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Biomass Burning Smoke Climatology of the United States: Implications for Particulate Matter Air Quality

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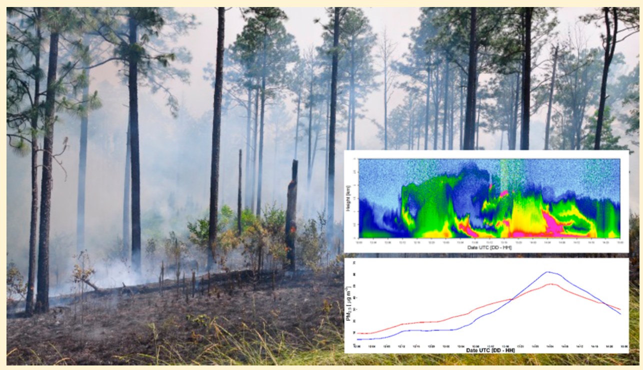
S *Supporting Information*



ABSTRACT: We utilize the NOAA Hazard Mapping System smoke product for the period of 2005 to 2016 to develop climatology of smoke occurrence over the Continental United States (CONUS) region and to study the impact of wildland fires on particulate matter air quality at the surface. Our results indicate that smoke is most frequently found over the Great Plains and western states during the summer months. Other hotspots of smoke occurrence are found over state and national parks in the southeast during winter and spring, in the Gulf of Mexico southwards of the Texas and Louisiana coastline during spring season and along the Mississippi River Delta during the fall season. A substantial portion (20%) of the 24 h federal standard for particulate pollution exceedance events in the CONUS region occur when smoke is present. If the U.S. Environmental Protection Agency regulations continue to reduce anthropogenic emissions, wildland fire emissions will become the major contributor to particulate pollution and exceedance events. In this context, we show that HMS smoke product is a valuable tool for analysis of exceptional events caused by wildland fires and our results indicate that these tools can be valuable for policy and decision makers.

■ INTRODUCTION

Wildland fires, including both wildfires and prescribed burning, are a major source of trace gas and aerosols in the atmosphere.[1−10] Annually, over 25% of primary $PM_{2.5}$ (particulate matter with a diameter less than 2.5 μm) emissions in the United States are from biomass burning.[11] Gaseous emissions in smoke plumes may also contribute to secondary formation of $PM_{2.5}$. Surface air quality degradation from biomass burning can be drastic near the source,[12,13] whereas large, long-lasting burning events can cause regional scale air quality degradation.[14−17] Biomass burning impacts on particulate pollution at the surface is dependent on a variety of factors including size, type, and duration of fires, and atmospheric conditions. Under stable conditions, smoke plumes from smoldering fires can be confined to a shallow atmospheric boundary layer, resulting in enhanced particulate pollution at the surface. In the case of high energy flaming fires, smoke can be injected to higher elevations. Within the convective boundary layer, smoke from both flaming and smoldering fires are mixed through a deeper atmospheric layer and potentially transported thousands of kilometers from the source and impact downwind air quality.[18,19] Long duration and large fires are able to inject smoke even under very stable conditions,[20] factors which also favor long-range transport of smoke plumes by counteracting reductions in concentrations due to atmospheric diffusion and chemical reactions.

Exposure to high concentrations of $PM_{2.5}$ from biomass burning smoke has been linked to increases in respiratory and cardiovascular related hospital admissions and emergency department visits.[21−28] In order to minimize the impact of particulate pollution on human health, the Environmental Protection Agency (EPA) mandates short and long-term particulate pollution primary standards that requires the 3 year averages of the 98th percentile of 24 h and annual average concentrations of $PM_{2.5}$ to not exceed concentrations of 35 μg m^{-3} and 12 μg m^{-3} respectively (78 FR Parts 50, 51, 52, 53, and 58).[29] However, the EPA Exceptional Events rule (81 FR Parts 50 and 51),[30] allows for the exclusion of air quality monitoring data impacted by exceptional events that could not reasonably be controlled when determining nonattainment of prescribed standards. Air quality degradation from wildfires and prescribed burning smoke is considered an exceptional event.[30]

Although emissions from wildfires and prescribed burning are both of relevance to attainment of particulate pollution

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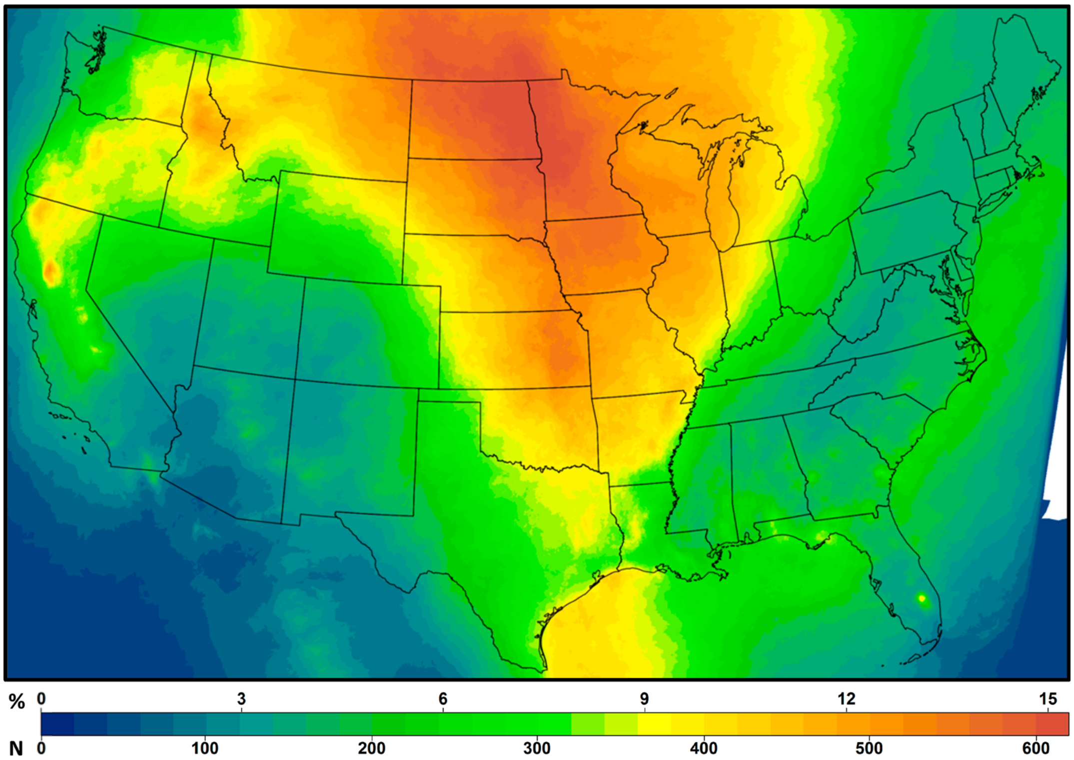

