RC1: Author 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. A significant effort was made to identify these breakpoints in the SPL data and correct them before trend analysis was performed. "

- 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, 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.

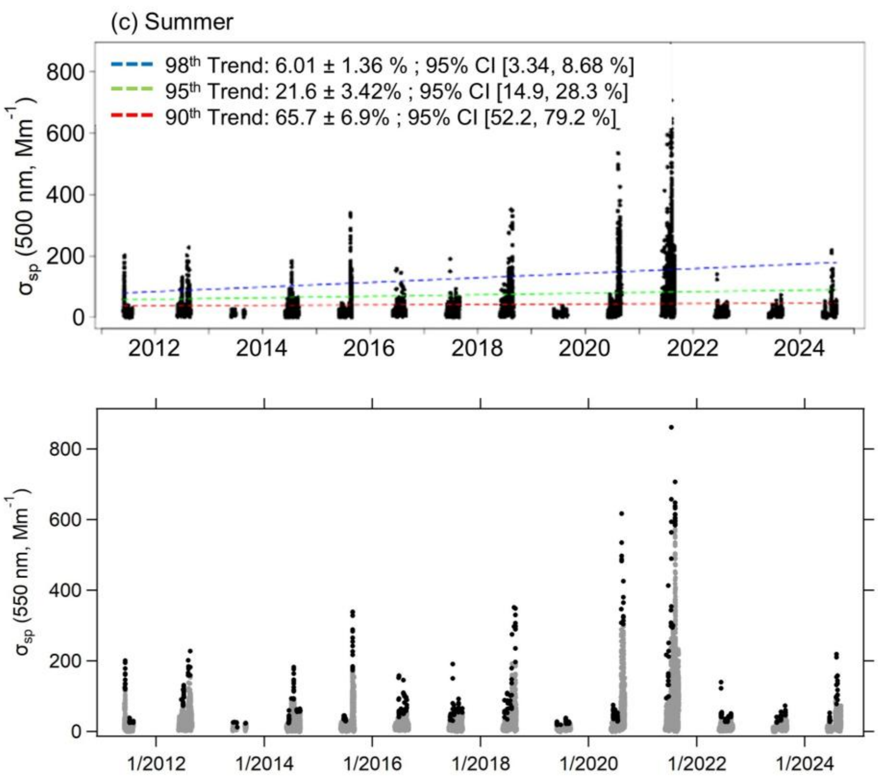

Figure RC1: Fig. S7a from the supplemental section along with a plot of hourly PM10 scattering data with 98th percentile values shown in black with all other data shown in grey.

