



Climatology of Aerosol Optical Properties at Storm Peak Laboratory

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ABSTRACT

Aerosols create large uncertainty in the planetary energy balance due to both direct and indirect radiative forcing. Understanding aerosol seasonal patterns is essential for accurate climate change prediction, but mountain regions are often difficult for climate models to resolve. Therefore, long-term observations collected at high elevations are particularly useful. In-situ surface aerosol optical measurements were analyzed for the years 2011–2016 at a mountain site located in western Colorado and tied to potential sources based on relationships among the aerosol properties.

The peak values for the scattering and absorption coefficients were observed during the summer, suggesting greater aerosol loading (likely due to wildfires), whereas the lowest values were observed during the winter, indicating cleaner conditions (due to less influence from the boundary layer). The scattering Ångström exponent, a property that provides information about size distributions, revealed the predominance of coarse-mode particles during the spring, which is consistent with the presence of dust. The aerosols observed during the summer, however, were mostly composed of fine-mode particles. This increase in the fine fraction points to combustion, likely wildfires during the dry season (Hallar, 2015), as a source, which is further supported by the absorption Ångström exponent dropping to its lowest value (close to 1) during the summer after exhibiting a slightly higher value (~1.3) during the spring. Schmeisser *et al.* (2017) suggests that, for in-situ aerosol, absorption Ångström exponents larger than 1.5 may be indicative of dust if they are associated with low (< 1.3) scattering Ångström exponents. The increase in combustion aerosols during the summer accompanied by high values for the single scattering albedo suggests that these aerosols underwent processing in the atmosphere before reaching Storm Peak Laboratory. These results are important for improving visibility and predicting future aerosol concentrations in the western U.S.

Keywords: Absorption; Scattering; In-situ measurements; Mountain; Climate.

INTRODUCTION

Atmospheric aerosols change rapidly over short time intervals, making future concentrations difficult to predict (Laj *et al.*, 2009). Establishing aerosol climatology is important for identifying aerosol sources, distributions, and transport (Yu *et al.*, 2009; Hand *et al.*, 2017). By revealing seasonal aerosol patterns of a region, improvements can be made to climate models (Chung *et al.*, 2005) and visibility by identifying aerosol sources (Hirdman *et al.*, 2010). There is large disagreement among climate models related to aerosol spatial distributions and the direct and indirect

radiative forcing effects of these aerosols (Reddington *et al.*, 2017).

Aerosols absorb and scatter radiation and thus have a direct effect on the planetary energy balance. These effects can be quantified by calculating direct radiative forcing from measured aerosol optical properties. Direct aerosol radiative forcing is dependent upon particle composition, size, and concentration (Haywood and Boucher, 2000). The current estimate from the Intergovernmental Panel on Climate Change of direct aerosol radiative forcing is $-0.35 \pm 0.5 \text{ W m}^{-2}$ (Stocker *et al.*, 2013). The large uncertainty associated with this value is a consequence of rapidly changing aerosol distributions with respect to time and location (Stocker *et al.*, 2013), model uncertainties (Reddington *et al.*, 2017), and varied aerosol composition (Jacobson, 2001).

Aerosols also influence the radiation balance by indirect mechanisms. Due to their role as potential cloud condensation nuclei (CCN), aerosols can change the energy balance by

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altering cloud properties. When CCN concentrations are high, clouds will be composed of more droplets of smaller diameter and thus have different optical properties (Twomey, 1977). Clouds composed of smaller droplets can persist longer due to delayed precipitation, resulting in further changes to the radiation balance (Albrecht, 1989).

Feedbacks that influence future aerosol climatology are another important consideration for numerical model accuracy. For example, wildfires are a common source of atmospheric aerosols during the summer months and are expected to increase as the climate warms (Spracklen *et al.*, 2009). Increased aerosol loading leads to diminished incoming solar radiation at the surface due to light extinction (Charlson *et al.*, 1992). Decreased solar radiation has been linked to milder summer monsoons in the southwestern U.S. (Diffenbaugh *et al.*, 2006). A strong correlation between drought and wildfire has also been observed (Westerling *et al.*, 2006), ultimately creating a positive feedback for aerosol loading and additional cooling of the climate system. Using numerical models, Spracklen *et al.* (2009) projected a 40% increase in organic carbon due to increased wildfire activity by the year 2050. Hallar *et al.* (2017) estimated this value to be lower, at 24% for the Colorado Rockies and 36% for the Wasatch and Uintah Mountains, an important consideration for numerical climate models since aerosols moderate warming. The net effect of these mechanisms is unclear due to competing feedbacks that are not well understood or quantified.