Figure 1. Spatial distribution of the number of smoke occurrence days over the CONUS derived using the HMS analysis for the time period of August 1, 2005 through August 31, 2016.

standards, uncertainties remain in characterizing the geographical distribution of smoke in the United States and its impact on surface particulate air quality. This study presents a satellite based climatology of smoke occurrence for the Continental United States (CONUS) and the implications for surface $PM_{2.5}$ concentrations.

■ DATA AND METHODS

The National Oceanic and Atmospheric Administration (NOAA) National Environmental Satellite, Data, and Information Service (NESDIS) Hazard Mapping System (HMS) Fire and Smoke Product[31−34] is utilized to identify the presence of smoke over a geographic location. The HMS smoke product consists of plumes identified by manual analysis of 1km spatial resolution visible channel imagery (0.63 μm) animations from seven NOAA and National Aeronautics and Space Administration (NASA) geostationary and polar orbiting satellites over the CONUS and adjacent regions. The analysis domain is seasonally adjusted, based on regional climatological burning seasons, to include Central America in the spring and Canada and Alaska in spring through early fall. The ability to identify a smoke plume in the visible channel imagery is dependent on smoke concentration and thus optically thin plumes may not be identifiable. Solar reflectance of smoke aerosols is very small at infrared wavelengths and therefore is utilized to help differentiate between smoke and clouds, especially visually similar cirrus clouds. Smoke is most easily observed at high solar zenith angles, which increases backscatter, of the early morning and late evening hours. Smoke plumes may not be discernible from anthropogenic haze particularly as plumes become optically thin with age and during stagnant atmospheric conditions. A plume must also be larger than the nominal satellite resolution to be identified. These factors combined with inability to identify smoke during cloudy conditions and during nighttime, makes the HMS smoke product a conservative estimate of smoke occurrence.

This study utilizes the HMS smoke product for the time period of August 1, 2005 through August 31, 2016 (133 months) to derive climatology of smoke occurrence over the CONUS region. For each day, the individual smoke polygons are merged creating a daily analysis of smoke areal extent. The daily smoke areal extent analysis is then utilized to the quantify number of days when smoke is present in the atmospheric column over a given location. The number of smoke occurrence days over CONUS is aggregated for the entire analysis period and also for different seasons. The percentage of smoke days for the entire analysis period and different seasons are provided as are percentages of the total smoke days observed during a given season. The time evolution of smoke area is evaluated by aggregating the spatial coverage for each EPA region excluding: (a) Puerto Rico and The Virgin Islands from Region 2, (b) Hawai'i, Guam, American Samoa, The Trust Territories, and Northern Mariana Islands from Region 9, and (c) Alaska from Region 10. The fractional regional monthly areal extent of smoke is calculated by summing the area of smoke covered daily for each month and dividing by the total possible coverage area for the region.

Seasonal atmospheric patterns over the CONUS that influence the transport of smoke are examined using climatologies derived from the North American Regional Reanalysis (NARR).[35] Vector wind fields and geopotential height fields at 850 hPa and 500 hPa (lower and middle troposphere respectively) at 32 km spatial resolution are utilized for this purpose. To aid in describing the transport of smoke, seasonal estimates of fire location densities are derived from MODIS Aqua and Terra (MCD14DL) fire locations.[36,37]

