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Drought Impacts on $PM_{2.5}$ Composition and Amount Over the US During 1988–2018

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Key Points:

- The increase in $PM_{2.5}$ in the northwestern US in summer is mainly due to the enhancement of total carbonaceous aerosols (TCAs) related to droughts
- Droughts affect TCA variations over the northwestern US mainly through its impact on wildfires
- The impacts of droughts on species concentrations in other regions and seasons are not sufficient to change $PM_{2.5}$ trends

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract

This study uses $PM_{2.5}$ species concentrations from Interagency Monitoring of Protected Visual Environments and Standardized Precipitation Evapotranspiration Index (SPEI) during 1988–2018 over the continental US to investigate the association of spatial-temporal variations of surface $PM_{2.5}$ species with droughts. Ubiquitous decreasing trends in seasonal mean reconstructed $PM_{2.5}$ are detected in all seasons except for the summer over the northwestern US. The increasing trend in the reconstructed $PM_{2.5}$ in summer over northwestern US is primary due to the positive trend in total carbonaceous aerosols (TCAs), which more than offsets the negative trends in sulfate and nitrate aerosols. This also causes the contribution of TCA to the reconstructed $PM_{2.5}$ to show an increasing trend in summer over the same region. The positive trend in TCA in summer over northwestern US is stronger under the drought than the wet conditions, hence resulting in a stronger positive trend in the contribution of TCA to the reconstructed $PM_{2.5}$ in the drought conditions. Drought enhances TCA over northwestern US through its impact on wildfire, and the temporal change of TCA mainly follows the non-linear variation of SPEI. Although droughts are found to have statistically significant impacts on the trends in sulfate, nitrate, and dust in other regions and seasons, the small contributions of these species to the reconstructed $PM_{2.5}$ make their trend variations not sufficient to affect trends in the reconstructed $PM_{2.5}$.

Plain Language Summary

Ubiquitous decreasing trends in surface mass concentrations of fine aerosol particles are observed in the past ~30 years in all seasons and regions except for the northwestern US in summer. The increasing trend in fine aerosol particle concentrations over the northwestern US in summer during 1988–2018 is primarily due to the positive trend in total carbonaceous aerosol concentrations associated with large fires in drought years. This positive trend is greater than the decreasing trends in sulfate and nitrate aerosols. As the frequency, duration, and spatial converge of droughts are expected to increase in a warming climate, the drought-induced air pollution through wildfire cannot be overlooked and could partly offset the benefit from reduction of anthropogenic emissions. To circumvent the model deficiencies of predicting wildfires, drought index has the potential to be a predicator for forecasting long-term trend of air pollution in future.

1. Introduction

Fine particulate matter (aerodynamic diameter ≤2.5 μm; $PM_{2.5}$) is recognized as a criteria pollutant by the US Environmental Protection Agency (EPA) and has adverse impacts on human health (Lelieveld et al., 2015). $PM_{2.5}$ mainly consists of ammonium, sulfate, nitrate, Black Carbon (BC), Organic Matter (OM), and fine dust aerosols. Carbonaceous aerosol, including BC and OM, has stronger toxicity than the rest of the $PM_{2.5}$ species (Lelieveld et al., 2015; Tuomisto et al., 2008). $PM_{2.5}$ concentrations over most US regions show negative trends in the range of −51% to −18% from 2000 to 2020 except for the northwestern US, where an increase of 6% exists (EPA, 2022). The reduction of carbonaceous and sulfate aerosols due to the control of anthropogenic emissions greatly contributes to the decreasing trends in $PM_{2.5}$ concentrations (Leibensperger et al., 2012; Malm et al., 2017; McClure & Jaffe, 2018; Murphy et al., 2011; Ridley et al., 2018). The positive trend in $PM_{2.5}$ over the northwestern US mainly exists in summer, and is thought to be largely attributed to the increase in wildfire carbonaceous aerosol emissions (McClure & Jaffe, 2018; O'Dell et al., 2019), although these studies only used data primarily collected during 1995–2016 (O'Dell et al., 2019) or confined to quantile trend analysis (McClure & Jaffe, 2018). Furthermore, the change of various $PM_{2.5}$ species and wildfires are both amenable to the change of weather patterns (Y. Wang et al., 2017), and from the observation-based point of view, it remains unclear how droughts, a major driver

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for the wildfires, may affect $PM_{2.5}$ composition and amount in different seasons. Establishing this association is important because the climate models still lack the processes or fidelity to predict the occurrence of fires and the change of $PM_{2.5}$ heretofore.

Over the past three decades, wildfire-burned areas show an increasing trend (CRS, 2021; Singleton et al., 2019), which is mainly linked to warmer temperatures (Westerling et al., 2006; Williams et al., 2014), lower atmospheric moisture content (Williams et al., 2014), and less precipitation (Holden et al., 2018). All these drivers are characteristics of drought and can be linked well with the drought index (Beguería et al., 2014). $PM_{2.5}$ from the drought-induced wildfire emissions can significantly increase children hospitalizations related to respiratory diseases (Smith et al., 2014). Hence, it is important to investigate the impact of droughts on the variations of aerosol concentrations from wildfires. Hallar et al. (2017) showed that the drought-related wildfires result in the increase of aerosol loading in the intermountain western US. Y. Wang et al. (2015) demonstrated that the increase of Organic Carbon (OC, the measurement of carbon associated with OM) emissions from wildfires is the major contributor to the enhanced $PM_{2.5}$ over the southern US during the 2011 drought from the perspectives of observation and simulation. Y. Wang et al. (2017) found the enhancement of OC over the US under the drought condition and ascribed it to the increase in OC emissions from wildfires through analyzing Global Fire Emission Database. However, quantitatively, how the frequency and severity of drought affect the variations of total carbonaceous aerosols (TCAs; OM and BC) and $PM_{2.5}$ mass remains elusive, especially from a long-term observation perspective. This is complicated by the possible impacts of drought on other $PM_{2.5}$ species (Y. Wang et al., 2017) in addition to wildfire-induced TCAs. Additionally, the temporal variation of drought can be divided into a linear part and a non-linear part (Y. Wang et al., 2021); their influence on the TCAs is also unknown. As more droughts are expected to increase from global warming (Cook et al., 2015; Y. Wang et al., 2017), analysis of historic data can help establish the benchmarks that are needed for better predicting future air quality of $PM_{2.5}$ and TCAs.