Previous studies and climate models relied heavily on aerosol optical depth (AOD) in order to observe aerosol loading and transport. AOD is an integrated measurement of aerosols throughout the entire atmospheric column that is measured using satellite or ground-based radiometers. Remote sensing measurements are particularly valuable because they can gather aerosol data over large horizontal areas and represent total vertical aerosol concentration (Chung *et al.*, 2005; Wang *et al.*, 2012). While useful, these measurements lack information regarding the vertical distribution of aerosols within the column. The vertical distribution has an effect on radiative forcing (Sanroma *et al.*, 2010) and can also provide information about aerosol transport and sources. For example, greater AODs have been observed during the spring with the transport of Asian dust to North America by extratropical cyclones (Yu *et al.*, 2008). Hallar *et al.* (2015) observed that, at their mountain site in Colorado, in-situ measurements made at the surface did not spike in the spring with AOD measurements, suggesting the aerosols contributing to the seasonal increase in AOD were located above the surface.

Surface in-situ measurements can provide additional aerosol information using different methodologies than those used to find AOD. Unlike remote sensing measurements, for in-situ measurements the air is sampled within instruments to obtain the aerosol absorption and scattering coefficients. This approach allows continuous data to be obtained at the surface at a specific location regardless of atmospheric conditions (e.g., clouds or nighttime). For some aerosol properties, such as absorption and single scattering albedo, remote sensing techniques can have high uncertainty,

emphasizing the importance of in-situ measurements (Dubovik, 2000). Combining remote sensing with in-situ measurements provides more comprehensive information for improving direct radiative forcing estimates (Andrews *et al.*, 2004; Yu *et al.*, 2009).

While global values of radiative forcing have large uncertainty, regional estimates are even more variable (Yu *et al.*, 2009). Gathering long-term in-situ data from various sites is one way to reduce this uncertainty (Andrews *et al.*, 2011; Pandolfi *et al.*, 2018). Mountain regions create unique difficulties for climate models due to complex terrain and insufficient model resolution (Giorgi, 2005). Therefore, data collected at high elevations will be especially valuable for improving climate models, particularly as the spatial resolution of the models improves.

In the following sections, in-situ aerosol optical property data collected from 2011 to 2016 at a high elevation site in the western U.S. are analyzed to identify seasonal aerosol trends. Scattering and absorption coefficients are used to calculate a variety of aerosol optical properties that can help to establish the aerosol climatology of the region and illuminate the possible sources of these aerosols.

METHODOLOGY

Location

Storm Peak Laboratory (SPL) is a mountain-top site (3220 m) located in western Colorado (40.455°N, 106.745°). Due to its remote location, SPL has little influence from traffic or industry of nearby cities (Hallar *et al.*, 2016) and has been used for aerosol research since the 1990s. All data for this project was collected at SPL and went through a quality assurance process to remove spikes due to local contamination (e.g., ski area vehicles) as well as periods when the instruments were not working or being maintained. The edited data was submitted to the EBAS Level 2 data archive (<http://ebas.nilu.no>). Measurements were taken in 1-minute intervals, but Level 2 data were reported as hourly averages. The Level 2 data were also reported at standard temperature (273.15 K) and pressure (1013.25 hPa), and the appropriate instrument corrections were applied as described below.

Instrumentation

The scattering data were collected using a TSI 3563 nephelometer that measures the scattering coefficient, σ_{sp} , at 3 wavelengths (λ ; 450 nm, 550 nm, and 700 nm). The nephelometer data were corrected for instrument angular truncation and light source non-idealities (Anderson and Ogren, 1998). A particle soot absorption photometer (PSAP) was used to measure the absorption coefficient, σ_{ap} , at slightly different visible wavelengths (467 nm, 530 nm, and 660 nm). Data from the PSAP was replaced in 2013 by data from a similar instrument known as a continuous light absorption photometer (CLAP; wavelengths: 467 nm, 528 nm, and 652 nm). This instrument measures absorption in the same manner as the PSAP but contains 8 filter spots (compared to 1 filter spot for the PSAP) through which it rotates before filter replacement is required (Ogren *et al.*,

2017). This is advantageous because SPL is a remote site with infrequent technician visits. PSAP and CLAP data were corrected using the schemes in Bond *et al.* (1999) to account for scattering artifacts, spot size, and flow, and Ogren (2010) to extend the Bond corrections for all three wavelengths. Data where the filter transmittance was less than 0.5 were automatically marked as invalid during the quality control process because of limitations of the Bond correction.