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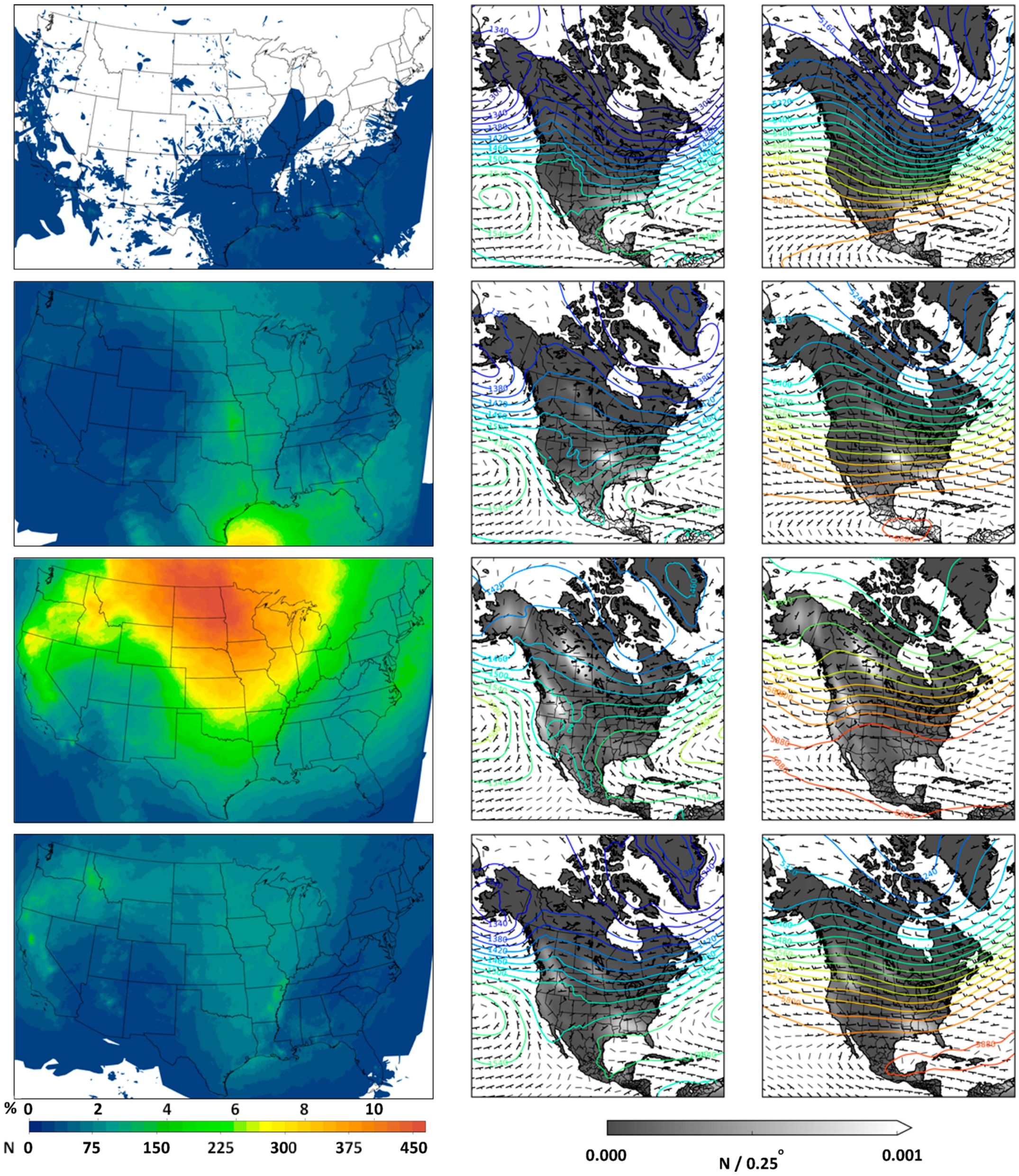

Figure 2. Spatial distribution of number of smoke occurrence days for the winter season (DJF) is show in the left panel on the top row. Winter season climatology of estimated fire density from MODIS (grayscale shading; number of fires per quarter degree), geopotential heights (color contours) and winds (barbs) at 850 and 500 hPa, derived from North American Regional Reanalysis are show in the middle and left panels on the top row, respectively. The second, third and fourth row of panels are the same as the first row, except for spring (MAM), summer (JJA), and fall seasons (SON).

Both federal reference method and acceptable $PM_{2.5}$ AQI (measurement codes 88101 and 88502 respectively) monitoring sites, from the EPA Air Quality System (AQS) repository, with at least two years of data during the study period are utilized in describing the impact of smoke on surface air quality. For colocated monitors, only the primary monitor (Parameter of Occurrence Code = 1) is utilized. Monitors are divided into rural (n = 582), suburban (n = 759), and urban (n = 678) settings. Monitors may collect data continuously but not less frequently than every third day depending on federal monitoring requirements. The fractional regional monthly areas are compared to the regional monthly distributions of $PM_{2.5}$ concentrations, represented by the median (50th percentile) bounded by the 10th and 90th percentiles, for each setting. Whereas the HMS smoke product does not provide information on the vertical placement of the smoke plume, it can be used to identify days when surface $PM_{2.5}$ concentrations are *potentially* influenced by smoke pollution. Using the HMS smoke product, observations of surface $PM_{2.5}$ concentrations are categorized into smoke free or smoke influenced days. Probability density functions (PDFs) of observed $PM_{2.5}$ for smoke free and smoke influenced conditions are computed as a function of season and land use (urban, suburban, and rural). Statistical significance of differences between smoke free and smoke influenced $PM_{2.5}$ distributions tested using the nonparametric Kolmogorov–Smirnov test (p < 0.05). Differences in mean surface $PM_{2.5}$ concentrations between smoke influenced and smoke free days

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and the percentage of smoke influenced days during which the surface $PM_{2.5}$ concentrations exceeded the 24 h federal standard (35 μg m^{-3}) are computed for each station and as a function of season and land use to examine the geographical distribution of the potential smoke impact on surface particulate pollution. Statistical significance is determined from monitor specific Kolmogorov−Smirnov test of smoke and smoke free distributions. Individual monitor analysis is performed only if five or more smoke days were present during the specified season.