TCAs are not only from wildfires, but also from anthropogenic combustion and the formation of secondary organic aerosol, whose formation process and precursors from biogenic sources are both affected by drought events (Zhao et al., 2019). To investigate the impact of drought on aerosols, it is important to distinguish TCAs from different sources. As non-soil potassium in smoke particles is a good indicator of wildfire activity (J. Wang et al., 2006; Kreidenweis et al., 2001), this study will analyze the covariation between non-soil potassium, TCAs, and other $PM_{2.5}$ species, and link their temporal variation with drought events. We introduce data and method in Section 2. Section 3 presents the relationships among droughts, wildfires, and $PM_{2.5}$ species concentrations. Discussion and conclusions are included in Sections 4 and 5, respectively.

2. Data and Method

2.1. IMPROVE Aerosol Concentrations

Mass concentrations of surface $PM_{2.5}$ species are routinely measured by the Interagency Monitoring of Protected Visual Environments (IMPROVE) network. As the ammonium concentrations are not analyzed by IMPROVE, $PM_{2.5}$ concentrations are calculated as the sum of sulfate, nitrate, BC, OM, and fine dust (aka fine soil); we call it the reconstructed $PM_{2.5}$. OM mass concentrations are obtained through multiplying OC mass concentrations by 1.8 (Bae et al., 2006) to calculate the reconstructed $PM_{2.5}$. Potassium (K) in smoke particles is an indicator of aerosols from wildfires, but the sources of K in the atmosphere include not only smoke but also soil. In the study, the technique of Kreidenweis et al. (2001) is used to estimate the mass of smoke K from non-soil sources, which is equal to the total mass of K minus 60% mass of Fe. The same technique is also used in J. Wang et al. (2006) that showed the mass concentration of non-soil K tracks well the smoke concentration from model simulation. Species concentrations data are available over a 24-hr period every third day in the IMPROVE observational network, and they are processed to form monthly mean data sets for every site if there are at least three every-third-day observations in 1 month. These monthly mean data sets will be processed to form seasonal and regional mean, median, and maximum as shown in Section 2.4.

2.2. SPEI Drought Index

Standardized Precipitation Evapotranspiration Index (SPEI) is selected in this study to investigate the impacts of drought events on spatial and temporal variations in aerosol concentrations across the contiguous US. SPEI is formulated through a water balance approach based on precipitation and air temperature data and can represent the temporal duration of drought events (Beguería et al., 2014). We select SPEI in this study as not only it is related to air temperature and precipitation, both of which play an important role in the occurrence of wildfires (Holden et al., 2018; Westerling et al., 2006; Williams et al., 2014), but also the relationship between drought events and aerosol concentrations derived from SPEI has been shown to be robust (Y. Wang et al., 2017). The SPEI data set (https://spei.csic.es/index.html) used in this study is based on Climatic Research Unit of the University of East Anglia's monthly precipitation and potential evapotranspiration data sets (https://catalogue.ceda.ac.uk/uuid/c26a65020a5e4b80b20018f148556681), which are based on observational data. The spatial and temporal resolutions of SPEI are 0.5° × 0.5° and 1 month, respectively. Negative and positive SPEI usually represents drought and wet events, respectively, and monthly SPEI is used to detect the drought and wet events that last at least 1 month.

2.3. Time Period and Site Selection

The time period for investigation in this study is 1988–2018, in which the SPEI and IMPROVE data sets have temporal overlaps. To investigate their seasonal characteristics, we group January, February, and March (JFM) as winter; April, May, and June (AMJ) as spring; July, August, and September (JAS) as summer; and October, November, and December (OND) as fall. We use this season definition as most active fires in the northwestern US occur in JAS (O'Dell et al., 2019), and the maximums of non-soil potassium, the indicator of aerosols from wildfires, are generally in August (Figure S1 in Supporting Information S1). In this way, the summer season (JAS) is also the most active fire months in the northwestern US (Figure 6b). This season definition to was also used in O'Dell et al. (2019). For the 31-year study period, we only analyze the data from those IMPROVE sites that have observational data available for at least 240-month (~65% or equivalent to 20 years). The monthly SPEI data sets at the grid boxes where the IMPROVE sites are located are paired with the monthly aerosol speciation data from the corresponding IMPROVE sites. The paired monthly data will be further processed to form seasonal data as shown in Section 2.4. As the dominant species vary by region and season, the contiguous US is divided into the northwestern (north of 37°N and west of 100°W), the southwestern (south of 37°N and west of 100°W), and the eastern (east of 100°W) US. There are 31, 15, and 21 IMPROVE sites meet the data selection criteria across the northwestern, southwestern, and eastern US, respectively.

2.4. Aerosol and SPEI Trends Analysis

To investigate the impacts of drought on the trends in aerosol concentrations, we processed the monthly mean IMPROVE data set that meets the completeness criteria in Section 2.3 into three conditions: one for the whole data record (i.e., the selected monthly mean data from Section 2.3) in 1988–2018, and the other two are the records under drought (SPEI <0.0) and wet (SPEI >0.0) conditions for every month and at every 0.5° × 0.5° grid; we name them the all, drought, and wet conditions, respectively. Thus, data under the drought and wet conditions are mutually exclusive, and the union of them is the all condition. For a certain month, there could be some grids under the drought condition and the rest under the wet condition, which is different from defining the whole area as drought or wet in the month. Based on the categorized IMPROVE monthly data sets, the seasonal mean and seasonal and regional mean, median, and maximum of each data set are computed and their trends are analyzed. The monthly SPEI data in the grids where the monthly IMPROVE data are available are averaged to form seasonal mean and seasonal and regional mean. The trends are calculated as the slopes derived from linear least-squares regression between time and variables to be analyzed, and the slopes are considered statistically significant in the two-tailed *t*-test when the corresponding *p*-value is less than 0.05. The calculation of the uncertainties (standard errors) of the slope (Wilks, 2011) is shown in Text S1 in Supporting Information S1. Meanwhile, with the assumption that if dryness conditions affect species trends, the impacts of drought and wet should be opposite, the one-tailed *t*-test is applied to evaluate the difference between the slopes calculated under different dryness conditions.