A switched impactor system separated aerosols into particulate matter less than 10 μm (PM_{10}) and less than 1 μm (PM_1) in diameter before measurements were made by the nephelometer and PSAP/CLAP. For measurements of this type, the sample air is often dried to achieve a relative humidity of 40% or less to avoid the confounding effects of water on aerosol properties. Because SPL is located in an arid region and the measurements were made within a heated building, relative humidity values above 40% were rare (Hallar *et al.*, 2015), eliminating the need for this step.

Calculations

The measured scattering and absorption coefficients offer information about aerosol loading at the surface. These values can also be used to calculate various intensive aerosol optical properties (intensive properties are independent of the amount of aerosol loading). The scattering Ångström exponent and absorption Ångström exponent can be calculated using measurements made at two wavelengths: $\alpha_x = -\log(\sigma_{\text{xp}}(\lambda_1)/\sigma_{\text{xp}}(\lambda_2)/\log(\lambda_1/\lambda_2))$, where $\sigma_{\text{xp}}(\lambda_i)$ is the scattering (s) (or absorption (a)) at wavelength i . The scattering Ångström exponent (α_s) can be used as an indicator of particle size, with low values corresponding to particles with greater diameter and higher values indicating smaller particles. Clarke and Kapustin (2010) experimentally obtained a threshold ($\alpha_s = 1.3$) for the scattering Ångström exponent (450 nm/700 nm) that differentiates between fine-mode aerosols ($\alpha_s > 1.3$) and coarse-mode aerosols ($\alpha_s < 1.3$). The absorption Ångström exponent (α_a) can provide information about the composition of aerosols (e.g., Bergstrom *et al.*, 2002, 2007). Black carbon has a theoretical value of $\alpha_a = 1$, while dust aerosol α_a tends to be larger than 2, and there is no clear consensus on typical values for biomass burning (Table 1 in Schmeisser *et al.*, 2017, and references therein). The single-scattering albedo (SSA) is the ratio of the scattering coefficient to the extinction coefficient (the sum of absorption and scattering). SSA is high (close to 1) for aerosols that readily scatter light and lower for aerosols that strongly absorb light (black carbon has $\text{SSA} \approx 0.3$), providing additional information about aerosol type.

RESULTS AND DISCUSSION

Fig. 1(a) shows the monthly means of scattering coefficients for PM_1 and PM_{10} at each wavelength (450 nm, 550 nm, and 700 nm). In all cases, the highest values are found during the summer months, with the peak in August. The lowest values occur during the winter months, when air is cleaner due to SPL being less influenced by the atmospheric boundary layer, which is generally the case

for mountain sites (Raatikainen *et al.*, 2014). The rate of change from spring to summer is more gradual than from summer to fall. It is probable that the spike in aerosol scattering during the summer is due to biomass burning during peak wildfire season (Hallar *et al.*, 2015).

Monthly averages for absorption coefficients are shown in Fig. 1(b). The PSAP or CLAP absorption data were adjusted to 450 nm, 550 nm, and 700 nm using the absorption Ångström exponent. This was done to accommodate calculation of SSA, which requires both absorption and scattering coefficients at the same wavelength. The absorption data follows a similar pattern to the scattering data, with the highest monthly values found during the summer season and lowest values during the winter months. As with scattering, the slope of the curve from spring to summer is not as steep as the slope from summer to fall.

While aerosol loading is an important factor for radiative forcing, information about the aerosol size distribution and composition can aid in attributing aerosol forcing to different sources. For example, natural particles such as dust tend to dominate the coarse mode (diameter $> 1 \mu\text{m}$), while anthropogenic and combustion aerosol are more dominant in the fine mode (diameter $< 1 \mu\text{m}$). When calculating the scattering Ångström exponent, an additional constraint on the scattering data was applied. Similar to Andrews *et al.* (2011), scattering values less than 0.5 Mm^{-1} were omitted when calculating the scattering Ångström exponent with the purpose of minimizing noise that can occur when taking the ratio of two small numbers. This method eliminated 7.45% of the scattering data for PM_1 and 6.69% of the data for PM_{10} . Fig. 2(a) shows the monthly averaged scattering Ångström exponent for PM_1 and PM_{10} . The scattering Ångström exponent is lowest during the spring months, particularly in March when the mean value drops to 1.3, indicating the presence of large particles at the surface; these particles are likely dust (Hallar *et al.*, 2015), although whether they are from regional sources or due to long-range transport is unclear. During the summer months, the scattering Ångström exponent is at its highest (~ 2 for PM_{10}), corresponding to a greater contribution from small particles. This is likely due to increased combustion aerosols during peak wildfire activity (Hallar *et al.*, 2015).

