systematic relationships from four North American regions, Atmos. Chem. Phys., 15, 12487–12517, https://doi.org/10.5194/acp-15-12487-2015, 2015.

- Line 170: What completeness criteria did the authors apply to calculate the parameters used in the trends? As well as in the climatological properties?

  In terms of both the climatology and the trend analysis we tried to ensure that there was enough data coverage in each season and month for the analysis to be considered representative. For both the σ_{sp} and the SAE at SPL, we have ≥50% hourly data coverage in every season and month even after filtering was applied to the calculated variables. There is similar average monthly coverage for σ_{ap} and BFR, however, coverage is low for SSA and AAE. Coverage for σ_{ap} is lower than that for σ_{sp} because the measurement is filter based, and at remote sites the filter does not always get changed right away. The percent coverage of hourly data for all of these variables is summarized in Table RC2 below. For the calculated variables, we confirmed that the overall 25th, 50th, and 75th percentiles did not change significantly as a result of the filtering (Table S2). Taking this further for the SSA and AAE, since these variables had the lowest coverage, it can be seen that a similar climatology is shown in both the filtered and unfiltered data even with the low data coverage (**Fig RC4)**. Altogether, this gives us confidence that the measurements are representative of conditions at the two sites.

  Table RC2: Average seasonal and monthly percent coverage of hourly data for σ_{sp} and σ_{ap}, as well as the calculated parameters, SSA, BFR, SAE, and AAE, after quality control filtering was applied.

	Winter	Spring	Summer	Fall
σ_{sp}	86.9	91.8	83.9	80.2
SAE	59.4	81.7	83.0	72.7

SPL	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
σ_{sp}	85.8	91.9	91.9	87.6	96.0	90.4	78.3	82.9	85.4	77.0	78.3	83.3
σ_{ap}	75.6	85.6	84.0	76.4	64.7	62.6	65.2	66.0	76.2	68.4	70.6	72.7
SSA	10.4	21.4	35.7	39.2	35.9	44.9	50.4	51.4	50.4	32.9	19.3	11.4
BFR	63.2	81.3	82.8	80.3	89.5	89.5	78.1	82.7	83.8	72.0	70.7	65.5
SAE	50.5	73.8	79.0	78.3	87.7	88.8	77.7	82.4	83.1	69.5	65.6	54.4
AAE	5.3	11.3	23.1	27.4	23.5	37.4	46.4	47.4	40.8	23.0	9.7	6.0

BOS	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
σ_{sp}	94.0	99.8	96.8	91.5	97.4	90.6	93.4	97.9	99.8	97.2	98.8	95.8
σ_{ap}	86.7	98.9	94.1	91.4	96.6	90.1	93.2	95.6	96.5	93.3	94.8	91.7
SSA	60.4	69.3	69.4	70.7	77.3	77.0	88.0	87.6	89.0	78.3	74.6	59.5
BFR	92.1	98.9	95.5	90.9	96.7	90.6	93.5	97.9	99.4	96.4	98.1	91.4
SAE	89.2	96.2	94.4	90.6	96.2	90.2	93.3	97.9	99.4	96.5	95.5	87.6
AAE	64.1	60.7	66.0	65.0	75.7	79.1	89.2	91.6	89.8	79.3	69.2	57.3