3. Results

3.1. Seasonal Mean and Trends of $PM_{2.5}$

Seasonal mean reconstructed $PM_{2.5}$ concentrations from 1988 to 2018 at the contiguous US IMPROVE sites are shown in Figure 1. The reconstructed $PM_{2.5}$ concentrations over the eastern US are generally larger than that over the northwestern and southwestern US in all seasons (Figure 1). Although the largest concentration occurs

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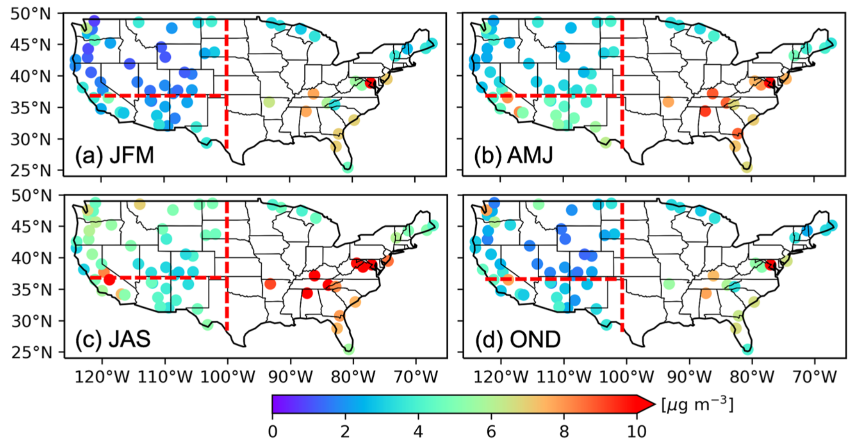

Figure 1. Seasonal mean reconstructed $PM_{2.5}$ concentrations at Interagency Monitoring of Protected Visual Environments sites from 1988 to 2018 in (a) winter (January, February, and March, JFM), (b) spring (April, May, and June, AMJ), (c) summer (July, August, and September, JAS), and (d) fall (October, November, and December, OND). Red dashed lines drawn along 100°W and 37°N are used in the analysis to distinguish between the northwestern, southwestern, and eastern US.

in summer for both the northwestern and eastern US, followed by spring (Figures 1 and 2), the contribution of species for the enhancements are different between these two regions (Figure 2). The large reconstructed $PM_{2.5}$ concentration across the eastern US in summer and spring is mainly caused by the enhancement of sulfate concentrations, followed by OM and dust concentrations; the magnitude of these increases largely surpasses the decrease of nitrate concentration (Figure 2c). Similar seasonal variations in sulfate, nitrate, and dust concentrations also occur in the northwestern US, but the enhancement of OM concentration is the most important contributor to the large reconstructed $PM_{2.5}$ concentration in summer (Figure 2a). As to the southwestern US, the largest reconstructed $PM_{2.5}$ concentration is observed in spring, followed by summer, which is caused by the increase of sulfate, OM, and dust concentrations (Figure 2b), and dust plays a more important role over the southwestern US than northwestern and eastern US (Figure 2).

Figure 3 shows the linear trends in seasonal mean reconstructed $PM_{2.5}$ concentrations during the same period. All the 21 sites across the eastern US observe statistically significant negative trends in winter and fall, and the decreasing reconstructed $PM_{2.5}$ trends are statistically significant at 95% and 85% of sites in spring and summer, respectively. In contrast to the ubiquitous statistically significant negative trends across the eastern US in all seasons, the percentage of sites observing significant decreasing trends over the northwestern US is much smaller. More than 93% of the 31 sites across the northwestern US have negative reconstructed $PM_{2.5}$ trends in winter, spring, and fall, and only 71%–87% is statistically significant. Moreover, only 42% and 10% of sites exhibit statistically insignificant and significant declines in the reconstructed $PM_{2.5}$, respectively in summer in the same region; of the 58% remaining sites that show positive trends, there are five sites that are statistically significant. As to the southwestern US, 73%–93% of sites observe statistically significant declines in the reconstructed $PM_{2.5}$ in winter, summer, and fall, while it decreases to 60% in spring.

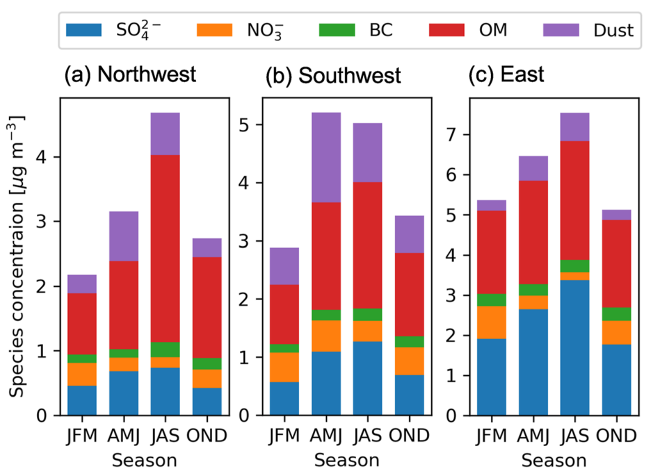

Figure 2. Regional mean concentration of sulfate (SO_4^{2-}), nitrate (NO_3^-), Black Carbon (BC), Organic Matter (OM), and dust during 1988–2018 in the northwestern (a), southwestern (b), and eastern (c) US from Interagency Monitoring of Protected Visual Environments in different seasons. The regions are defined in Figure 1. January, February, and March (JFM), April, May, and June (AMJ), July, August, and September (JAS), and October, November, and December (OND) are months of winter, spring, summer, and fall, respectively.

