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Quantifying long-term seasonal and regional impacts of North American fire activity on continental boundary layer aerosols and cloud condensation nuclei

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Submitted to *Journal of Earth and Space Science*

This article has been accepted for publication and undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process which may lead to differences between this version and the Version of Record. Please cite this article as doi: 10.1029/2020EA001113

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Abstract

An intimate knowledge of aerosol transport is essential in reducing the uncertainty of the impacts of aerosols on cloud development. Datasets from the U. S. Department of Energy (DOE) Atmospheric Radiation Measurement platform in the Southern Great Plains region (ARM-SGP) and the NASA Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2) showed seasonal increases in aerosol loading and total carbon concentration during the spring and summer months (2008-2016) which was attributed to fire activity and smoke transport within North America. The monthly mean MERRA-2 surface carbonaceous aerosol mass concentration and ARM-SGP total carbon products were strongly correlated ($R=0.82$, $p<0.01$) along with a moderate correlation with the ARM-SGP cloud condensation nuclei (N_{CCN}) product (0.5, $p\sim0.1$). The monthly mean ARM-SGP total carbon and N_{CCN} products were strongly correlated (0.7, $p\sim0.01$). An additional product denoting fire number and coverage taken from the National Interagency Fire Center (NIFC) showed a moderate correlation with the MERRA-2 carbonaceous product (0.45, $p<0.01$) during the 1981-2016 warm season months (March-September). With respect to meteorological conditions, the correlation between the NIFC fire product and MERRA-2 850 hPa isobaric height anomalies was lower (0.26, $p\sim0.13$) due to the variability in the frequency, intensity, and number of fires in North America. An observed increase in the isobaric height anomaly during the past decade may lead to frequent synoptic ridging and drier conditions with more fires, thereby potentially impacting cloud/precipitation processes and decreasing air quality.

Plain Language Summary

Aerosols have complex impacts on cloud development and air quality. This study seeks to illustrate and quantify climatological trends and impacts of carbonaceous smoke aerosols generated from fire activity in North America. In the presence of rising air motion (e.g., updrafts) and sufficient moisture, the smoke aerosols can become cloud condensation nuclei (CCN). The CCN eventually become cloud droplets which gather together to form clouds which may or may not precipitate depending on available moisture and updraft strength. Long-term, surface-based observation data from the Department of Energy (DOE) Atmospheric Radiation Measurement facility in the Southern Great Plains region (ARM-SGP), the NASA 2nd Generation Modern Era Retrospective Reanalysis (MERRA-2) product, and the National Interagency Fire Product show the regional and seasonal behavior and transport of the smoke aerosols derived from fires in the United States, Mexico, and Canada. Since 2010, during the warm season months (March-September), numerous fires due to anomalously dry weather patterns have been observed in North America and likely increased the amount of carbonaceous smoke aerosols and potentially CCN, though the overall atmospheric aerosol burden has noticeably decreased.

Keywords: boundary layer aerosols, aerosol-cloud interactions, aerosol chemistry, biomass burning, climatology

1 Introduction

In North America, carbonaceous aerosols derived from biomass burning are not only known to indirectly influence cloud development but have been increasing in magnitude since

the 2000s (Marlon et al., 2012; Westerling, 2016; Provençal et al., 2017; Logan et al., 2018; Zheng et al., 2020). For example, North America is impacted by long-range wildfire smoke transport from Mexico during the spring months (Lyons et al., 1998; Wang et al., 2009; Saide et al., 2015; Logan et al., 2018; Zheng et al., 2020). The source region then shifts to the western United States and Canada during the summer and autumn months (Chin et al., 2014; Hallar et al., 2017; Brey et al., 2018; Campos-Ruiz et al., 2018; O'Dell et al., 2019; Zheng et al., 2020). A major prediction of the effects of climate change is a likely increase in coverage and magnitude of wildfires thereby increasing the amount of carbonaceous biomass burning aerosols (Westerling et al., 2006; McClure & Jaffe, 2018; O'Dell et al., 2019). Therefore, a robust set of observations is needed to quantify and constrain model simulations of the seasonal and spatial behavior of carbonaceous aerosols.

There are extensive networks such as the Interagency Monitoring of Protected Visual Environments (IMPROVE), Aerosol Robotic Network (AERONET, Holben et al., 1998), and the National Emissions Inventory (NEI), which provide nationwide coverage and continuous retrievals of aerosol chemical and physical data primarily in rural areas (Hallar et al., 2017). Previous studies have used these platforms to measure aerosol optical depth (AOD), aerosol chemical speciation (total or elemental carbon), and particulate matter (PM) products to quantify the seasonal and regional behavior of aerosols (Hand et al., 2013; Raman et al., 2016; Hallar et al., 2017; Keeley & Syphard, 2018; McClure & Jaffe, 2018; O'Dell et al., 2019). Based on their findings, many studies concluded a decreasing trend in column AOD and surface $PM_{2.5}$ (e.g., particulate matter with aerodynamic diameters less than 2.5 μm) but an increase in the contribution of carbonaceous aerosols to $PM_{2.5}$ (Hand et al., 2013; Wang et al., 2015; Provençal et al., 2017; McClure & Jaffe 2018; O'Dell et al., 2019; Hand et al., 2019) during the past two decades. In addition, Hallar et al. (2017) and O'Dell et al. (2019) concluded that the frequency of occurrence of wildfires was higher in the western United States during the

summer months and was strongly correlated with increased arid conditions, especially in the higher terrain of the Intermountain West.

Aerosols contribute to cloud development due to their inherent ability to activate as cloud condensation nuclei (CCN) under favorable dynamic and thermodynamic conditions (Rosenfeld et al., 2008; Liu & Li 2014, Logan et al., 2014; Fan et al., 2015; Fan et al., 2016; Logan et al., 2018; Zhang et al., 2019). Rising motion resulting from synoptic and mesoscale dynamics along with sufficient moisture are needed for CCN to undergo net condensational growth to become cloud droplets that can eventually become raindrops via the collision/coalescence process (Pruppacher & Klett, 1997; Rosenfeld et al., 2008; Liu & Li 2014). This study seeks to build upon previous research (e.g., Spracklen et al., 2011) by seeking to not only quantify the spatiotemporal trends of carbonaceous aerosols derived from fire activity and the meteorological conditions responsible for their transport, but also their impacts boundary layer CCN. The U. S. Department of Energy (DOE) operates the Atmospheric Radiation Measurement (ARM) observation facility located in the Southern Great Plains region (ARM-SGP) The ARM-SGP site has been providing continuous atmospheric, meteorological, and boundary layer aerosol observations for over 20 years (Sisterson et al., 2016). Since the ARM-SGP site represents a point measurement, datasets from the larger spatiotemporal NASA Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2) product (Randles et al., 2017; Buchard et al., 2017) are used in conjunction with the ARM-SGP observations (Logan et al., 2018). The results from the ARM-SGP and MERRA-2 datasets will help to answer the following scientific questions (SQs):

SQ1: How can the trend in carbonaceous aerosol loading with respect to synoptic forcing be useful in investigating long-term aerosol-climate effects on cloud condensation nuclei within the continental boundary layer?