The monthly averaged absorption Ångström exponent is presented in Fig. 2(b). The α_a at SPL exhibits less seasonal variation than other aerosol optical properties, ranging only between 1 and 1.3 throughout the year. The lowest α_a (near 1) occurred during the summer months. The highest values for α_a occurred during spring season. Some winter months also exhibit higher values for α_a ; however, SPL is an extremely remote site, with the cleanest conditions occurring during the winter months, which can lead to noise in the α calculation. The springtime values for α_a found at SPL are lower than typically suggested for dust (e.g., Table 1 in Schmeisser *et al.*, 2017); however, the monthly α_a values represent all data for that month, not just during dust events, which tend to be sporadic and short-lived (less than 1, to 5 days; Hallar *et al.*, 2015).

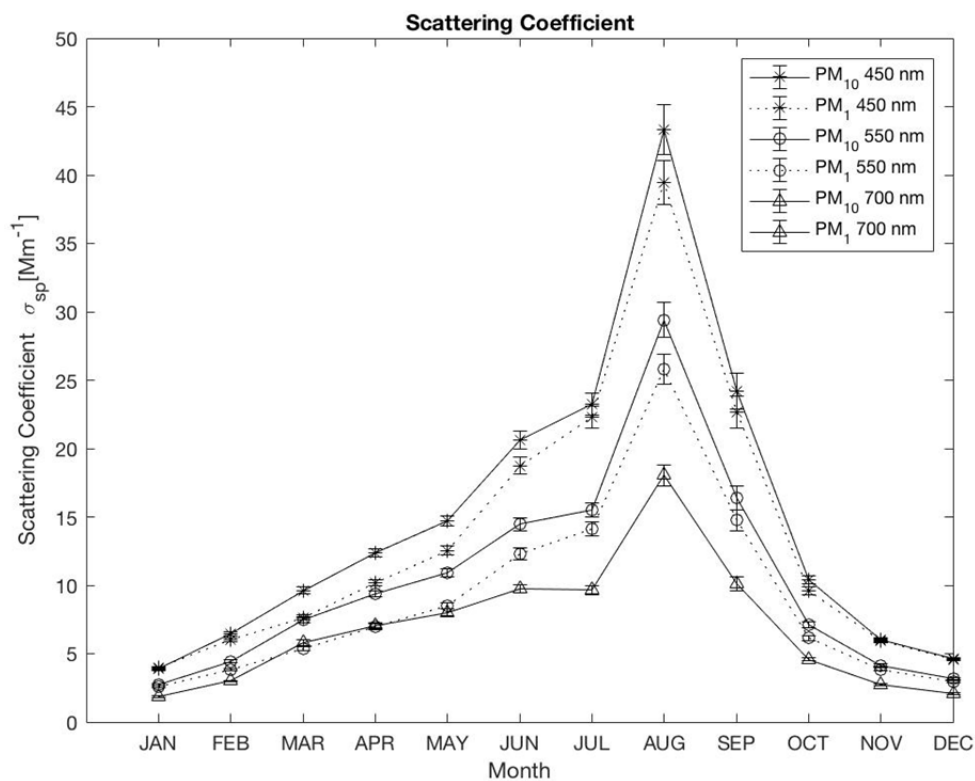


Fig. 1(a). Mean monthly scattering coefficients for years 2011–2016. Measurements at 450, 550, and 700 nm are represented by asterisk, open circle, and triangle markers, respectively. Solid lines are PM₁, and dashed lines are PM₁₀. The 95% confidence intervals of the monthly means are displayed using error bars.