3.2. Drought Impact on Aerosol Composition

The trends in regional mean reconstructed IMPROVE $PM_{2.5}$ concentrations under the all, drought, and wet conditions are shown in Figure 4. The reconstructed $PM_{2.5}$ trends are always negative regardless of dryness conditions in any seasons and any regions except for the northwestern US in summer (Figures 4a–4c). The trend in the reconstructed $PM_{2.5}$ over the northwestern US in summer is $0.078 \pm 0.041\ \mu g\ m^{-3}\ a^{-1}$ under the drought condition, which is much larger than $0.034 \pm 0.023\ \mu g\ m^{-3}\ a^{-1}$ under the all condition,

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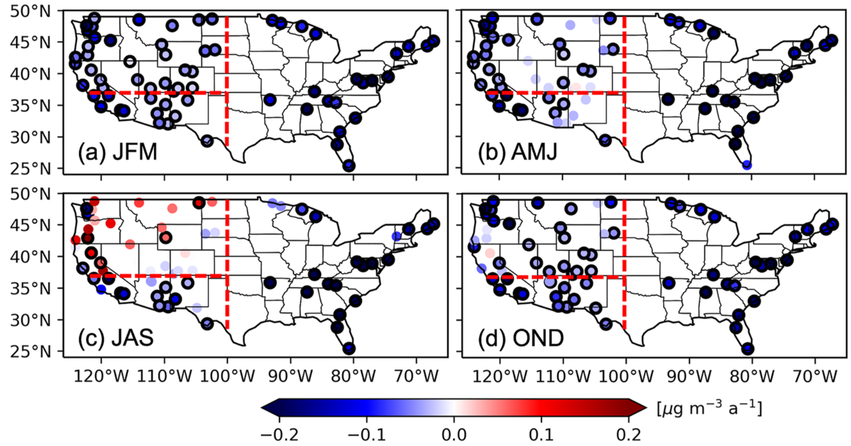

Figure 3. Trends in seasonal mean reconstructed $PM_{2.5}$ concentrations at Interagency Monitoring of Protected Visual Environments sites from 1988 to 2018 in (a) winter (January, February, and March, JFM), (b) spring (April, May, and June, AMJ), (c) summer (July, August, and September, JAS), and (d) fall (October, November, and December, OND). Sites with trends that are significant at a 95% confidence level are outlined in black, and sites with insignificant trends are not outlined. Red dashed lines drawn along the 100°W and 37°N are used in the analysis to distinguish between northwestern, southwestern, and eastern US.

and the trend decreases to 0.004 ± 0.020 μg m^{-3} a^{-1} under the wet condition (Figure 4a). Drought and wet events can significantly modify the trend when the *p*-value for the trend difference between drought and wet events is less than 0.1. The difference between the trends in the reconstructed $PM_{2.5}$ under drought and wet conditions

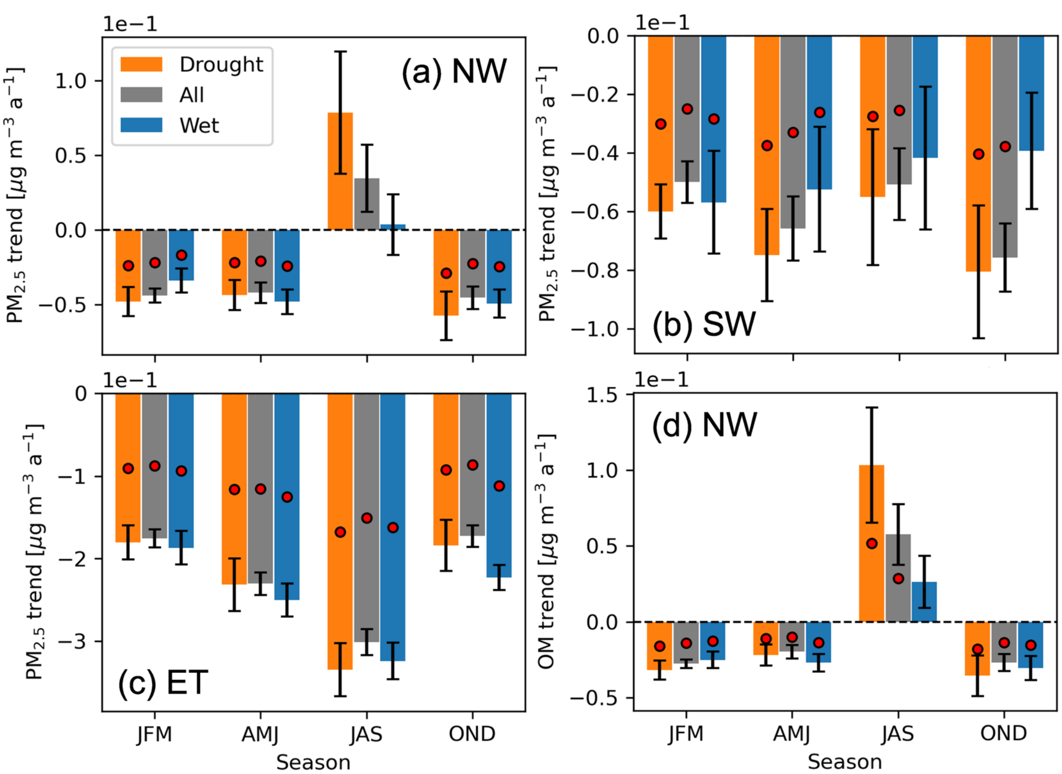

Figure 4. The 1988–2018 trends in mean reconstructed $PM_{2.5}$ over the (a) northwestern (NW), (b) southwestern (SW), and (c) eastern (ET) US and Organic Matter (OM) over the northwestern US (d) in various seasons derived under drought (orange), all data (gray), and wet (blue) conditions. Vertical black lines represent the standard error of trends. Solid red circles represent that the trends are statistically significant. The numbers at the top left corner of panels indicate the magnitude of *y*-axis.