SQ2: What is the nature of the major synoptic forcing that influences aerosol transport on a climatological scale?

SQ3: How well do the ARM-SGP and MERRA-2 datasets quantify the seasonal variation in aerosols derived from fire activity in North America?

Our previous work focused on aerosol-cloud-meteorology relationships during the 2011 Midlatitude Continental Convective Clouds Experiment (MC3E) field campaign (Logan et al., 2018; Zheng et al., 2020). MC3E was conducted over the SGP region from March through June (Jensen et al., 2015). In this study, we use a longer-term dataset in conjunction with MERRA-2 reanalysis products to address the SQs. In particular, we focus on extreme aerosol events with respect to the long-term average of the ARM-SGP and MERRA-2 datasets. An additional, independent database of fire number and burn acreage from the National Interagency Fire Center (NIFC) is used to support and validate the findings of the ARM-SGP and MERRA-2 data. Section 2 details the ARM-SGP Aerosol Observation System (AOS) aerosol and chemical data which are used as "ground truth" to help validate the MERRA-2 datasets along with a brief discussion of the NIFC dataset. The results of the analysis are presented in Section 3. A brief summary along with ideas for future research regarding boundary layer aerosol-cloud interactions are presented in Section 4.

2 Materials and Methods

2.1 ARM-SGP Facility

The ARM-SGP central facility (36.6°N, 97.5°W) hosts a variety of ground-based instruments and remote sensing equipment which provide continuous physical and chemical measurements of the atmosphere (Jensen et al., 2015; Parworth et al., 2015; Sherman et al., 2015; Sisterson et al., 2016). These measurements include: total aerosol number concentration

(N_a), CCN number concentration (N_{CCN}), and carbonaceous/inorganic chemical species. The facility is located in a central region of the United States where it can sample boundary layer air parcels from regional as well as distant source regions such as the Gulf of Mexico, Mexico, Canada, Asia, and Africa (Sherman et al., 2015; Sisterson et al., 2016; Logan et al., 2018; Zheng et al., 2020).

2.1.1 Aerosol Observation System

The AOS platform features a TSI Model 3563 nephelometer, TSI Model 3772 Condensation Particle Counter, Data Management Team (DMT) optical particle counter (OPC), and Aerodyne model aerosol chemical speciation monitor (ACSM). The nephelometer measures the aerosol scattering coefficient (σ_{sp}) at three wavelengths (450, 550, and 700 nm) for particles of 10 μm size or less at 40% relative humidity to minimize uncertainties due to hygroscopic effects (Jefferson, 2011). Retrievals of sub-10 μm aerodynamic size range particles (PM_{10}) are used in this study. The data are averaged daily and have been quality assured following Anderson and Ogren (1998). The green wavelength aerosol scattering coefficient (σ_{sp550}) is used to denote aerosol loading where clean conditions have values less than 20 Mm^{-1} and values greater than 60 Mm^{-1} are considered as polluted conditions (Bergin, 2000; Logan et al., 2018).

The CPC measures surface N_a in units of # cm^{-3} and the data are averaged every hour (Kuang & Mei, 2019). The OPC measures surface N_{CCN} at seven supersaturation levels (Jefferson, 2011) each hour. This study uses the 0.2% supersaturation level to represent optimal low-level cloud development under conditions of moderate to strong aerosol loading (Hudson & Noble, 2013; Logan et al., 2018). The ACSM provides information on the non-refractory, submicron aerosol composition with respect to the following species: total carbonaceous (black and organic carbon), and inorganic ions of sulfate, nitrate, ammonium, and chloride (Ng et al.,

2011; Parworth et al., 2015). The total carbonaceous product is used as a proxy for urban/industrial pollution and biomass burning smoke (Hudson et al., 2004; Liu & Li, 2014; Parworth et al., 2015; Logan et al., 2018). Note that the smoke particles are a highly complex mixture of soot, brown, and aged organic carbon compounds. However, the total carbonaceous product is well correlated with biomass burning smoke (e.g. Logan et al., 2018). ACSM data are available from 2011-2016 while the remaining AOS instruments have data available from 2008-2016.

2.2 NASA MERRA-2

MERRA-2 is the latest reanalysis for the satellite era (1980 onward) using the Goddard Earth Observing System version 5 (GEOS-5) Earth system model (Randles et al., 2017; Buchard et al., 2017). The MERRA-2 aerosol chemistry product contains a variety of species which contribute to the total atmospheric aerosol burden. The MERRA-2 domain is bounded by a rectangle ranging from 15-85°N and 65-170°W with the ARM-SGP site located near the center. The domain includes Alaska, Canada, the contiguous United States, and Mexico as well as the eastern Pacific Ocean and Gulf of Mexico. This study considers boundary layer aerosols and assumes a well-mixed distribution from the surface to the sub-cloud layer (e.g., Randles et al., 2017; Buchard et al., 2017) to be comparable with observations from the surface-based ARM-SGP AOS site. The gas products (e.g., ozone, sulfur dioxide, carbon monoxide, and carbon dioxide) are not considered.

A detailed explanation of the MERRA-2 aerosol data assimilation techniques is provided by Randles et al. (2017) and Buchard et al. (2017). We do note that both papers emphasized that caution should be taken when interpreting the multi-decadal aerosol datasets with respect to trend analyses since the product highly relies on satellite AOD in the data

assimilation process. Uncertainties also occur in the geolocation of various aerosol types due to meteorological activities. For example, smoke and dust particles can have similar physical properties that are at times not well distinguished by the MERRA-2 dataset (see Buchard et al., 2017). Therefore, this study takes great care in validating the reanalysis dataset with ground-based measurements to generate realistic results and conclusions.