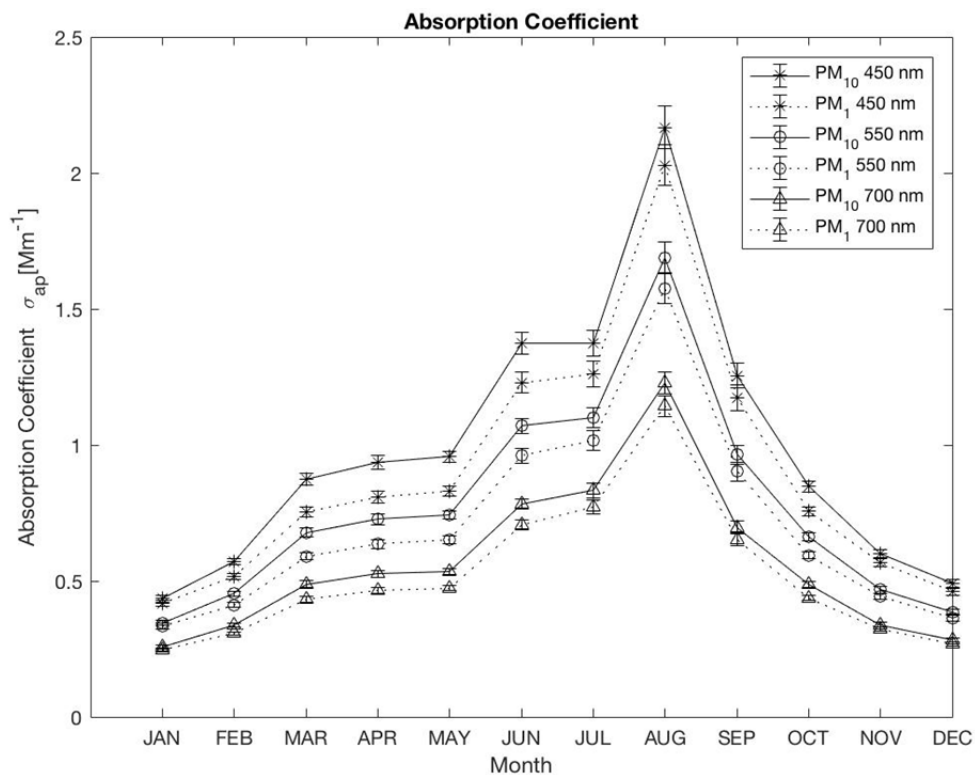


Fig. 1(b). Mean monthly absorption coefficients for years 2011–2016. Measurements at 450, 550, and 700 nm are represented by asterisk, open circle, and triangle markers, respectively. Solid lines are PM₁, and dashed lines are PM₁₀. The 95% confidence intervals of the monthly means are displayed using error bars.

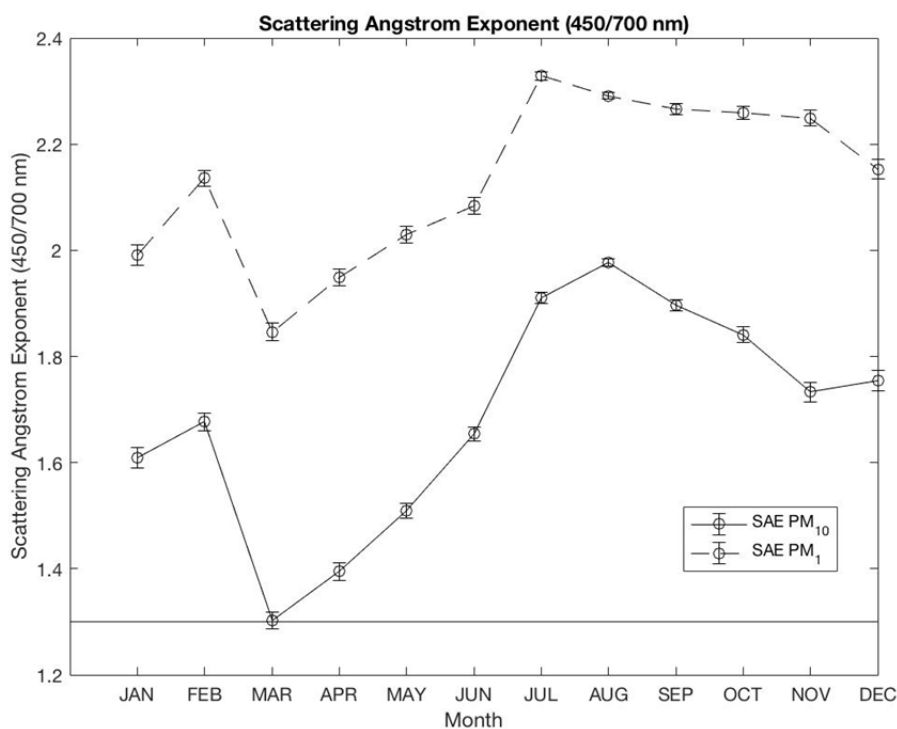


Fig. 2(a). Mean monthly scattering Ångström exponents (400/700 nm). The solid line represents PM₁₀ and dashed line, PM₁. The 95% confidence intervals of the monthly means are displayed using error bars.