■ RESULTS

The spatial distribution of smoke occurrence days for the study period show that the Great Plains states are the most impacted (Figure 1), with a maximum number of ∼600 days potentially influenced by smoke (∼15% of total days during the study period) found over the border region of North Dakota and Minnesota. The local maximum centered on the plains states region extends southward into Oklahoma, Arkansas, and north Texas and is also contiguous to enhanced smoke occurrence found in the western states of California, Oregon, Washington and Rocky Mountain states of Idaho and Montana. Other prominent local maximums in smoke occurrence include a large area in the Gulf of Mexico to the south of Texas coast line and numerous small areas across the southeast. A gradient of smoke occurrence is observed along the coastline in the southern Gulf of Mexico, extending from Louisiana to Florida pan handle. A similar feature is also observed along the Atlantic coast, extending from Florida to Maine. Three dominant local minimum of smoke occurrence are identified, one along the Appalachian region in the Eastern U.S. and two in the Four Corners area and Sonoran Desert regions in the Western U.S. respectively. The local minimum over the Sonoran region extends westwards into southern regions of California and Nevada. The minimum number of smoke occurrence days of ∼80 (2%) in the CONUS is found in the Sonoran desert region.

The above-described pattern of smoke occurrence is a composite of seasonal variations in wildfire and prescribed burns and atmospheric conditions. In order to better understand spatial and temporal variation of smoke occurrence, seasonal patterns are examined (Figure 2). Widespread smoke occurrence is not observed over the United States during the winter months (December, January, February; DJF) (Figure 2, top row and left column), except across the southern states. In specific, winter local maximums of smoke occurrence are observed in western Louisiana (60 days, 15.8% of all smoke days, 1.5% of study period days), in the Florida panhandle and in the Lake Okeechobee regions (135 days, 33.8% of all smoke days, 3.3% of study period days). The local maximum in Louisiana is in the vicinity of the Kisatchie National Forest (Supporting Information (SI) Figure S1), where prescribed burning is used for land management.[38] Enhanced smoke occurrence in the Lake Okeechobee region is associated with the Florida Everglades, where fire is an important ecological[39] and agricultural[40] process. Prescribed burning is responsible for the majority of smoke occurring in the Okeechobee region during the winter.[41,42] Smoke transport to ocean areas is also evident along the coastlines of Louisiana, Florida, Georgia and South Carolina. The flow regimes during the winter months are primarily westerly and thus there is minimal meridional transport of smoke. Localized maximums are consistent with relatively small, short duration controlled fires and plumes that diffuse on daily temporal scales.

Transition from winter to spring (March, April, May; MAM) is characterized by an increase in days of smoke occurrence over the southern states and the Gulf of Mexico, with the maximum number of smoke days increasing from 60 to more than 300 (75% of all smoke days, 7.4% of study period days). Smoke occurrence is most frequent over the Gulf of Mexico, south of the Texas coast. This feature is as result of agricultural burning in the Meso and Central American countries.[7,43−45] The local smoke occurrence maximum over eastern Kansas is associated with tallgrass prairie ecosystem conservation efforts in the Flint Hills.[46] During the spring season, atmospheric flow in the Eastern U.S. is dominated by the establishment of the Bermuda high pressure system to the southeast of Florida.[47] The anticyclonic circulation associated with the Bermuda high transports the smoke from the Central American fires northward toward the Southern U.S., potentially causing the local maxima in Texas and Louisiana and the band of enhanced smoke extending into the Midwest and eastwards from the Gulf Coast into the Atlantic.

Smoke occurrences across the CONUS reaches a seasonal maximum during the summer (June, July, August; JJA). This seasonal smoke maximum coincides with the maximum in the western United States and high latitude boreal forests wildfires.[48] Over the Central and Western U.S., summer smoke accounts for over half of the total smoke days observed (450 days, 75% of all smoke days, 11.1% of study period days). The well-established Bermuda high in the Gulf of Mexico governing flow over the eastern region and an upper level anticyclone dominating the southwest contribute to easterly transport of smoke across the CONUS. This zonal transport combined with a southerly component in the upper troposphere transporting smoke from high latitude North American[49−51] and Siberian fires[52,53] results in a convergence of plumes and the smoke occurrence maximum over the North Central U.S. A relative minimum in smoke is observed over the Appalachian region (120 days, 75% of all smoke days, 3% of study period days). Compared with the western United States and the boreal forest of Canada, relatively few fires occur over the northeastern United States.[48] Given that the smoke over this region is primarily the result of transport from high latitude fires[51,54−56] by southwesterly flow, the local minimum over Appalachia is potentially an artifact of the smoke identification capabilities. Frequent occurrence of cloudiness in this region (SI Figure S2), limiting the identification of smoke plumes, is the potential cause for the minimum in smoke occurrence over Appalachia during summer.

During the fall season (September, October, November; SON) the spatial pattern of smoke occurrence remains similar to summer, but with a reduced number of smoke occurrence days. Maximums in smoke occurrence are found across northern California (150 days, 34.1% of all smoke days, 3.7% of study period days) and other western states (widespread >45 days and up to 135), but the number of observed days have decreased by a factor of 2 from summer. A smoke occurrence maximum is observed in the southwest during the fall (75 days, 41.7% of all smoke days, 1.9% of study period days) as a result of managed ecosystem restoration efforts and wildfires in the ponderosa pine forests.[57] There is a maximum in the Mississippi River Valley (150 days, 35.7% of all smoke days, 3.7% of study period days) caused by agricultural burning.[58]