Note that the correlation between the MERRA-2 black (BC) and organic carbon (OC) products rarely falls below 0.9. In addition, the BC concentration is typically an order of magnitude less than the OC concentration. Furthermore, after a period of several hours to days after an initial fire episode, water soluble organic carbon compounds (e.g., "brown" carbon) are assumed to be the primary contributors to carbonaceous aerosols within the aerosol plumes during transport (e.g., aging and oxidation mechanism of black carbon). Therefore, only the MERRA-2 OC product will be used in this study. Under polluted conditions, BC aging also involves significant secondary inorganic aerosol formation according to the recent laboratory and modeling studies (Wang et al., 2018; Zhang et al., 2020).

The geopotential heights in MERRA-2 can be used as a proxy for aerosol transport via large-scale, synoptic weather patterns. Climate anomalies (positive or negative) in the synoptic weather pattern can affect how aerosols impact other weather phenomena such as wildfires, hurricanes, severe weather outbreaks, floods, and droughts (Prospero & Lamb, 2003; Saide et al., 2015). This study utilizes 850 hPa geopotential heights (sub-cloud, planetary boundary layer level) from 1981-2016 as a climatological mean which encompasses the period of operation of the ARM-SGP site used in this study (2008-2016). The aerosol and 850 hPa geopotential height products have a spatial resolution of 0.5º x 0.625º.

2.3 National Fire Interagency Center

The National Fire Interagency Center (NIFC; www.nifc.gov) has been collecting yearly data and situation reports on the number of active fires and burn acreage in the United States since 1926 (Westerling, 2016). The NIFC dataset has greatly evolved over the past 90 years. It should be noted that the fire reports have undergone more scrutiny and quality control since 2000. The ratio of burn acreage to fire number is used in conjunction with the MERRA-2 organic carbon surface mass concentration (proxy for wildfire smoke) to illustrate a robust connection between observed fire activity and the impact of biomass burning smoke aerosols on the atmosphere. Though the NIFC dataset covers primarily the contiguous United States this study focuses on smoke aerosols from fire activity upwind of the ARM-SGP site.

3 Results

3.1 Monthly aerosol and chemical analysis

Figure 1 shows the monthly means, medians, and interquartile ranges (IQRs) of the ARM-SGP ground-based aerosol scattering coefficient (σ_{sp}), aerosol number concentration (N_a), and N_{CCN} data for the 2011-2016 period. The data are positively skewed for all three AOS products during much of the year. The monthly trends are similar with maximum values occurring during the spring and summer seasons. However, the months when the maxima occur are different. For example, the greatest magnitude of aerosol loading at the ARM-SGP site occurs in July and August where the data skewness and IQR are the largest (Figure 1a). Drier conditions in the late autumn and winter months (in the absence of snow cover) can lead to sporadic, extreme aerosol events which add to statistical variability.

The N_a amount represents the total number of boundary layer aerosols that can serve as centers for condensation while the N_{CCN} product is the fraction of N_a that can activate as cloud condensation nuclei. The seasonal trends for both products are similar with spring and summer

maxima. The peak values observed in March and October suggest a larger number of hygroscopic aerosols transported to the ARM-SGP site. Though the aerosol loading is higher in July and August, a lower number concentration of hygroscopic aerosols was inferred from the decrease in N_{CCN}. Figure 1c shows the amount of N_a that can eventually become CCN. The aerosols that are advected to the ARM-SGP site during the spring and early summer (March-June) tend to readily activate as CCN under humid conditions brought about by mesoscale and synoptic weather events (e.g., deep convection and frontal passages) (Logan et al., 2018; Zheng et al., 2020). Fewer CCN are observed in the winter months due to fewer aerosol intrusions. A further discussion of the frequency of clean and polluted aerosol events which lead to the variability in the datasets is presented in Section 3.2.

Figure 2 shows the ARM-SGP monthly mean and MERRA-2 domain monthly mean carbonaceous concentration data for 2011-2016. The two platforms are strongly correlated in their monthly trends (R-value=0.82; $p<0.01$). Though the ARM-SGP site is a point measurement within the larger MERRA-2 domain, both platforms show a summer mean maximum. The ARM-SGP site has a larger IQR since it is more sensitive to regional transport due to its smaller scale of observation (e.g., mesoscale) compared to the larger synoptic domain of MERRA-2. In addition, the spring season contribution of biomass burning smoke from Mexico is dampened by the large domain of study while the summer season signal is more robust due to a greater number of active wildfires in the western United States and Canada (see Section 3.3).

In terms of the monthly mean N_{CCN}, there is a moderate, somewhat statistically significant relationship with the monthly mean MERRA-2 carbonaceous data (0.50; $p\sim0.1$) along with a strong significant correlation with the monthly mean ARM-SGP AOS total carbon (0.7; $p\sim0.01$). Logan et al. (2018) previously found similar correlations using hourly mean N_{CCN}, total carbon, and MERRA-2 carbonaceous aerosol data during the three-month 2011

MC3E field campaign. The study concluded that factors such as aerosol-to-CCN conversion efficiency, aerosol hygroscopicity, and uncertainties in measurement representativeness between the ARM-SGP and MERRA-2 platforms can lead to lower correlations. Though the brevity of the aerosol and chemical datasets can raise many concerns, other studies conducted over the ARM-SGP site have found similar temporal relationships. For example, Sherman et al. (2015) found a similar statistically significant decreasing trend in σ_{sp} with a corresponding increasing trend in scattering Ångström exponent from 1997-2013. Furthermore, absorption Ångström exponent values near unity observed during the warm season months of May-September denoted influences of carbonaceous aerosols derived from biomass burning (Sherman et al., 2015). Parworth et al. (2015) employed an ACSM to analyze long-term aerosol chemical data and concluded that biomass burning aerosols and biogenic emissions also contributed to the overall carbonaceous concentration observed at the ARM-SGP site during the spring and summer months. In context of this study, the long-term MERRA-2 and AOS trends in this study support previous work.

3.2 ARM-SGP frequency analysis

To explain the monthly variability in aerosol scattering coefficient and N_{CCN} at the ARM-SGP site during the past decade, a frequency analysis of clean and polluted airmass intrusions is conducted. Figure 3 shows the frequency of clean and polluted aerosol and CCN conditions over the ARM-SGP site from 2008-2016. The summer maxima in aerosol loading and N_{CCN} frequency support the previous results in Section 3.1. The summer months tend to feature more extreme aerosol events (e.g., fire activity) which supply aerosols that may activate as CCN under moist conditions prior to a synoptic or deep convective event. The frequency of clean airmass episodes is highest during the winter (Figure 3a). In fact, the spring, autumn, and

winter months typically feature more progressive weather patterns which shift the airflow from southerly (polluted, moist Gulf air) to northerly (clean, dry continental air) (Zheng et al., 2020).