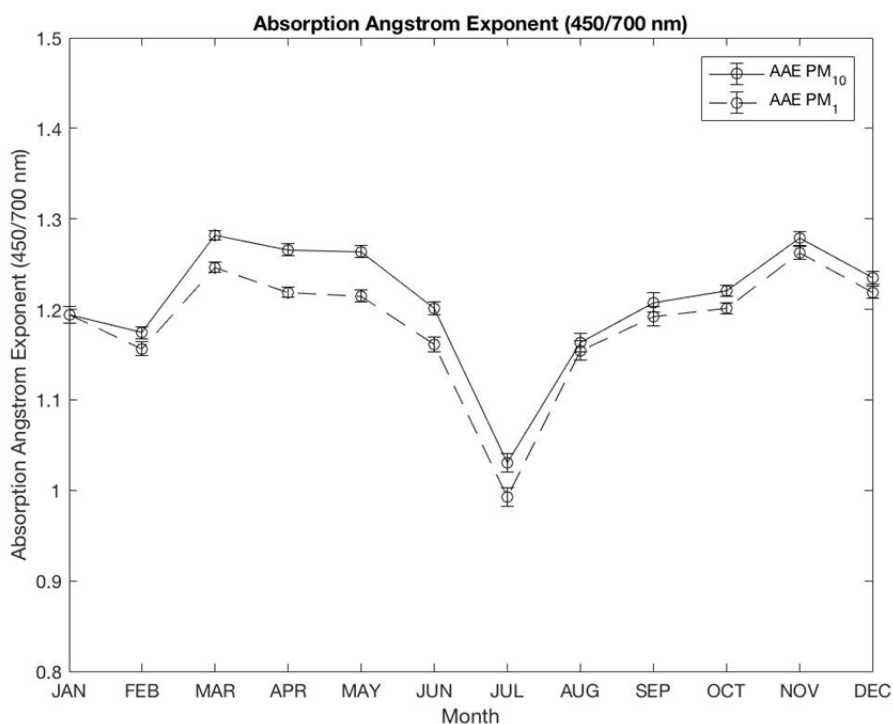


Fig. 2(b). Mean monthly absorption Ångström exponents (400/700 nm). The solid line represents PM₁₀ and dashed line PM₁. The 95% confidence intervals of the monthly means are displayed using error bars.

Fig. 3 shows a plot of the scattering Ångström exponent (450 nm/700 nm) versus scattering coefficient at 550 nm for PM₁₀. The scattering Ångström exponent was calculated for each hourly data point for the years 2011–2016. The black

dots represent all data, green dots represent data collected during the peak dust season days (April 1–May 15) at SPL (Hallar et al., 2015), and red dots represent values from the summer season defined as days (June 19–August 13).