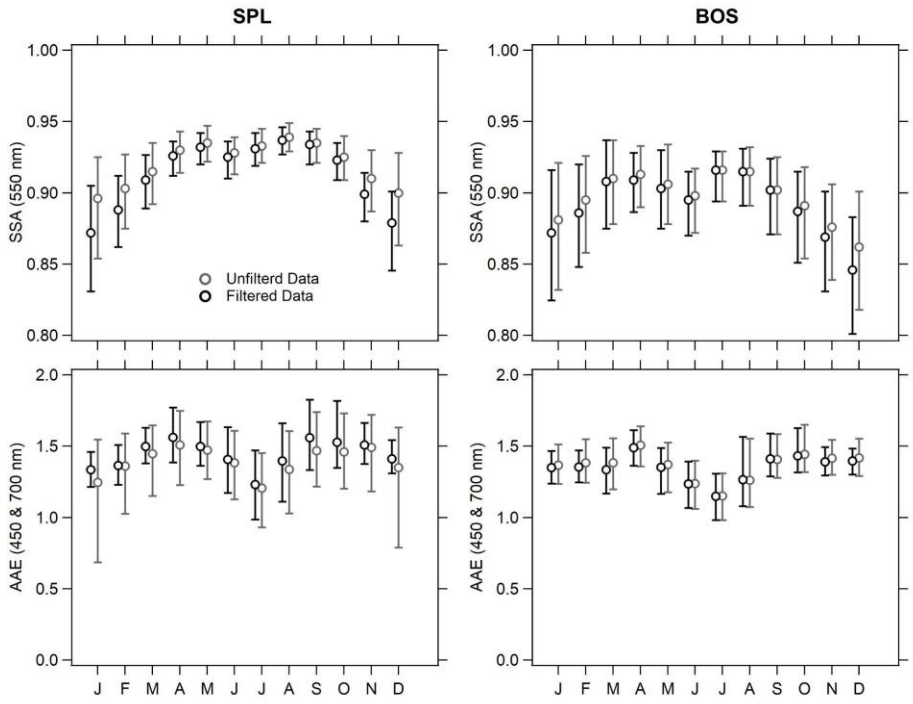

Fig RC4: Monthly 25th, 50th (median), and 75th percentiles for the SSA (550 nm) and AAE (450 & 700 nm) at SPL (left) and BOS (right). Grey markers include all measurements, while black markers show statistical values after filtering was applied based on total scattering and absorption.

- Line 173-174: Explicitly mention what is shown in Figure 2 and 3 (rather than group them together in the text).
  Line 186: The following statement has been added to differentiate between the plots:
  "This is visible in both the timeseries of average monthly σ_{sp} and σ_{ap} values, shown in **Fig. 2**, and the annual climatology, shown in **Fig. 3**."

- Line 175: Add "respectively" after "BOS"
  Done

- Line 207: It appears contradictory that in Lines 199-200 the authors state that during the May event limited upward and westward transport to SPL occurred.

  By limited we did not mean there was no transport to SPL for the May event, rather that transport of smoke to SPL was reduced relative to BOS because the mountains acted as vertical and horizontal barriers. BOS sampled higher concentrations for a longer period of time than were sampled by SPL. The text has been changed to read:

  Line214: "...smoke being channeled down the Front Range of the Rocky Mountains with less upward

and westward transport to SPL (see 2023 BOS σ_{sp} and σ_{ap} spike in **Fig. 2**). ”

- Line 220: Why isn’t May included?
  This was a typo, May is included. All analysis referencing the ‘spring’ in this work includes measurements for March – May.

- Line 227: Presumably all the light scattering data used for these trends are for RH<40% so the impacts of RH on scattering are not influencing these trends. Did the authors investigate these trends as a function of RH even below 40%? It is important to remind the reader that these trends are for aerosol properties considered to be “dry”.
  Line 227: That is correct, and trends were not investigated as a function of RH, even <40%. To give context we have provided an overall and monthly histograms using data from 01/2022 – 07/2025, of the hourly RH data form inside the nephelometer at BOS (Fig. RC5). The majority of data is below an RH of 20%, and even in the summer when RH is highest the sample RH does not get higher than 35%. The RH at SPL would be expected to be lower than that at BOS, given its lower ambient temperatures. This just doesn’t leave much room to look at the trends in the context of RH. Additionally, the composition and loading changes throughout the year along with RH would make it difficult to separate changes caused by the different factors.

  Following other comments by the reviewer, reminders of the dry sampling conditions have been added at various points throughout the text and in figure captions.