3.3 The impacts of anomalous synoptic forcing

Favorable meteorological patterns are needed to (1) transport polluted airmasses, (2) mix aerosols down to the surface, and (3) transport clean, continental air to the ARM-SGP site (Miller et al., 2011; Logan et al., 2018; Zheng et al., 2020). Figure 4 shows the yearly mean scattering coefficient at the ARM-SGP site for 2008-2016. There has been an overall decline in aerosol loading for the spring and winter seasons. The summer season dominated the statistical average of the yearly means in all years except for 2010. The spring monthly mean values are typically equal to or just below the summer mean values with a few exceptions (e.g., 2010, 2013, and 2016). Note that the 2010 and 2013 winter seasons featured exceptional drought conditions within the SGP region which exacerbated the likelihood for wildfire activity near the ARM-SGP site.

Three examples are chosen from this dataset: (a) the spring season of 2011, (b) the summer season of 2015, and the spring/summer seasons of 2016. During March and April of 2011, large smoke plumes were observed by satellite transporting to the SGP via the Gulf of Mexico. The 2015 late spring and summer months featured extreme smoke generation from fire activity in Alaska and Canada which was advected southward and eastward to the United States. The 2016 spring/summer months were among the lowest aerosol loading magnitudes observed at the ARM-SGP site and thus represent a "clean contrast" to the 2011 and 2015 seasons.

Figure 5 shows the MERRA-2 850 hPa isobaric height anomalies calculated using the difference in the 2011, 2015, and 2016 seasonal and the 1981-2016 climatological domain means. The 2011 spring season shows a broad negative height anomaly (shaded in blue)

exhibiting a cyclonic wind pattern stretching from Alaska southeastward toward the SGP region (Figure 5a). This season featured frequent lee-side low pressure troughs ("Colorado Low") which contributed to numerous outbreaks of severe weather in the SGP, the Gulf Coast, and the eastern United States. The positive anomaly (10-20 m above average) in the Gulf of Mexico facilitated dry conditions in Mexico which led to frequent wildfire episodes as well as frequent northward smoke transport to the SGP region, as indicated by the wind vectors passing over the dark red contours (positive anomalous fire activity) in Mexico (Wang et al., 2009; Saide et al., 2015; Fan et al., 2015; Logan et al., 2018). In contrast, Figure 5b shows a broad positive height anomaly region surrounded by an area of weaker smoke transport over the Gulf of Mexico combined with anomalous weaker smoke activity (dark blue contours) in Mexico during the 2016 spring season. This pattern led to fewer aerosols being observed at the ARM-SGP site.

Figure 5c shows a narrow, negative isobaric height anomaly (15-20 m below average) stretching from Alaska to the northeastern United States with a slightly negative anomaly (~5 m below average) exhibiting northerly winds draped over the SGP. In addition, anomalously strong wildfire activity was present in the Pacific states and Idaho within the positive anomaly. The carbonaceous aerosols were well captured by the ARM-SGP site as a result. It appears that the ARM-SGP site exhibits a dependency on the isobaric anomaly in that negative anomalies allows for more intrusions of carbonaceous aerosols from Mexico and Canada while positive anomalies allow for more intrusions from the west.

During the spring and summer months of 2016, persistent anticyclonic wind patterns were evident over much of North America including the SGP region (Figures 5d). In fact, Figure 5d shows regions of anomalous carbonaceous aerosols (red and blue contours) being advected north and east of the SGP. This led to fewer instances of aerosols being advected to and observed by the ARM-SGP site, though there were most certainly wildfire outbreaks in the

western United States and Canada (e.g., Fort McMurray fire in May 2016). An additional method to analyze the ARM-SGP and MERRA-2 aerosol datasets with respect to fire activity climatology is presented in the next section.

3.4 NIFC data product comparison with ARM-SGP and MERRA-2

Figure 6 shows the yearly domain mean MERRA-2 organic carbon (OC) surface concentration (green circle line) and NIFC fire number ratio data (gold diamond line) from 1981-2016. Note that the NIFC product is being used as an independent dataset to compare with the AOS and MERRA-2 data to (a) provide an additional viewpoint on how fire activity can influence carbonaceous aerosol contributions to the atmosphere and (b) strengthen the connection between synoptic conditions and fire activity. Moreover, there is an underlying assumption that the ARM-SGP site will retrieve information about the carbonaceous aerosols derived from fire activity north and west of the SGP region where previous studies have suggested much of the fire activity in North America will increase in future decades (Hallar et al., 2017; Malm et al., 2017; McClure & Jaffe, 2018).

There is a moderate correlation between the mean MERRA-2 OC and NIFC datasets ($R=0.40$; $p\sim0.01$). When focusing on the warm season months (blue triangle line) of March-September (i.e., most probable months of fire activity in North America), the correlation increases to 0.45 ($p<0.01$). Interestingly, both trends have shown an increase in carbonaceous concentration and fire ratio since 2000 with similar decreases in 2010, 2014, and 2016. As previously shown in Figure 4, the ARM-SGP site illustrated an increasing trend in aerosol scattering coefficient that occurred primarily during the summer season months from 2009-2015. Furthermore, there is a similar observed decrease in aerosol concentration over the ARM-SGP site in 2016.

A brief analysis of satellite imagery from the Moderate Resolution Imaging Spectroradiometer (MODIS) platform shows intermittent smoke plumes from smaller agricultural burns and wildfires located upwind of the ARM-SGP site (MODIS imagery is available at NASA EOSDIS Worldview; https://worldview.earthdata.nasa.gov/). An additional analysis of the correlation between the NIFC fire number ratio and MERRA-2 850 hPa isobaric height anomaly reveals R-values of 0.30 ($p\sim0.06$) and 0.26 ($p\sim0.13$) for the yearly and warm season means, respectively. The instances where the height anomalies are negative typically correspond to weaker (but non-zero) fire activity while positive anomalies suggest more favorable conditions for drought. Since 2010, the height anomaly has become increasingly positive denoting stronger ridging that can facilitate frequent arid conditions which are necessary for wildfire activity. The related increase in carbonaceous concentration is indicative of the impacts of increased fire activity on the atmospheric aerosol burden.