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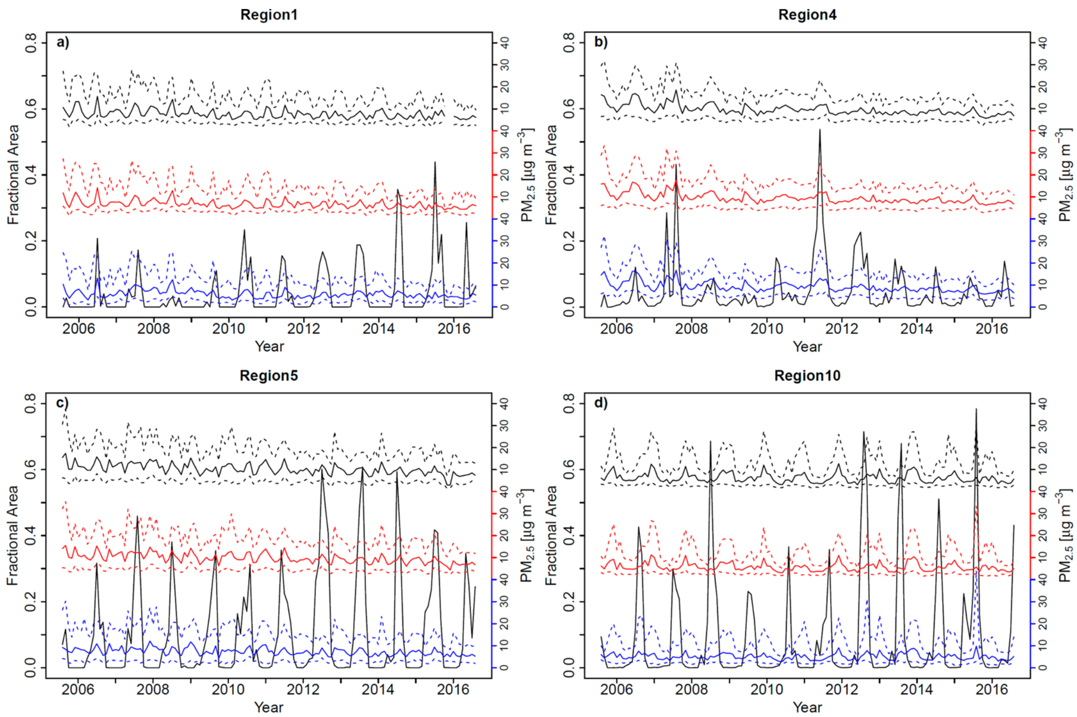

Figure 3. (a) Time series of monthly distributions of observed 24 h average $PM_{2.5}$ concentrations, represented by the median (solid lines) bound by the 10th and 90th percentile of observations (lower and upper dashed lines) for rural (blue line), suburban (red line), and urban (black line) stations in EPA region 1. The proportion of the EPA region that experienced smoke occurrence on a monthly basis is shown using the solid black line. Panels b−d are the same, except for region 4, 5, and 10 respectively. Similar plots for the other EPA regions are shown in Figure 2 included in the Supporting Information.

Climatologically weak winds in the Mississippi River Valley restrict the transport of smoke to near the source.

The temporal variability of smoke and dependence of $PM_{2.5}$ concentrations on the areal extent of smoke is further characterized in Figure 3. Statistically significant downward trends of the median and 90th percentile of the distribution of observed $PM_{2.5}$ are observed for all regions and monitor settings excluding the following combinations for which no trend is observed: (1) rural monitors in region 9 and 10 (50th and 90th percentile of $PM_{2.5}$ distributions), (2) suburban monitors in region 8 and 10 (90th percentile), and (3) urban monitors in region 10 (90th percentile). Concurrently, positive trends in smoke area are observed at both monthly and yearly time scales for all regions. In the Northeast U.S. (EPA Region 1, Figure 3a), distributions of $PM_{2.5}$ concentrations in urban and suburban settings often tend toward higher concentrations during maximums in smoke coverage, however; annually surface $PM_{2.5}$ distributions observed in the winter, when smoke extent is at a minimum, are comparable or have more numerous high observed concentrations. Wintertime pollutant mass concentration maximums are attributed to compounding conditions of reduced atmospheric mixing resulting from a lower planetary boundary layer heights and weaker winds when compared to the other seasons[59] and increased fuel combustion for heat including both large point sources and residential fuel combustion.[60−62] A low bias in the smoke area calculations, resulting from the limited ability to identify smoke in the presence of clouds which are at a maximum in the winter,[63] may alter the perceived lack of impact during the winter. Secondary maximums in smoke extent such as those observed in 2007, 2010, 2011, and 2015 correspond with increases in the 90th percentile of $PM_{2.5}$ across monitor settings.

The areal coverage of smoke is seldom zero in the Southeast U.S. (EPA Region 4, Figure 3b) but large wildfires are atypical and evident only twice during the analysis time period (2007 and 2011). Percentiles of $PM_{2.5}$ are positively correlated to the monthly areal extent of smoke are (R range from 0.49 to 0.59, lag 0; $p < 0.05$). Unlike region 1, distinct increases of $PM_{2.5}$ concentrations distributions in the absence of smoke for all monitor settings are not typically observed during the winter months. Provided there is limited observed plume dispersion (Figure 2), numerous small fires during the winter, with smoke confined to near the surface, have a greater potential to impact $PM_{2.5}$ than fires occurring during the warm seasons in the southeast. Conversely, smoke management practices in the development of prescribed burn practices aid in limiting surface impacts during observed maximums in smoke coverage.