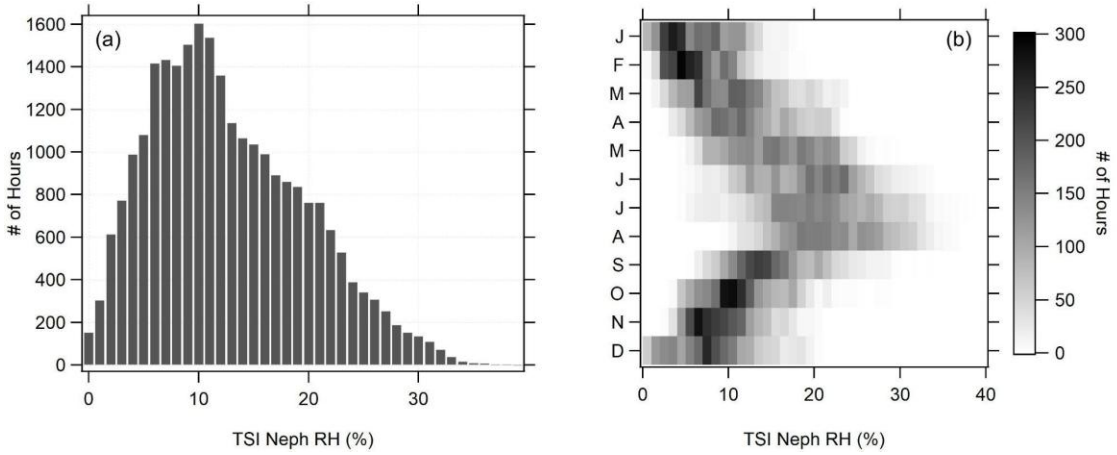

Figure RC5: (a) Total and (b) monthly distribution of hourly median measurement cell RH for the nephelometer for the 01/2022 – 07/2025 period. For all histograms shown, bin width was 1%.

- Line 237: Trends in SAE in spring are ~0. If dust has decreased, wouldn’t SAE likely increase? Since winter and spring trends diverge over the 80th percentile (and presumably the increase in spring is related to dust) wouldn’t you expect to see this in the SAE? Do you see this impact on other size-related parameters?
  Trends at the highest quantiles (70th and above) of SAE in the spring are ~0 and not significant. This indicates that there are not significant changes in the springtime periods that are dominated by smaller particles. Increased dust contributions would lead to lower SAE so changes in the lower percentiles of

SAE will be indicative of enhanced dust. Figure 4 in the submitted manuscript shows that the lower percentile trends in SAE (below the 80th quantile, and even more obviously below the 50th quantile) for winter, spring and fall exhibit statistically significant negative trends. These negative trends suggest an increasing contribution of coarse mode aerosol during these seasons, which is likely from decreases in concentrations of smaller background aerosols.

Looking at the scattering trends in the winter and spring above the 80th percentile, we do see a divergence. While that is likely related to dust, it's important to point out that those trends in both seasons are still negative, the ones in the spring are just less negative and veer towards zero. As we stated in the text that could indicate a slight decrease in dust intensity which would change total aerosol loading but may not lead to large changes in the SAE, since SAE is independent of concentration. Alternatively, it could be an artifact of an overall decrease in aerosol loading in the spring that would simply be less apparent in the upper quantiles when larger sources / events are adding to the total loading.

Since SAE and Rsp are the primary variables sensitive to dust and Figure RC3 indicates they are positively correlated we would expect trends in Rsp to be consistent with the SAE trend analysis.

- Line 240: Light scattering in winter is quite low (according to Figure 3). What is the uncertainty in SAE during these months? Are the trends in the lowest percentiles reliable?
  Table RC2 provides uncertainty values for all the variables for both clean conditions and the upper scattering threshold (50 Mm^{-1}) used to identify extreme events. Fractional uncertainty in σ_{sp} & SAE during low scattering ($\sigma_{sp} \leq 1$ Mm^{-1}) are 10% and 6.3% respectively. So even though scattering is low in the winter the values with the highest uncertainty would not be larger than that given our filtering parameters.

  That said, we feel it's important to emphasize here that the lowest SAE values are not necessarily tied to the lowest values of the light scattering coefficients and therefore the largest uncertainties. The SAE depends on the difference in scattering between the two wavelengths used in the calculation. The lowest SAE values have the smallest difference between scattering at the two wavelengths, so the lowest percentiles simply correspond to the periods with the largest particles which can be periods of high loading/ scattering.

- Line 248: “Consistent attributable” appears to be a typo.
  Yes, ‘consistent’ has been removed.

- Line 275: These have been defined and the acronyms could be used here.
  Line 275: “scattering Ångström exponent and backscatter fraction” have been replaced by SAE and BFR

- Line 275: and also for fall for BFR.
  Line 275: The statement concerning the late summer increase in SAE / decrease in BFR has been extended to include the fall where this relationship also occurs:
  “As BFR and SAE are sensitive to different parts of the aerosol size distribution, the higher sub-

micron contribution (increased SAE) with increased large accumulation mode particles (decreased BFR) later in the summer and fall could indicate a narrowing of the size distribution (Collaud Coen et al., 2007)."