It is likely that a combination of the variability in the magnitude, location, and season of the fires plus synoptic variability contribute to the decreased correlation and statistical significance between the domain mean MERRA-2 and NIFC datasets. Hence, the increasing trend in fire burn acreage and the atmospheric contribution of carbonaceous aerosols with respect to synoptic forcing should continue to be investigated in future research efforts involving aerosol-cloud-climate interactions (Zheng et al., 2019). In addition, air quality concerns typically increase in areas downwind of fire activity which also warrant further investigation (Miller et al., 2011; Hand et al., 2013; Chin et al., 2014; Malm et al., 2017; Brey et al., 2018). Moreover, the strong correlation between the long-term MERRA-2 and ARM-SGP carbonaceous aerosol data ($R\sim0.82$) can prove to be useful when constraining aerosol concentrations in data assimilation platforms that are computationally taxed due to resolution issues.

4 Conclusions and future work

It is evident that though atmospheric aerosol loading shows a declining trend in North America, carbonaceous biomass burning aerosol concentration derived from wildfire activity has apparently increased, especially during the past decade. We present the following conclusions to address three scientific questions (SQs):

(SQ1) The 2008-2016 ARM-SGP measurements of boundary layer aerosol and CCN number concentrations showed similar seasonal increases during the spring and summer months in comparison to the larger MERRA-2 domain surface carbonaceous concentration. Despite the differing scales representativeness of both platforms, the monthly mean AOS total carbon and MERRA-2 surface carbonaceous concentrations were strongly correlated (R-value=0.82, $p<0.01$). There was a dominant seasonal signal in the summer months due to fire activity in northern and western North America in addition to a weaker signal in the spring months owing to long-range smoke transport from agricultural burns and wildfire activity in Mexico. The corresponding increases in N_{CCN} during the spring and summer months reflect the relative ease of the smoke aerosols to activate as CCN under humid conditions which precede episodes of synoptic and mesoscale events (e.g., frontal passages and deep convection). This is supported by the moderate correlation between the monthly mean ARM-SGP N_{CCN} and MERRA-2 surface carbonaceous concentration products (0.5, $p<0.1$) and the strong correlation between the ARM-SGP total carbon and N_{CCN} (0.7, $p\sim0.01$).

(SQ2) Using case studies, the 850 hPa isobaric height anomalies were found to be responsible for aerosol transport to the ARM-SGP site. Strong ridging (positive height anomalies) positioned over the Gulf of Mexico during the spring months likely led to

droughts which facilitated frequent wildfires in addition to regular seasonal agricultural burning in Mexico. Carbonaceous aerosols derived from the fire activity were transported northward by negative height anomalies (~15 m below 1981-2016 climate mean) positioned over the SGP. However, during the summer months when the negative anomaly was centered north of the SGP, carbonaceous aerosols were transported southward and eastward from fires in Alaska, the western United States, and western Canada, to the ARM-SGP site. When positive isobaric height anomalies were present over the North American continent, long-range transport of carbonaceous aerosols to the SGP was less frequent although local and regional fire activity could still be observed by the ARM-SGP site.

(SQ3) The NIFC fire ratio product has shown a noticeable increase in fire burn acreage since 2010. There has been a corresponding increase in surface carbonaceous aerosol mass concentration which was moderately correlated with the NIFC product (0.40; p~0.01). During the warm season months (March-September) the correlation was higher (0.45; p<0.01). In addition, the ARM-SGP N_{CCN} measurements increased (decreased) in direct response to the seasonal increases (decreases) in carbonaceous aerosols. Moreover, there was a weak correlation between the NIFC fire number ratio and MERRA-2 850 hPa isobaric height anomaly as given by R-values of 0.30 (p~0.06) and 0.26 (p~0.13) for the yearly and warm season means, respectively. This was likely due to variabilities in fire aerosol emission, fire size and location, and positions of the synoptic ridges within the domain of study. More input from emission inventory and meteorological data is certainly needed in future work.

A secondary point to conclude from SQ3 is the apparent increase in the positive isobaric height anomaly with respect to the corresponding increase in fire activity since 2010. In modern times, a growing percentage of population and infrastructure is being impacted by these fires (e.g., loss of life/property). Thus, there is a need to better quantify the long-term effects of carbonaceous aerosols in the vein of climate change.

The ARM-SGP site is centrally located within the North American continent. Its long-term operation continues to offer a unique perspective on the direct and indirect effects of aerosols derived from fire activity under certain meteorological conditions. The ARM facilities (e.g., SGP, ENA, and NSA) can present viable constraints as well as a "ground-truth" to platforms having larger spatial domains (e.g., satellite and reanalysis) and will continue to be used in future studies regarding long-term aerosol-cloud-climate interactions within continental and marine boundary layer regimes.

Acknowledgments

The surface aerosol data were obtained from the ARM-SGP central facility sponsored by the U. S. DOE Office of Energy Research, Office of Health and Environmental Research, and Environmental Sciences Division. Analyses and visualizations used to generate the MERRA-2 figures were produced using the NASA Giovanni online data system, developed and maintained by the NASA GES DISC (found at https://giovanni.gsfc.nasa.gov/giovanni/). The wind data used in Figure 5 are available from NOAA ESRL data repository website (found at https://www.esrl.noaa.gov/psd/data/composites/day/). The authors wish to thank the anonymous reviewers for their insightful comments to improve the manuscript. This research was supported by National Science Foundation Collaborative Research under award number AGS-1700796 at Texas A&M University, AGS-1700728 at the University of Arizona, and

AGS-1700727 at California Institute of Technology. Eleanor Marlow and James Maddux were supported by the Texas A&M University High Impact Learning Program grant.