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

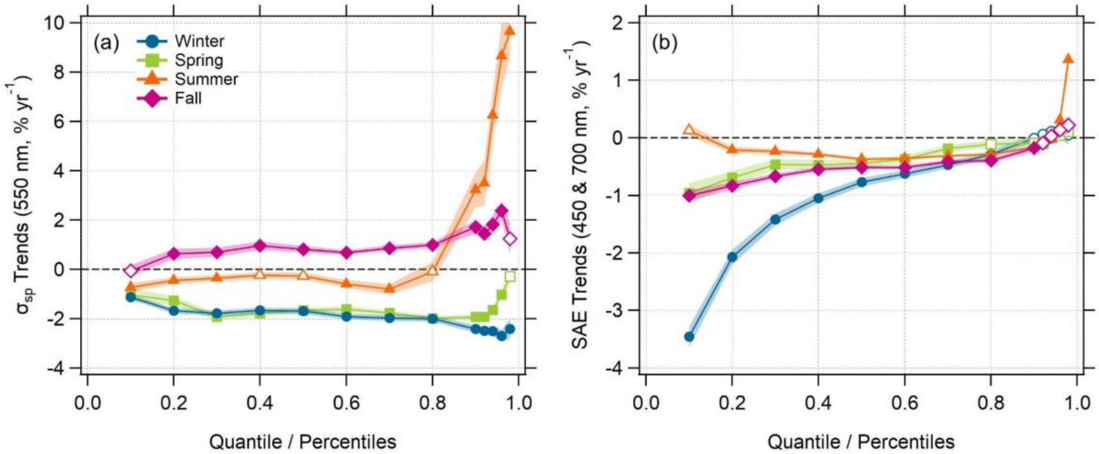

Fig. 4: Quantile regression trends for PM10 (a) σsp and (b) SAE at SPL over all seasons from 2011 to 2024. Winter = Dec – Feb, Spring = March - May, Summer = June - Aug, Fall = Sept - Nov. Open markers show trends that were not significant at (p-value ≥ 0.05). Numerical values and p-values for the quantile 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.

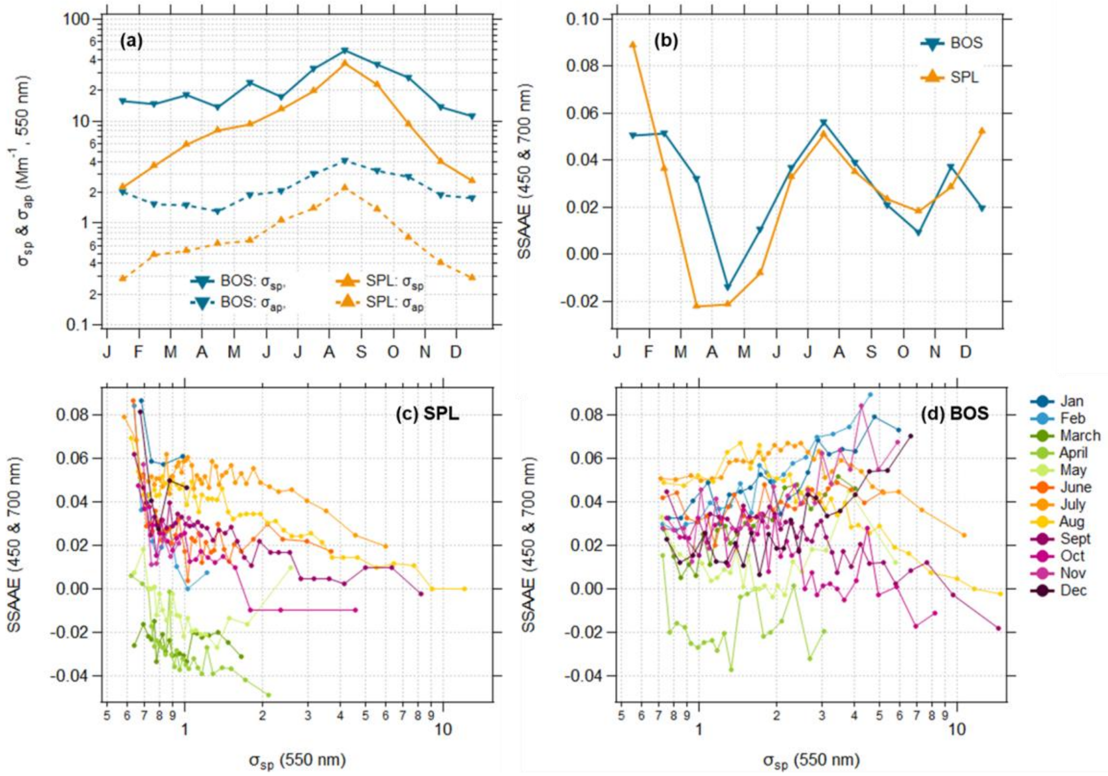

Fig RC2: (a) Seasonal cycles of median PM_{10} σ_{sp} and σ_{ap} (550 nm) for both SPL and BOS. (b) Monthly median PM_{10} SSAAE for 450 & 700 nm for both SPL and BOS. PM_{10} SSAAE (450 & 700 nm) binned by σ_{sp} (550 nm) for (a) SPL and (b) BOS. For all plots, each data point represents the median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

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.