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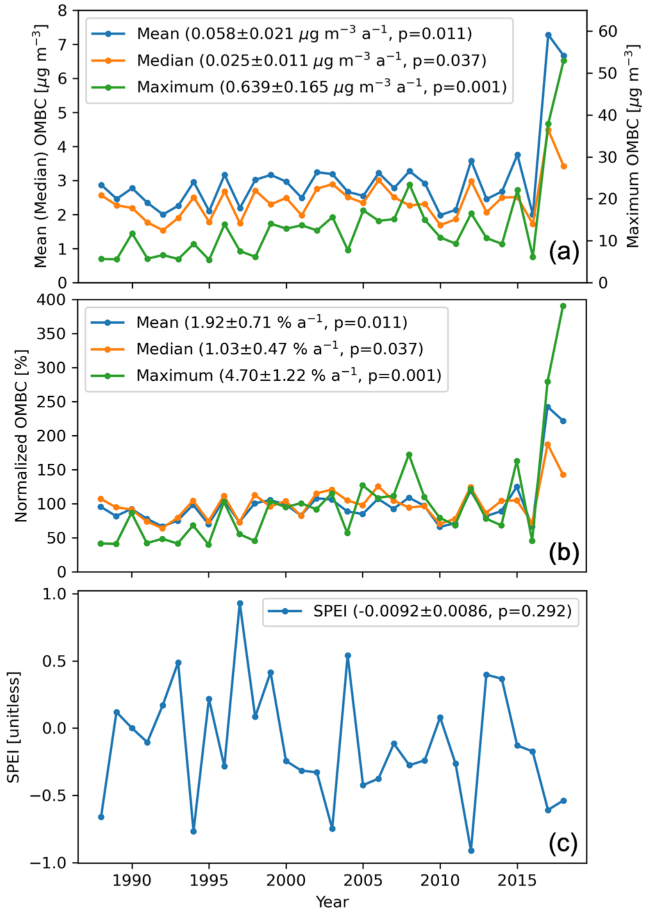

Figure 5. Interannual variations in (a) total carbonaceous aerosols (OMBC) of seasonal and regional mean (blue), median (orange), and maximum (green) derived from monthly mean Interagency Monitoring of Protected Visual Environments data and (c) Standardized Precipitation Evapotranspiration Index (SPEI) over the northwestern US in summer; linear trends, their standard errors, and *p*-values are shown in the legend. Panel (b) is similar to panel (a), but expressed as the percentage of 31-year average of the corresponding time series.

over the northwestern US in summer is statistically significant ($p = 0.05$), while for the rest of the seasons and regions, p values are in the range of 0.11–0.44. These variations in $PM_{2.5}$ trends over the northwestern US in summer under different dryness conditions are mainly caused by the fact that drought and wet events have significant impacts on the trends in OM ($p = 0.04$, Figure 4d). Similar trend variation occurs in BC ($p = 0.07$, Figure S2c in Supporting Information S1, note that the number at the top left corner indicates the magnitude of y axis), but the BC trend difference between drought and wet conditions is around an order of magnitude smaller than that of OM (Figure 4d, note that the number at the top left corner indicates the magnitude of y axis) as the concentrations of BC are much smaller than OM (Figure 2a). As OM (Figure 4d) and BC (Figure S2c in Supporting Information S1) show a similar response to droughts, and the combination has smaller observational error than separate species (Schmid et al., 2001), we combine OM and BC as TCAs (Figure S2e in Supporting Information S1), and it shows that the total carbonaceous trends are very similar to OM trends. Although the trends in sulfate (Figure S2a in Supporting Information S1) in fall, nitrate (Figure S2b in Supporting Information S1) in winter, and dust (Figure S2d in Supporting Information S1) in summer and fall over the northwestern US under drought condition are different from that under wet condition with statistical significance, these differences are not sufficient to lead to statistically significant difference of trends in the reconstructed $PM_{2.5}$ under different dryness conditions over the northwestern US. Similarly, the significant trend differences between drought and wet events for sulfate and dust in fall over the southwestern US, and for nitrate in spring over the eastern US do not significantly change the corresponding trends in the reconstructed $PM_{2.5}$. For certain regions, seasons, and species that are not mentioned above, the trend differences between drought and wet conditions are not statistically significant.

As the drought-induced change of trends in TCAs can significantly affect the trend in the reconstructed $PM_{2.5}$ (Figure 4 and Figure S2 in Supporting Information S1), we further investigate how the linear trend of drought severity affects TCA concentrations. The interannual variations in the regional mean, median, and maximum of TCAs in summer over the northwestern US are shown in Figure 5a. The trend in the maximum during 1988–2018 is 0.639 ± 0.165 μg m^{-3} a^{-1}, which is much stronger than 0.058 ± 0.021 and 0.025 ± 0.011 μg m^{-3} a^{-1} for the mean and the median, respectively (Figure 5a and Table 1). To remove the interference caused by the magnitude difference of these time series in the comparison of their trends, Figure 5b shows the seasonal mean, median, and maximum TCAs expressed as the percentage of 31-year average of the corresponding time series, and their corresponding trends are analyzed. The normalized trend in the maximum is $4.70\% \pm 1.22\%$ a^{-1}, which is still much larger than $1.92\% \pm 0.71\%$ a^{-1} for the mean and $1.03\% \pm 0.47\%$ a^{-1} for the median (Figure 5b); all these

Table 1
Linear Trends and Their Standard Errors in the Mean, Median, and Maximum of Total Carbonaceous Aerosols (TCA; Units: μg m^{-3} a^{-1}) and Mean Standardized Precipitation Evapotranspiration Index (SPEI; Unitless) in Summer Over the Northwestern US During Various Periods

Year	Mean TCA	Median TCA	Maximum TCA	Mean SPEI
1988–2018	0.058 ± 0.021	0.025 ± 0.011	0.639 ± 0.165	−0.0092[a] ± 0.0086
1988–2009	0.028 ± 0.012	0.025[a] ± 0.013	0.575 ± 0.110	−0.0106[a] ± 0.0147
2010–2018	0.538 ± 0.189	0.212[a] ± 0.099	4.094 ± 1.553	−0.0426[a] ± 0.0586

[a]All the trends are statistically significant at a 95% confidence level except for those denoted by a superscript letter.