Region 5 (Great Lakes region, Figure 3c) has one of the highest areal coverage of smoke but disproportionately small effects on surface monthly $PM_{2.5}$ are realized, with the 90th percentile of observations never above 17 $\mu g\ m^{-3}$ during peak smoke extents observed during the summers of 2012, 2013, and 2014. Often (i.e., 2014), average $PM_{2.5}$ concentrations are at a maximum during the winter and do not coincide with smoke areal extent maximums. The lack of correlation between smoke and $PM_{2.5}$ concentrations (R ranges from −0.01 to 0.16, lag 0 and lag 1 for the 90th percentile of observations; $p > 0.05$) suggest that the HMS smoke detected in this region are elevated plumes[19,64] that seldom reach the surface. Alternatively, under sampling of $PM_{2.5}$ by monitors that collect data every third day and the aforementioned smoke detection limitation will contribute to lowering correlations.

Distributions of $PM_{2.5}$ in the Northwestern U.S. (Region 10, Figure 3d) reveal distribution features common to regions 1

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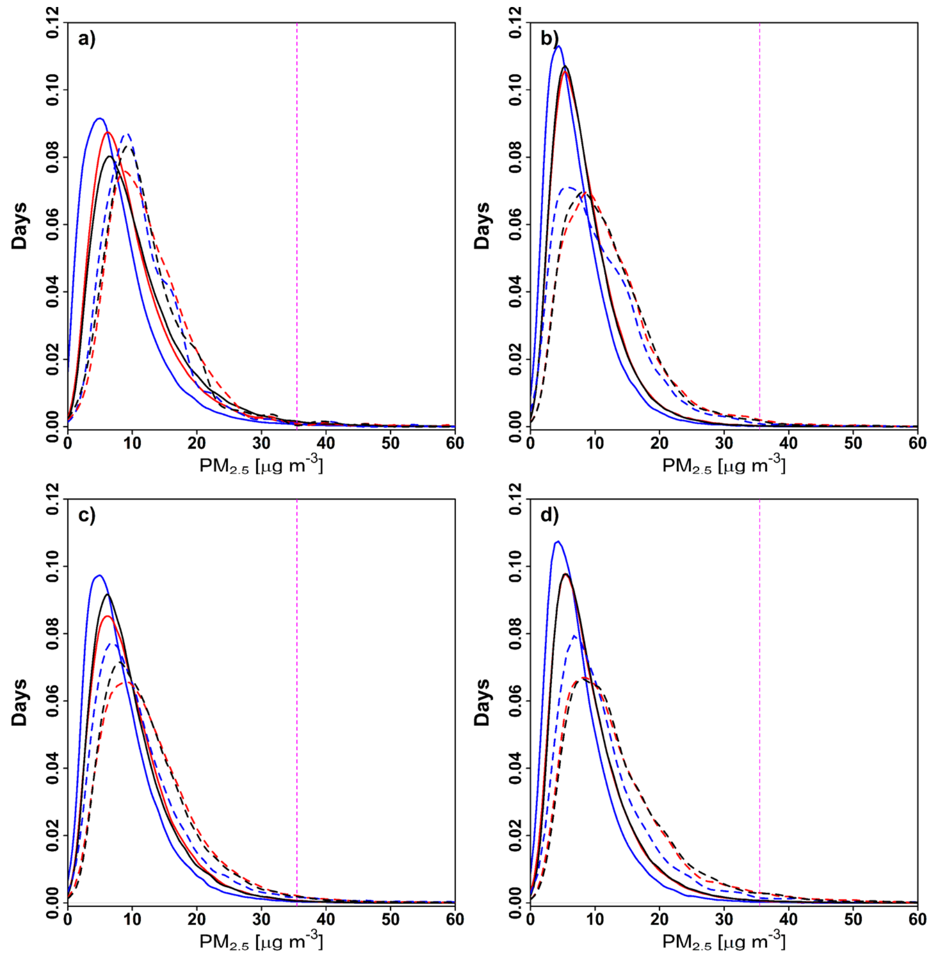

Figure 4. (a) Probability density estimates of the observed 24 h average $PM_{2.5}$ for smoke free (solid line) and smoke influenced (dashed line) conditions for the winter months (DJF). Urban, suburban, and rural observations are shown using black, red and blue colors, respectively. The 24 h average $PM_{2.5}$ federal standard of 35 μg m^{-3} is indicated by the vertical magenta line; (b−d) same as panel a, except for spring, summer, and fall seasons.

Table 1. Summary of $PM_{2.5}$ Concentrations for the United States from August 2005 through August 2016[a]

		with smoke			without smoke		
		N (%)	μ (μg m^{-3})	Mo (μg m^{-3})	N (%)	μ (μg m^{-3})	Mo (μg m^{-3})
rural (number of monitors = 582)	DJF	0.010 (0.74)	11.96	8.90	1.390 (99.26)	8.03	5.01
	MAM	0.137 (29.09)	10.89	5.51	0.335 (70.91)	7.08	4.39
	JJA	1.088 (70.12)	11.83	7.10	0.462 (29.82)	8.30	4.96
	SON	0.347 (45.81)	12.73	6.73	0.411 (54.19)	7.57	4.33
suburban (number of monitors = 759)	DJF	0.012 (0.29)	12.95	8.82	4.105 (99.71)	10.32	6.06
	MAM	0.275 (29.94)	12.38	9.00	0.644 (70.06)	8.38	5.27
	JJA	0.904 (45.43)	13.13	9.55	1.086 (54.57)	9.86	6.05
	SON	0.378 (19.43)	13.53	8.32	1.568 (80.57)	9.18	5.35
urban (number of monitors = 678)	DJF	0.009 (0.21)	12.47	9.23	4.111 (99.79)	11.05	6.33
	MAM	0.159 (22.13)	12.04	8.22	0.560 (77.87)	8.30	5.37
	JJA	0.822 (45.73)	12.78	8.09	0.975 (54.27)	9.65	6.03
	SON	0.379 (19.27)	14.01	8.22	1.588 (80.74)	9.32	5.63