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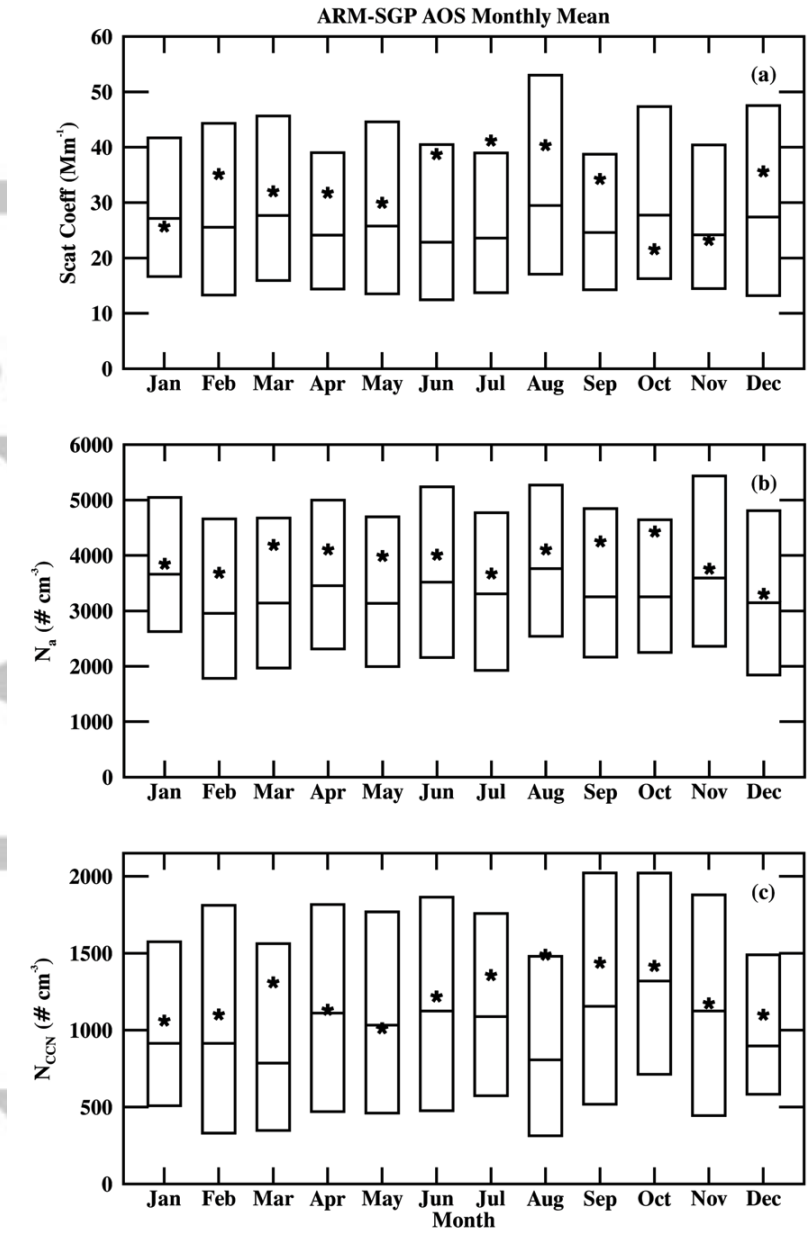

Figure 1. 2008-2016 monthly mean (asterisk) and median Aerosol Observation System (AOS) (a) scattering coefficient, (b) aerosol number concentration (N_a), and (c) cloud condensation nuclei number concentration (N_{CCN}). The interquartile range (IQR) is added to show the spread of the AOS measurements. Note the seasonality of the dataset which is more positively skewed during the summer and autumn (JJASO) months with a second maximum from late winter through the spring (FMAM).

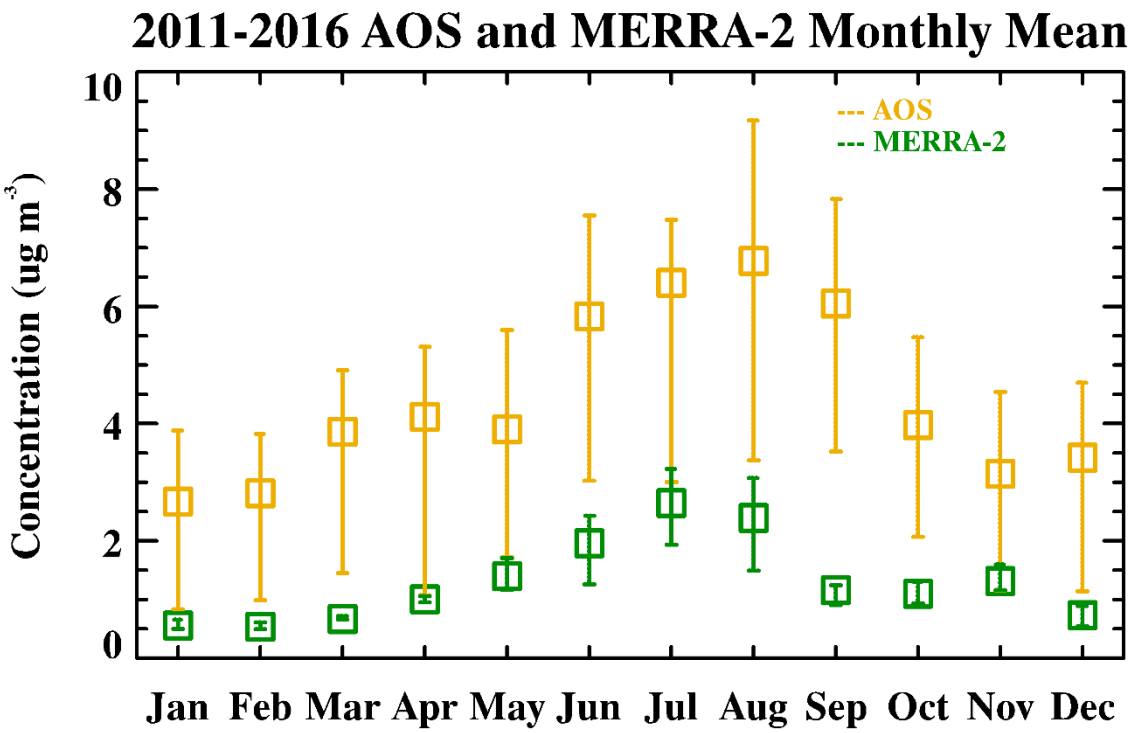

Figure 2. Monthly mean (box) carbonaceous concentrations from the (a) Aerosol Observation System (AOS) (2011-2016) and (b) MERRA-2 (2011-2016) platforms. Note the summer maximum in carbonaceous aerosols is captured by both platforms due to wildfire activity within North America. The bars denote the IQR as in Figure 1.