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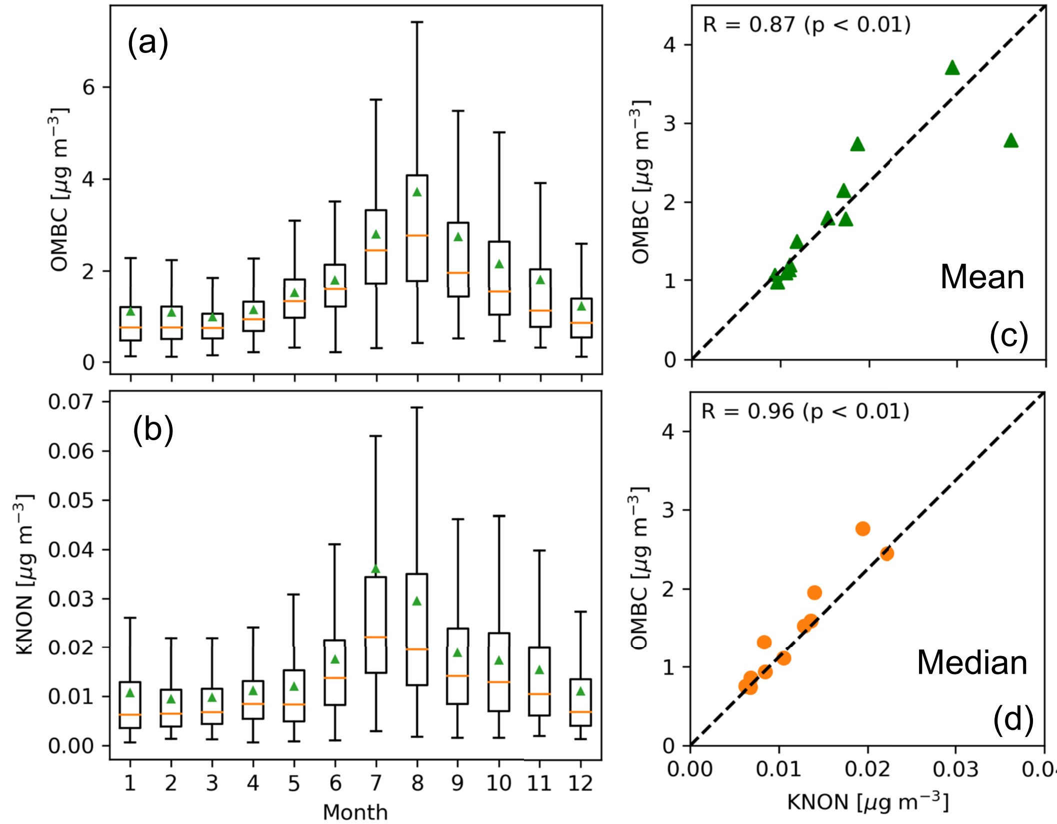

Figure 6. Boxplots of (a) total carbonaceous aerosols (OMBC) and (b) non-soil potassium (KNON) during 1988–2018 in every month over the northwestern US. Scatter plots of the monthly mean (c) and median (d) of total carbonaceous aerosols from (a) versus non-soil potassium from panel (b). Linear correlation coefficients and *p*-values are shown in the scatter plots.

trends are statistically significant. As the trend in maximum is much larger than that in mean and median, the increasing trend in extremely high TCA concentration is a major contributor to the upward trend in total mean carbonaceous aerosol concentration over the northwestern US in summer. Meanwhile, the linear trend in seasonal and regional mean SPEI is negative over the northwestern US during 1988–2018 summer, but it is not statistically significant (Figure 5c).

To further analyze how the linear trend in TCAs is affected by drought, we divide the whole time period into 1988–2009 and 2010–2018. The trends in the mean, median, and maximum of TCA from 2010 to 2018 are 0.538 ± 0.189, 0.212 ± 0.099, and 4.094 ± 1.553 μg m^{-3} a^{-1}, respectively, which are a factor 7–19 larger than that during 1988–2009 (Table 1). Correspondingly, the trend in the seasonal and regional mean of SPEI in summer over the northwestern US during 2010–2018 is -0.0426 ± 0.0586 a^{-1}, which is a factor of 4 stronger than -0.0106 ± 0.0147 a^{-1} from 1988 to 2018, although they are not statistically significant. The trends in SPEI and TCAs during 2010–2018 are simultaneously larger than that during 1988–2009, which means the linear trend in drought partly enhance the TCAs in summer over the northwestern US. We also notice that the TCA concentrations in 2017 and 2018 summers are much larger than that in the rest of the summers, and have important impacts on TCAs during 2010–2018 summers; when the two-summer data are removed, the linear trends in the mean, median, and maximum of TCAs are 0.085 ± 0.147, 0.033 ± 0.100, and 0.164 ± 1.113 a^{-1} during 2010–2016, respectively, which are much weaker than the corresponding 0.538 ± 0.189, 0.212 ± 0.099, and 4.094 ± 1.553 a^{-1} during 2010–2018. Since air pollution extreme events and uncertainties of trends are both important for assessing air quality, we include the two-summer data in the analysis.

According to the method to calculate the standard error (Text S1 in Supporting Information S1) of a linear trend, the large non-linear variation of SPEI leads to the large uncertainty (± 0.0086 a^{-1}, Figure 5c) of SPEI trend (-0.0092 a^{-1}, Figure 5c). Thus, we investigate the following question: does the non-linear variation of SPEI impacts TCA concentrations in addition to its impact on the standard error of SPEI linear trend? The linear

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correlation coefficients between the regional mean, median, and maximum of TCAs and regional mean of SPEI in summer over the northwestern US are −0.45, −0.54, and −0.42, respectively, which are all statistically significant. When the linear trends of these time series are removed by subtracting them from the original time series (Text S2 in Supporting Information S1), the linear correlation coefficients of the residue are −0.41, −0.51, and −0.39, and are still statistically significant. The correlation coefficients for the non-linear part (−0.41, −0.51, and −0.39) are only a little weaker than that for the original time series (−0.45, −0.54, and −0.42); thus, the non-linear temporal variation of SPEI plays a more important role than the linear trend for the increase of TCAs in summer over the northwestern US.