[a]Number of $PM_{2.5}$ concentrations observations that exceed 35 μg m^{-3} (24 h Average NAAQS Standard) per number of monitors (N), mean (μ) of $PM_{2.5}$ concentrations and mode (Mo) of $PM_{2.5}$ concentrations with and without the potential influence of smoke. For each setting and season, the percent of observations with and without the influence of smoke are provided in parentheses.

and 4 but these features are more pronounced. Namely a greater number of elevated $PM_{2.5}$ concentrations are observed in the winter, compared to spring and fall, and when the area covered by smoke is high. Similar to the northeast, the 50th and 90th percentiles of $PM_{2.5}$ concentrations are highest during the winter and as is observed in the southeast, the presence of

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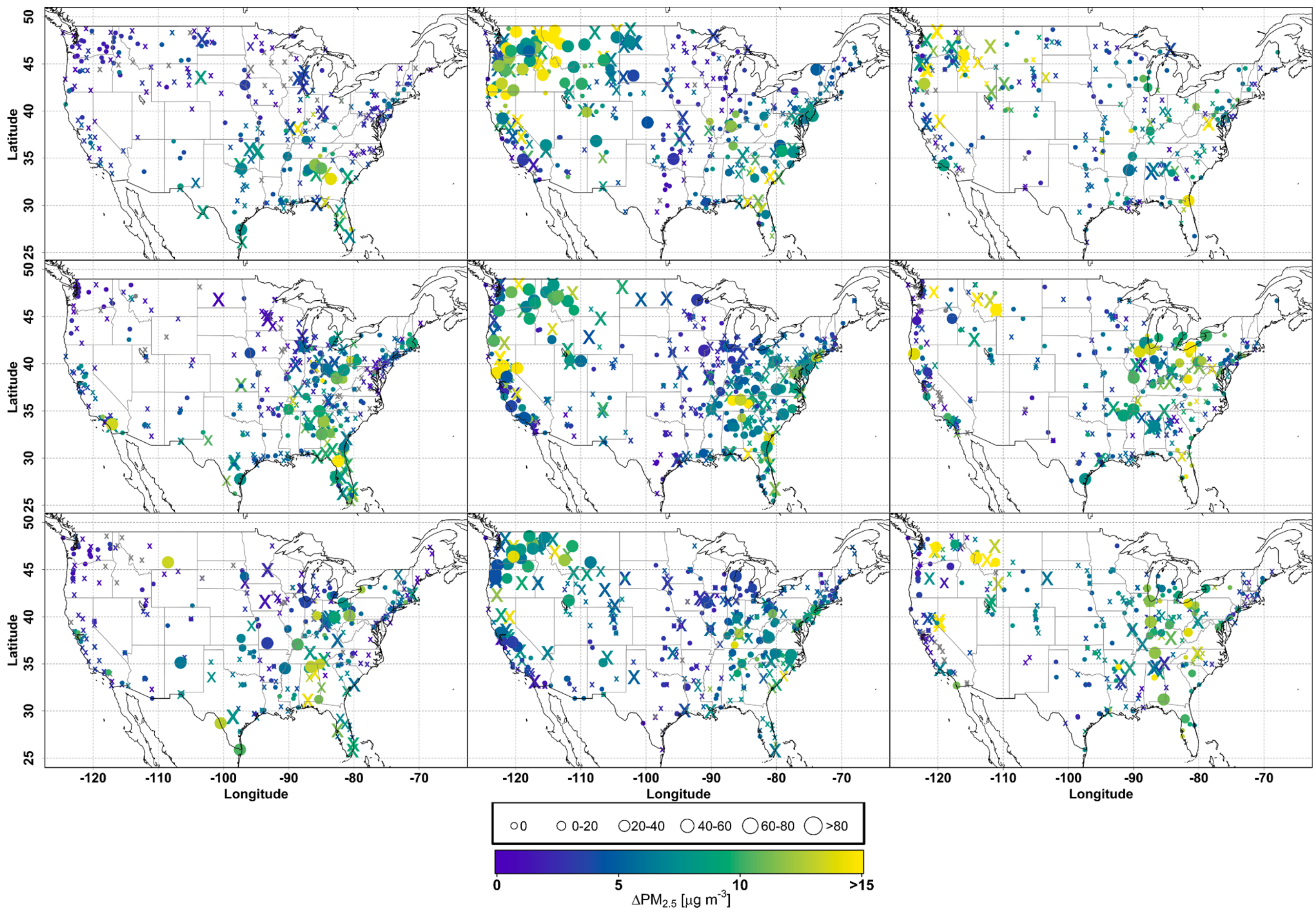

Figure 5. Spatial distribution of number of days of exceedances in the spring season (MAM; left column) that occur when smoke is detected over rural (top row) $PM_{2.5}$ monitors. A circle (O) denotes $PM_{2.5}$ monitors for which there is a statistically significant difference in distributions of $PM_{2.5}$ concentrations between the smoke influenced and smoke free days. An X denotes monitors with nonstatistically significant distributions. The size of the symbol and color shading indicates the number of days of exceedance and the enhancement in mean $PM_{2.5}$ concentration when smoke is present, respectively. Negative enhancements are colored gray. Suburban and urban monitors are shown in the middle and bottom rows, respectively, while the middle and right column are for summer (JJA) and fall (SON) seasons.

smoke can under low ventilation conditions can dramatically alter the distributions. However, there are multiple instances (i.e., summer 2008) where smoke coverage exceeds 30% of the possible area with minimal apparent impact on surface $PM_{2.5}$. These narrow, low concentration distributions are a combination of sparse $PM_{2.5}$ observations in mountainous terrain and elevated smoke plumes. Because of these aforementioned variabilities in smoke and surface observations, the correlations between percentiles of observed $PM_{2.5}$ concentrations and smoke extent is low and highly variable (R = 0.28 and 0.30, −0.12 and −0.01, −0.09, and −0.04; for the 50th and 90th percentiles of 24 h average $PM_{2.5}$ observations for rural, suburban and urban monitors; $p < 0.05$ only for rural monitors). Regional data excluded from Figure 3 can be found in the SI (Figure S3).