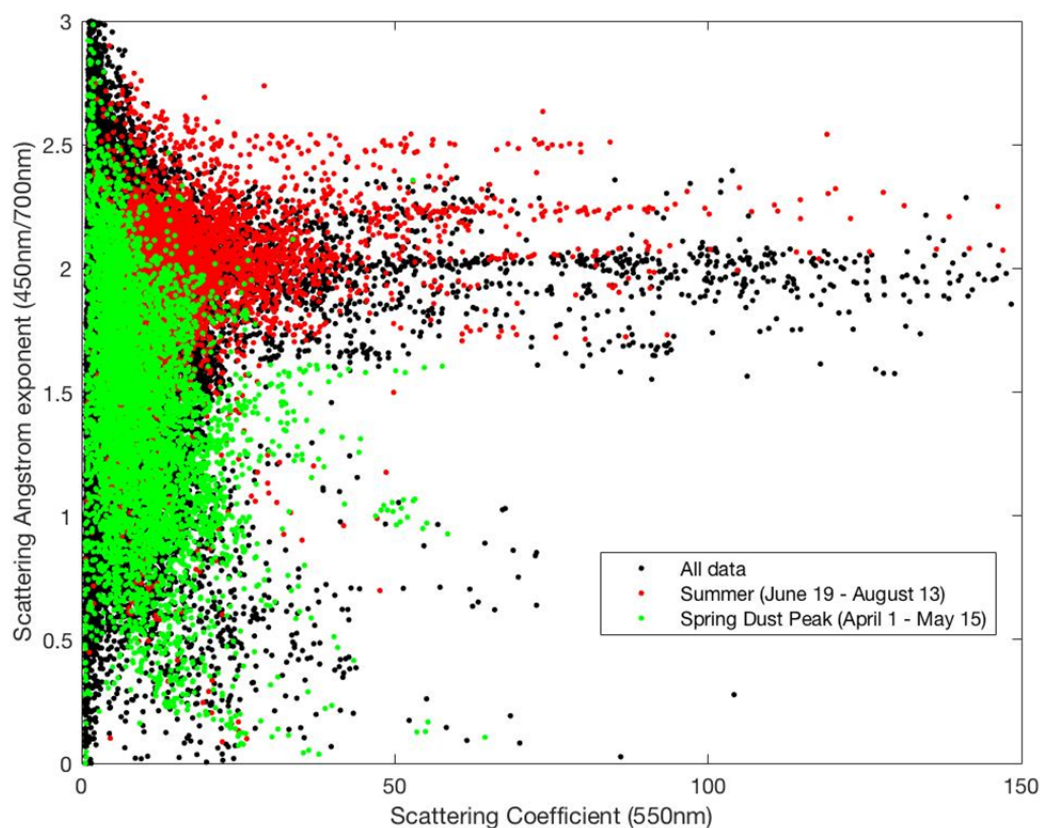


Fig. 3. Ångström exponent (450/700 nm) vs. scattering coefficient at 550 nm for years 2011–2016. Black dots represent all data. Green dots occur during peak dust season (Hallar *et al.*, 2015) from April 1–May 15 for all years. Red dots represent summer season for days June 19–August 13 of all years.

By comparing these two quantities, a relationship between aerosol loading and aerosol size with respect to season can be discerned. Fig. 3 shows two major aerosol categories. One group consists of small size particles and high aerosol loading, located in the top portion of the graph, and is associated largely with the summer months (combustion aerosols). The summer season is shown with red points in Fig. 3, and these points are primarily found with a scattering Ångström exponent greater than 1.5. Data collected during the Spring, found primarily on the lower portion of the plot, is composed of mostly coarse-mode particles (dust), as demonstrated with the lower scattering Ångström exponent. Additionally, a lower scattering coefficient was observed during the Spring season. Hallar *et al.* (2015) presented similar results; however, nephelometer and PSAP measurements in that study were limited to the years 2011–2013, compared to 2011–2016 for this project. In Hallar *et al.* (2015), aerosol optical depth values combined with surface level nephelometer data suggested that high altitude long-range transport may be primarily responsible for the strong dust signal observed at SPL in the spring. The expanded dataset presented here suggests otherwise, as there is a strong dust signal observed at the surface that is apparent from the decrease in in-situ scattering Ångström exponent during the spring.

The single scattering albedo for the 450 nm, 550 nm, and 700 nm wavelengths are shown in Fig. 4. SSA was

highest during the spring and summer (with the exception of June) and lowest during the winter season. We hypothesize that the seasonal variability of SSA is due to the impact of wildfires in the late spring and summer, along with an increase in emissions from home heating via wood burning during the winter months (Zhao *et al.*, 2013).

While Fig. 3 suggests that the summer months are associated with wildfires (i.e., combustion aerosol), the SSA observed during the summer is quite high. This has previously been observed for long-range transport of smoke and is possibly associated with the condensation of gases from wildfire smoke, which can enhance the scattering of aerosol (Andrews *et al.*, 2004).

Though dust is observed at SPL in the spring, it is difficult to identify specific dust sources without comparison to known dust events. While infrequent, Asian dust has previously been detected at SPL, though, as noted above, it was typically above the surface (Hallar *et al.*, 2015). Hallar *et al.* (2015) also observed a number of dust events originating from regional (inter-mountain west) sources during the spring months. In the future, the incorporation of meteorological conditions may also yield important information regarding dust sources. Different methodologies, including aerosol chemistry, could also be utilized to identify aerosol types with higher specificity and thus help identify aerosol sources.