Figure 6 shows that TCAs over the northwestern US mainly originate from wildfires, and Holden et al. (2018) has shown decreasing precipitation, a phenomenon of drought, leads to the increase of wildfire area burned over the western US. Therefore, drought events lead to the enhancement of TCAs through the increase of wildfires over the northwestern US. The median and mean of both TCAs (Figure 6a) and non-soil potassium (Figure 6b) for each month have their minimum in JFM, show increases in AMJ, reach their maximum in JAS, and then decrease in OND. The linear correlation coefficients between non-soil potassium and TCAs for monthly mean and median are 0.87 (Figures 6c) and 0.96 (Figure 6d), respectively, both of which are at a 99% statistical confidence level. In the meanwhile, the temporal variations in BC (Figure S3c in Supporting Information S1), and OM (Figure S3d in Supporting Information S1) follow the same pattern. Although sulfate (Figure S3a in Supporting Information S1), TCAs (Figure 6a) and non-soil potassium (Figure 6b) have a similar pattern for seasonal variation, the change of sulfate is smother than that for the other two species. In contrast, monthly variations of sulfate (Figure S3a in Supporting Information S1), nitrate (Figure S3b in Supporting Information S1), and dust (Figure S3e in Supporting Information S1) are different from that of TCAs and non-soil potassium.

3.3. The Contribution of Total Carbonaceous Aerosols

To investigate the contribution of TCAs to reconstructed $PM_{2.5}$ over the northwestern US, and how drought and wet events affect it, we calculate the percentage of TCAs to reconstructed $PM_{2.5}$ in every season and dryness conditions, and analyze their temporal variations from 1988 to 2018 (Figure 7). Under the all condition, the contribution of TCAs to reconstructed $PM_{2.5}$ shows a statistically significant decreasing trend in winter during 1988–2018 (Figure 7a), and there are no trends in spring (Figure 7b) and fall (Figure 7d); the same holds true under the wet and drought conditions (Figures 7a, 7b and 7d). Thus, drought and wet events do not affect the trends in the contribution of TCAs to reconstructed $PM_{2.5}$ in winter, spring, and fall. Conversely, regardless of dryness conditions, the contribution of TCAs to reconstructed $PM_{2.5}$ show statistically significant increasing trends in summer (Figure 7c). The increasing trends in the contribution of TCAs to reconstructed $PM_{2.5}$ in summer during 1988–2018 result from the combination of the increasing trend in TCA concentrations (Figure S2e in Supporting Information S1) and the decreasing trends in sulfate (Figure S2a in Supporting Information S1) and nitrate (Figure S2b in Supporting Information S1) concentrations. Moreover, the positive trends increase from 0.45% ± 0.09% a^{-1} under the wet condition to 0.60% ± 0.09% and 0.73% ± 0.14% a^{-1} under the all condition and the drought condition, respectively (Figure 7c), which can be explained by the facts that (a) drought events significantly enhance the positive trend in TCA concentrations (Figure 4 and Figure S2 in Supporting Information S1); (b) the impact on reconstructed $PM_{2.5}$ trends from sulfate and nitrate concentrations is not significant (Figure S2 in Supporting Information S1); (c) although the impact on the reconstructed $PM_{2.5}$ trend from dust is statistically significant, it is an order magnitude smaller than that from TCAs (Figure 4 and Figure S2 in Supporting Information S1). We also notice that the contribution of TCA concentrations to the reconstructed $PM_{2.5}$ in 2017 and 2018 summers are much larger than that in the rest of the summers. When the two-summer data are removed, the trends in the contribution are 0.39% ± 0.09%, 0.44% ± 0.08%, and 0.52% ± 0.13% a^{-1} under the wet, all, and drought conditions, respectively. The two-summer data have a larger impact under the drought condition than the wet condition.

4. Discussion

Dry versus wet month separation is conducted for every IMPROVE site rather than for the selected regions as a whole in this study, as there are both drought and wet in any of the three selected regions. With this separation, however, if TCAs at a site mainly come from the long-range transport (from emissions of fires in the upper wind), dryness condition at this site alone cannot explain the TCA concentration at this site. To tackle this situation,

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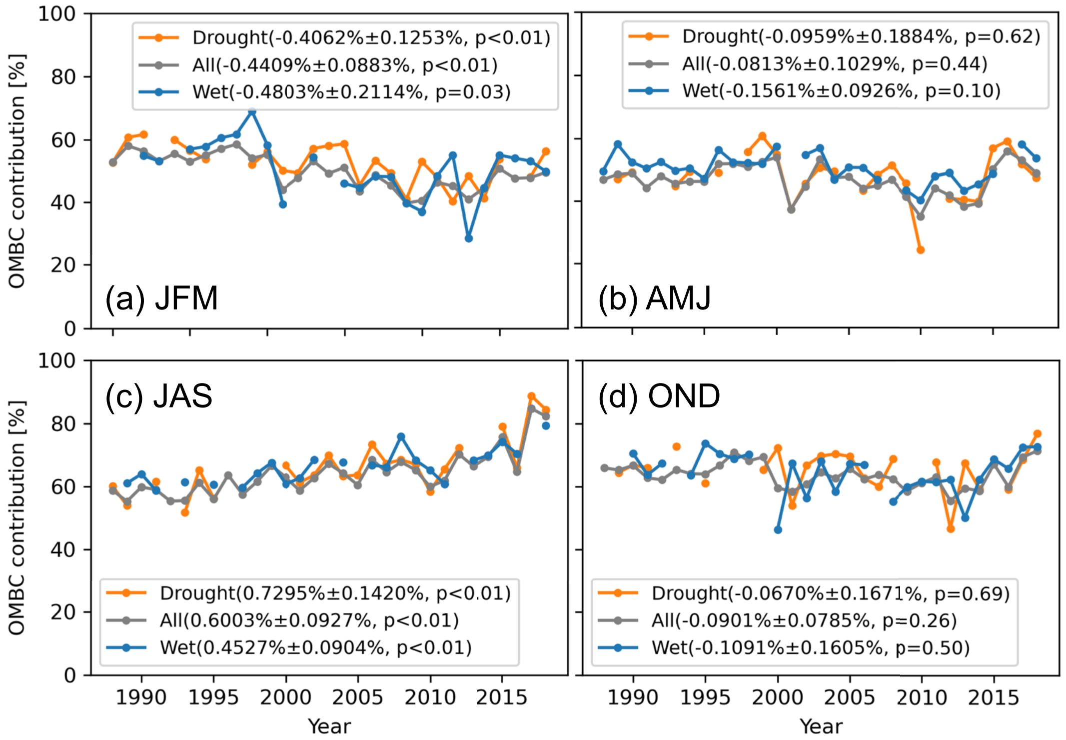