- Line 286: The acronym could be used here.
  Line 286: "single scattering albedo " has been replaced by SSA

- Line 298: What do the authors mean by "anthropogenic biomass burning"?
  Line 298: This was poor wording, this has been changed to "... mixture of fossil fuel, biomass burning, and BC dominated aerosol particles."

- Line 303: Figure S11 shows number of hours- number of hours of what?
  Line 303 / Fig. S11: Number of average hourly periods where AAE data was available over the entire sampling period. The figure caption has been updated to make this more clear: "Histograms of hourly AAE data, with the y-axis showing a total count of the number of hourly periods (# hours), for the spring and summer periods over all years at (a, c) SPL (2011-2024) and (b, d) BOS (2019-2024). Note that the y-axis is a log scale."

- Line 323: Can the authors explain why the particles have become less absorbing but there appears to be more smoke in the later period?
  A similar comment was made by Reviewer #1, and so we give a similar comment. Biomass burning (BB) from wildfires has been shown to be more scattering overall, with higher measured mass scattering efficiencies (MSE) compared to mass absorption efficiencies (MAE) leading to higher SSA values. This was shown during the Fire Laboratory at Missoula Experiments (Levin et. al., 2010; Mack et. al., 2010), and is consistent with measured MSE and SSA for BB sources under ambient conditions (Laing et. al., 2016). It's also been shown that SSA can increase as the age/ transport time of BB plumes increase. Using flights with repeated transects of BB plumes, Kleinman et. at. (2020) showed increases in both SSA and MSE for aged wildfire smoke. Selimovic et. al (2019) also observed increases in SSA as a function of time over a prolonged period of biomass burning. With these results we do expect the SSA to be high when the sites are measuring BB sources, even with the increase in BB / BC in the summer as indicated by the AAE. So with these results we do expect there to be increases in total scattering when BB is a major source.

Levin, E. J. T., McMeeking, G. R., Carrico, C., Mack, L., Kreidenweis, S. M., Wold, C. E., Moosmuller, H., Arnott, W. P., Hao, W. M., Collett, J. L., and Malm, W. C.:Biomass burning smoke aerosol properties measured during Fire Laboratory at Missoula Experiments (FLAME), J. Geophys. Res.-Atmos.,115, doi:10.1029/2009JD013601, 2010.

Laing, J. R., Jaffe, D. A., and Hee, J. R.: Physical and optical properties of aged biomass burning aerosol from wildfires in Siberia and the Western USA at the Mt. Bachelor Observatory, Atmos. Chem. Phys., 16, 15185–15197, https://doi.org/10.5194/acp-16-15185-2016, 2016.

Mack, L. A., Levin, E. J. T., Kreidenweis, S. M., Obrist, D., Moosmüller, H., Lewis, K. A., Arnott, W. P., McMeeking, G. R., Sullivan, A. P., Wold, C. E., Hao, W.-M., Collett Jr., J. L., and

Malm, W. C.: Optical closure experiments for biomass smoke aerosols, Atmos. Chem. Phys., 10, 9017–9026, https://doi.org/10.5194/acp-10-9017-2010, 2010.

Kleinman, L. I., Sedlacek III, A. J., Adachi, K., Buseck, P. R., Collier, S., Dubey, M. K., Hodshire, A. L., Lewis, E., Onasch, T. B., Pierce, J. R., Shilling, J., Springston, S. R., Wang, J., Zhang, Q., Zhou, S., and Yokelson, R. J.: Rapid evolution of aerosol particles and their optical properties downwind of wildfires in the western US, Atmos. Chem. Phys., 20, 13319–13341, https://doi.org/10.5194/acp-20-13319-2020, 2020.

Selimovic, V., Yokelson, R. J., McMeeking, G. R., and Coefield, S.: In situ measurements of trace gases, PM, and aerosol optical properties during the 2017 NW US wildfire smoke event, Atmos. Chem. Phys., 19, 3905–3926, https://doi.org/10.5194/acp-19-3905-2019, 2019.