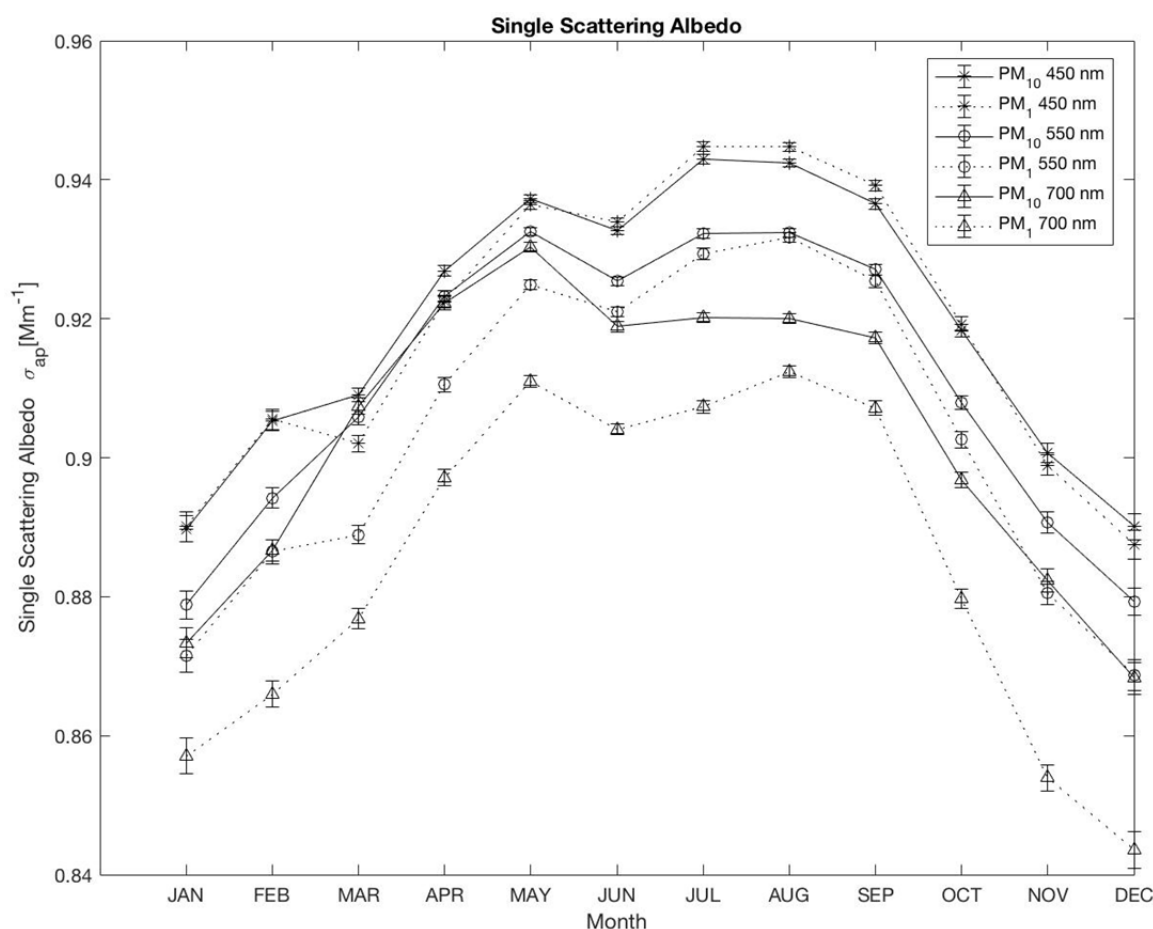


Fig. 4. Single scattering albedo at 450 nm (asterisk), 550 nm (open circle), and 700 nm (triangle). The solid lines represent PM₁₀, and the dashed lines represent PM₁. The 95% confidence intervals of the monthly means are displayed using error bars.

CONCLUSIONS

The identification of aerosol seasonal patterns and sources in the western U.S. is crucial to improving visibility and climate model accuracy. In this study, the annual cycles for aerosol optical properties (the absorption, scattering, single scattering albedo, and absorption and scattering Ångström exponents) were presented and tied to known aerosol types. A strong wildfire signal was observed in the summer at SPL, yet these events were not associated with a significant decrease in SSA. These combined measurements suggest that wildfire smoke is aged and processed, reducing the direct radiative forcing impact, as the smoke is transported from the initial source.

Additionally, the aerosol climatology based on in-situ measurements at SPL presented by Hallar *et al.* (2015) has been updated to include the years 2013–2016. A notable new finding is the detection of a strong dust signal at the surface by in-situ measurements, whereas previous comparisons of the column and surface data suggested that most of the springtime dust remained above the surface. These results point to the impacts of regional climate change on visibility with an increasingly arid environment in the western U.S. (Hallar *et al.*, 2017).

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