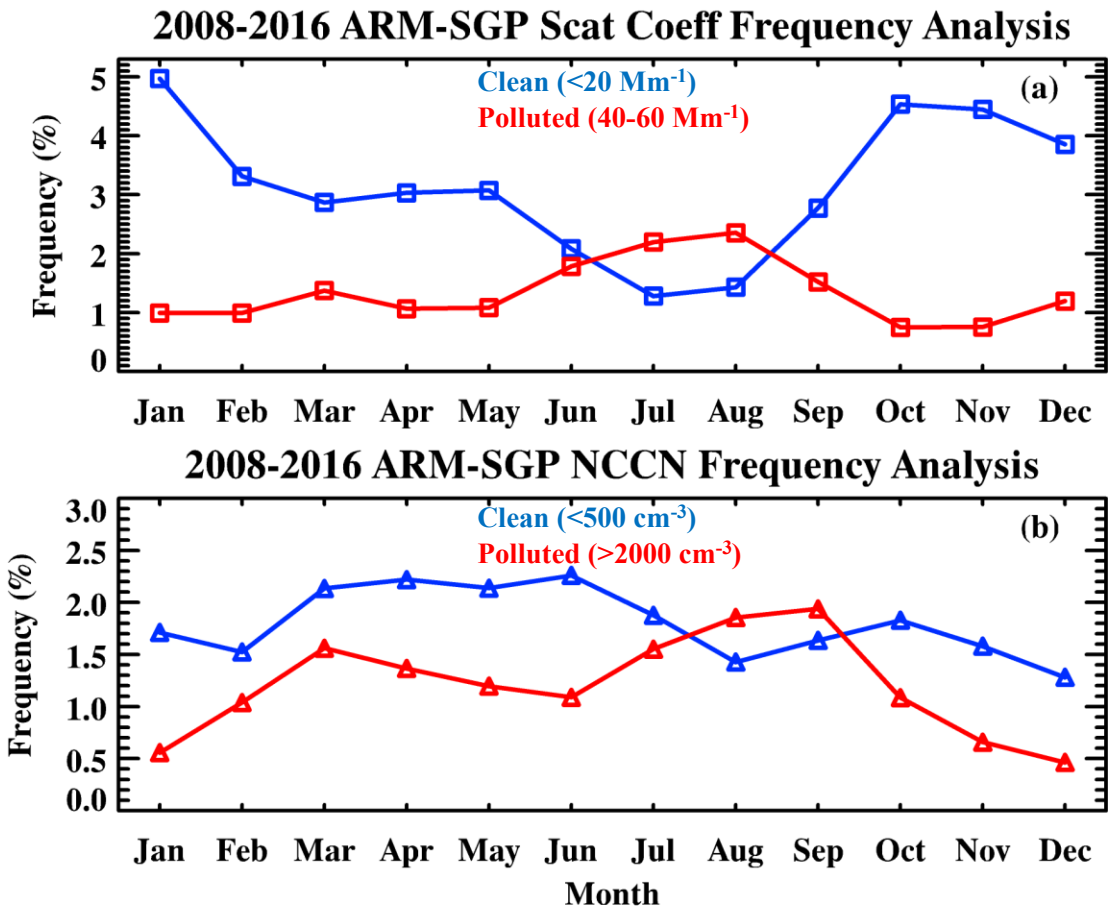

Figure 3. Frequency analysis of the mean monthly Aerosol Observation System (AOS) (a) scattering coefficient and (b) number concentration of CCN. Note that clean continental conditions are more frequent for most of the year except during the summer months.

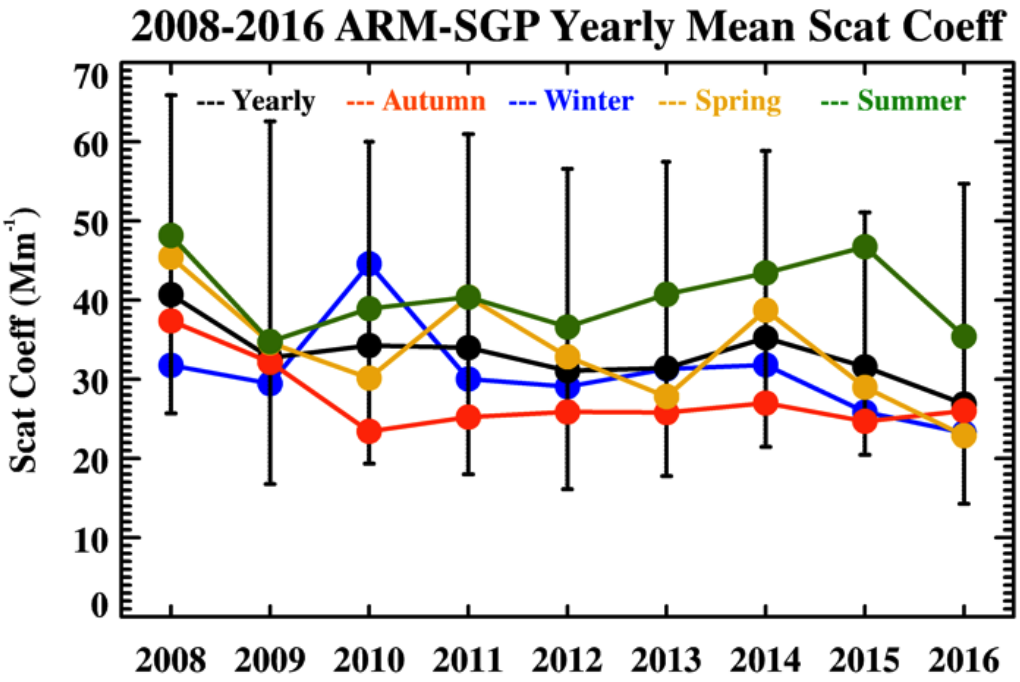

Figure 4. Yearly (black) and seasonal (winter-blue, spring-gold, summer-green, and autumn-orange) mean Aerosol Observation System (AOS) scattering coefficient trend. Note the decreasing trend of mean yearly and seasonal aerosol loading except for the summer season. In addition, the summer maxima in biomass burning smoke aerosols (North American wildfire activity) are also captured by the AOS. The black bars denote the IQR for the year.