- Line 325: Is the SPL site influenced by residential wood combustion in Steamboat?
  SPL's mountaintop location is more isolated from anthropogenic emissions in the valley during cold months because the thermal gradients driving upslope/downslope flow are not as strong (e.g., Collaud Coen et al., 2018). Thus SPL is less likely to sample smoke related to home heating in the seasons when residential combustion is most prevalent. A more likely source of absorbing aerosol are the ski area activities (snow mobiles and snow grooming vehicles) but those data are marked as contamination during the manual editing process and not included in the data analyzed here.

  Collaud Coen, M., Andrews, E., Aliaga, D., Andrade, M., Angelov, H., Bukowiecki, N., Ealo, M., Fialho, P., Flentje, H., Hallar, A. G., Hooda, R., Kalapov, I., Krejci, R., Lin, N.-H., Marinoni, A., Ming, J., Nguyen, N. A., Pandolfi, M., Pont, V., Ries, L., Rodríguez, S., Schauer, G., Sellegri, K., Sharma, S., Sun, J., Tunved, P., Velasquez, P., and Ruffieux, D.: Identification of topographic features influencing aerosol observations at high altitude stations, Atmos. Chem. Phys., 18, 12289–12313, https://doi.org/10.5194/acp-18-12289-2018, 2018.

- Line 377 and associated discussions of clusters: Please include the cluster number after each mention as it's easier to find on the plot.
  Line 377: Thank you for this suggestion. Numbers have been added

- Line 381: Figure S16: This seems like a step change for all seasons. Does this make sense that BC dominates all seasons and all seasons see this change?
  Line 381: We note in the text (Line 405) that given the distribution of the data and the decreases in the BC dominated group over all seasons, that it's likely that the background aerosol for the sites is also falling into this category even though the background aerosol is not necessarily BC dominated. As we state, this classification has limitations and there's clearly not a classification for all aerosol types. So when we consider all the data, as we do in Fig. S15 & S16, then the classifications become less definitive especially since we do not have chemical data to parse what all is encompassed in that grouping.

- Line 428/9: The acronyms for scattering can be used here.
  These instances have been replaced with the scattering symbol (σ_{sp}).

Figure Caption Comments:

- Figure 2: Add "a" and "b" in the caption. Include that these data in (a) and (b) correspond to RH< 40%.
  Fig 2: Added label to caption along with the following statement on RH: "The average σsp and σap values represent dry sampling conditions (RH < 40%)."

- Figure 3: Include the years in the caption.
  Fig 3: done

- Figure 4: Provide years for trends and include RH < 40%. Here the significance is given by alpha but the rest of the text it is referred to as a p-value (like line 167).
  This has been noted in the caption, and significance has been put in terms of the p-value:
  "Quantile regression trends for PM10 (a) σsp and (b) SAE at SPL over all seasons from 2011 to 2024. Winter = Dec – Feb, Spring = March - May, Summer = June - Aug, Fall = Sept - Nov. Open markers show trends that were not significant (p-value ≥ 0.05). Numerical values and p-values for the quantile regression trends are provided in Table S5 & S6. As with other analyses presented, these trends are for dry sampling conditions (RH < 40%)."

- Figure 1: Change S14 to S12.
  There is no reference to Fig S14 in Fig 1 or Fig S1. We request clarification on this comment before making changes.

- Figure S7: Include the location in the caption.
  Fig S7: done

- Figure S8: Include (a), (b), etc in the caption. What is the difference between a-b and c-d?
  Fig S8: Apologies for not including this distinction originally. Panels (a) & (b) show data for the entire Rocky Mountain Area – including Colorado, Kansas, Nebraska, South Dakota, and Wyoming – while (c) & (d) show data only for Colorado. This has been added to the caption.

- Figure S11: Captions states "histogram" but axis reads "# of hours" ?
  Fig S11: Yes, the distribution is not normalized in any way so the axis is just a total count of the hourly averaged periods in the spring and summer periods over the entire date range (2011 - 2024 for SPL and 2019 - 2024 for BOS) where AAE was in each bin range.

  To make this more clear the caption has been changed to: "Histograms of hourly AAE data, with the y-axis showing a total count of the number of hourly periods (# hours), for the spring and summer periods over all years with measurements at (a, c) SPL (2011-2024) and (b, d) BOS (2019-2024). Note that the y-axis is a log scale."