Figure 7. The 1988–2018 trends in the percentage of total carbonaceous aerosols (OMBC) in the reconstructed $PM_{2.5}$ mass in (a) winter (January, February, and March, JFM), (b) spring (April, May, and June, AMJ), (c) summer (July, August, and September, JAS), and (d) fall (October, November, and December, OND) derived under drought (orange), all (gray), and wet (blue) conditions over the northwestern US; linear trends, their standard errors, and *p*-values are shown in the legend.

we further calculate regional and seasonal mean SPEI over the northwestern US in every summer using SPEI at every grid in the region rather than only using SPEI that is paired with IMPROVE sites (Section 2.4). The linear correlation coefficient of the regional summer mean SPEI calculated by using SPEI at every gird and regional summer mean TCA is −0.36 over the northwestern US, which is weaker than −0.45 (Section 3.2) calculated by using SPEI paired with IMPROVE sites. Hence, this contrast suggests that the impact of local dryness condition on TCAs is more important than that of remote dryness condition (and its associated transport of fire emissions through the atmosphere).

In this study, data from IMPROVE sites are used to calculate regional mean, which is further applied for trend analysis. As there are only 31, 15, and 21 IMPROVE sites that can be used across the northwestern, southwestern, and eastern US, respectively, and all these sites are in remote regions, our analysis has the possibility to lack the regional representativeness. However, IMPROVE program has the longest record of aerosol composition data in the U.S. with a focus on rural areas. Unlike urban areas that have undergone a large change of anthropogenic emissions in the past three decades, rural areas can be more susceptible to the impact of drought and the resultant aerosols from wildfires. Future studies may infuse satellite-based aerosol optical depth data with IMPROVE data and study how droughts impact aerosols in urban areas.

5. Conclusions

This study investigates the spatial distributions of both the reconstructed $PM_{2.5}$ and their species concentrations, as well as their respective trends across the US in different seasons in 1988–2018 using IMPROVE data. During the four seasons, the largest reconstructed $PM_{2.5}$ concentrations occur in summer over the northwestern and eastern US mainly due to the enhancement of OM and sulfate aerosols, respectively. In the southwestern US, the increase of sulfate, OM, and dust concentrations lead to the largest $PM_{2.5}$ concentrations in spring. Ubiquitous decreasing trends in the reconstructed $PM_{2.5}$ concentrations are observed in all seasons and regions except for the northwestern US in summer. The increasing trend in $PM_{2.5}$ concentrations over the northwestern US in summer

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during 1988–2018 is primarily due to the positive trend in TCA concentrations, which is more than compensates for the negative trends in sulfate and nitrate aerosols. The value of the negative trends in dust aerosol is much smaller than their standard errors and is negligible.

According to SPEI, IMPROVE data are grouped into drought, wet, and all conditions to investigate how the drought and wet events affect species trends over the northwestern US. The trends in OM and BC over the northwestern US under the drought condition are significantly different from that under the wet condition. Positive trends in OM are observed under the all conditions and are enhanced by droughts, while trends in BC change from negative under the wet condition to positive under the drought condition. Due to the impact of drought and wet events on BC and OM, we combine them as TCAs for further analysis. The monthly variations of TCAs follow the temporal pattern of non-soil potassium, which is a good indicator of wildfire smoke. Hence, droughts affect the temporal variations of total carbon aerosols primarily via its impact on wildfire emissions. The linear correlation coefficient between the regional median of TCAs and the regional mean of SPEI in summer over the northwestern US is −0.54, which is stronger than that for the regional mean (−0.45) and regional maximum (−0.42) of TCAs. When linear trends are removed, these correlation coefficients become −0.41, −0.51, and −0.39, respectively, but are still significant. Thus, non-linear variation of SPEI plays a more important role than its linear trend in changing TCAs over the northwestern US in summer. Due to the fact that drought events significantly enhance the positive trend in TCA concentrations, the percentage contribution of TCAs to $PM_{2.5}$ has an increasing trend of 0.45% ± 0.09% a^{-1} under wet conditions and increases to 0.60% ± 0.09% and 0.73% ± 0.14% a^{-1} in the all and drought scenarios, respectively.

The observational analysis presented here has shown that droughts enhance the TCA pollution through the impacts on wildfires over the northwestern US in summer, hence increase the contribution of TCAs to $PM_{2.5}$. Droughts are expected to increase in the context of global warming, thus this study implicate that drought-induced air pollution through wildfire cannot be overlooked and could partly offset the benefit from reduction of anthropogenic emissions. To circumvent the model deficiencies of predicting wildfires, drought index is likely a promising predicator for forecasting long-term air pollution.

Data Availability Statement

The Interagency Monitoring of Protected Visual Environments data used for species analysis in this study were from Federal Land Manager Environmental Database (Federal Land Manager Environmental Database, 2022). The Standardized Precipitation Evapotranspiration Index (SPEI) data used for drought analysis in this study were from Global SPEI database (Beguería et al., 2022). Figures were made with Matplotlib version 3.2.1 (Caswell et al., 2020; Hunter, 2007).

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