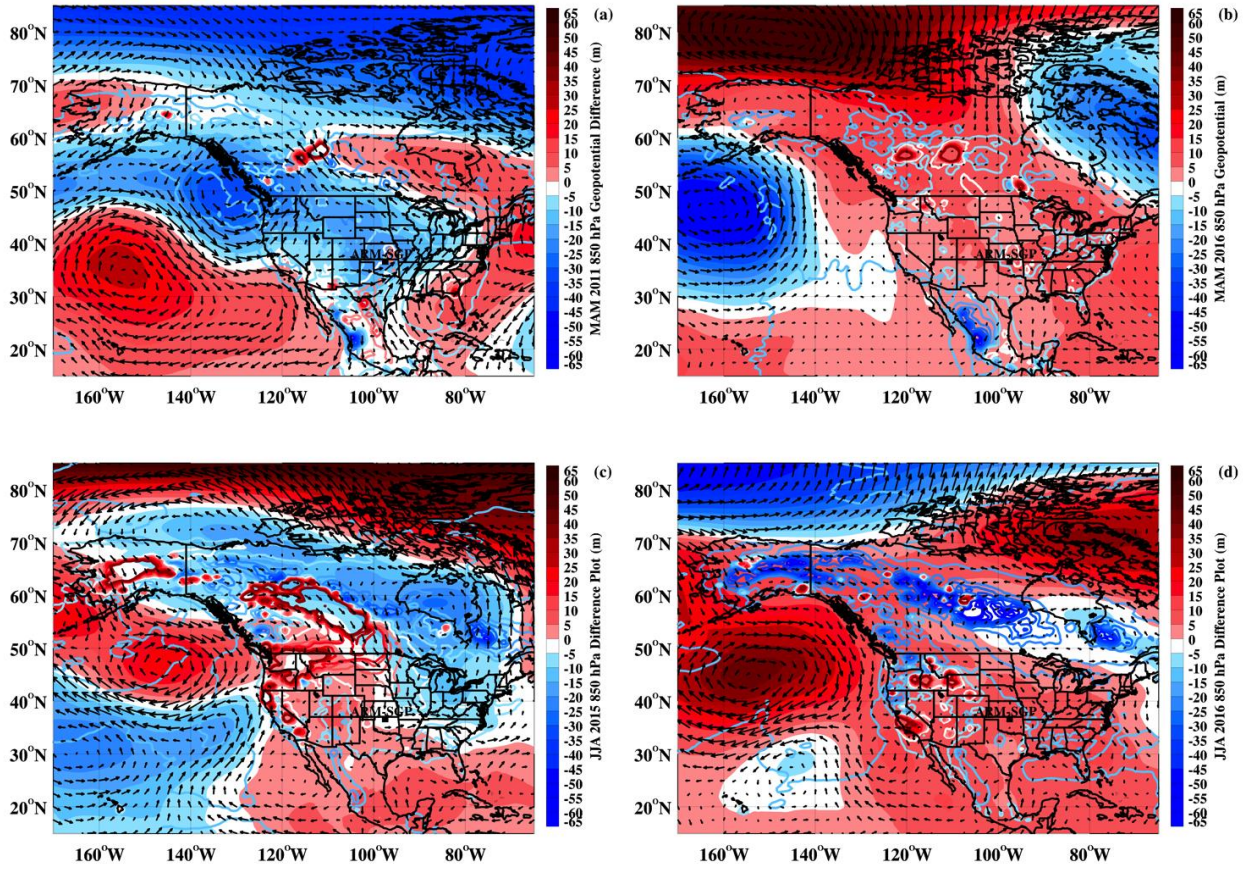

Figure 5. MERRA-2 domain isobaric height anomalies for (a) Spring 2011, (b) Spring 2016, (c) Summer 2015, and (d) Summer 2016. The anomalies are derived by subtracting the 1981-2016 climate domain mean. The red (blue) regions denote positive (negative) height anomalies in which wind motions are anticyclonic (cyclonic) as given by the black arrows. The red (blue) colored line contours denote positive (negative) anomalous MERRA-2 carbonaceous concentrations derived from wildfire activity. Wind data are taken from the NOAA ESRL data repository (https://www.esrl.noaa.gov/psd/data/composites/day/).

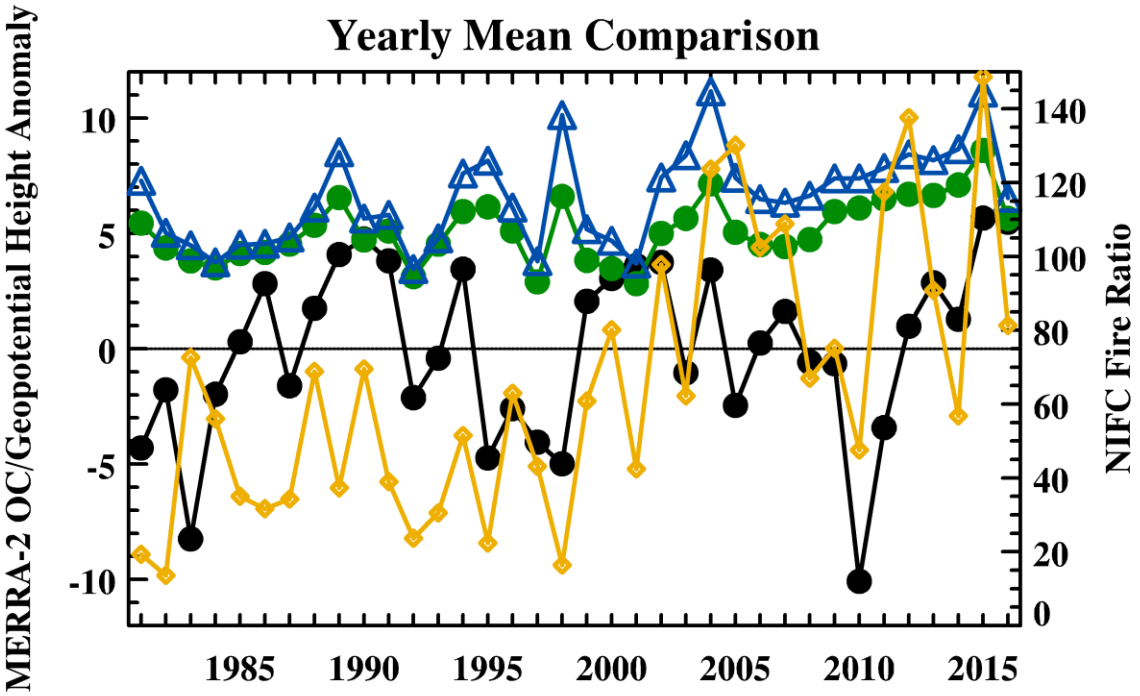

Figure 6. 1981-2016 MERRA-2 domain mean yearly surface organic carbon mass concentration (green circle line), warm season domain mean OC concentration (blue triangle line), and 850 hPa isobaric height domain mean anomaly (black circle line). The National Interagency Fire Center (NIFC) fire number ratio (gold diamond line) is plotted on the secondary y-axis. Note that the NIFC product does not include fire data from Canada and Mexico which likely contributes to the lower correlation with the MERRA-2 products. However, positive isobaric anomalies (increased synoptic ridging) do seem to facilitate conditions of carbonaceous aerosol abundance more often than negative anomalies, especially since 2010.