The smoke free and smoke influenced $PM_{2.5}$ distributions for all observations, categorized according to seasons and land use, have a positive skew (Figure 4). Application of Kolmogorov−Smimov test show that the smoke influenced and smoke free observations are not draw from the same distribution, and thus the differences between these distributions are statistically significant ($p < 0.05$). The skewness of the distributions are generally enhanced in the presence of smoke thus there is an increase in the percentage of observations with higher surface $PM_{2.5}$ concentrations when smoke is present. Clear sky conditions, which are required for smoke detection, in general are associated with low ventilation and enhanced photochemistry introducing a positive bias in the smoke influence distribution. The maximum change in skew of the distributions due to smoke influence occurs for rural sites. The maximum number of 24 h average observations exceeding 35 $\mu g\ m^{-3}$ are found at rural regions occurs during the summer season and ∼70% of these events occur with the presence of smoke (Table 1). Over urban and suburban regions, the maximum number of exceedances occur during the winter however the number of events associated with the presence of smoke is minimal (<1%). Excluding winter, the highest number of air quality exceedances for suburban monitors occur during the summer time and ∼45% of these events are associated with the presence of smoke. The total number of exceedances over suburban regions remains high during the fall season, but the percentage of smoke influenced events fall to ∼19%. Over urban regions, the total number of exceedances is at a maximum during fall season and slightly higher compared to the summer season. However, the percentage of smoke influenced events are similar to that of suburban regions. The above-discussed results show particulate air quality in rural regions to be most impacted by smoke.

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The spatial distribution of $PM_{2.5}$ enhancement resulting from the presence of smoke also shows differences depending upon season and nature of monitor setting. The total number of exceedance days (Table 1) is highest during the winter, but ~75% of these events are confined to urban and suburban areas. Of these, only a small number of events in the Louisiana and Florida regions co-occur with smoke and monitors that have statistically significant differences in distributions of smoke influenced and smoke free $PM_{2.5}$ concentrations.

During the spring season, differences in mean $PM_{2.5}$ concentrations between smoke influenced and smoke free days (referred as $\Delta PM_{2.5}$ from this point onward) are highest over the Southeastern U.S. (Figure 5). Maximum $\Delta PM_{2.5}$ of ~15 $\mu g\ m^{-3}$ is found in suburban regions of Florida (Figure 5). The percentage of air quality exceedance events during for which smoke was present (referred from here on as smoke influenced exceedance) is also highest in the southeast and at suburban locations. The smoke influenced exceedances in the eastern regions potentially result from prescribed burns with contributions from wildfires across the southeast. A small percentage of smoke influenced exceedances and modest enhancements are observed in the vicinity of the tallgrass prairie burning (Kansas). Smoke influenced exceedances are also found along coastal regions of south Texas and caused by transport of smoke from agricultural fires in Meso and Central American.

Even though the climatological maximum in smoke occurrence is found over the Central U.S. during the summer, there is a minimal impact on the surface $PM_{2.5}$ (Figure 5, middle column). This pattern is consistent with the hypothesis that the majority of the observed smoke is elevated and a result of long-range transport from the Western U.S. and high latitudes. During summer season, the number of smoke influence exceedance days and $\Delta PM_{2.5}$are maximized over the northwest. Over rural regions in the northwest, $\Delta PM_{2.5}$ exceeds 15 $\mu g\ m^{-3}$ at some locations. Several suburban and urban locations along the east coast also show smoke influenced exceedances of the 24 h $PM_{2.5}$ standard. During summer time, ~52% of all exceedance events in the CONUS region occur when smoke is present, whereas it is greater than 80% for individual monitors in the western United States.

The number of monitors experiencing high enhancements (>15 $\mu g\ m^{-3}$) during the fall are fewer than is observed during the summer and almost exclusively confined to the northern Rocky Mountains (Figure 5, right column). Both the number of monitors with exceedances and the number of exceedances with contributions from smoke decrease in the northwest. In the Midwest, $\Delta PM_{2.5}$ is higher in the fall compared to summer potentially due to an increase in upwind prescribed and agricultural burning in the Mississippi River Valley and Central U.S.[40,65] Despite larger enhancements, the percentage of exceedance days in the presence of smoke remains steady in the Midwest. Exceedances across Arkansas, Mississippi, and Alabama almost exclusively occur when smoke is potentially present.

■ DISCUSSION

The importance of fire and smoke management is of concern because of the increasing trend in occurrence of large wildland fire occurrences in the Western U.S. and Canada[66−69] and their projected influence on summertime particulate pollution.[70] Also of concern is the projected growth of the wildland-urban interface leading to more population facing exposure to smoke pollution[71] increasing health risks and care costs.[26,72−74] Despite $PM_{2.5}$ concentrations increasing more at rural sites, compared to urban and suburban, in the presence of smoke (Figures 5), nonattainment designations of the $PM_{2.5}$ NAAQS are overwhelming found in more densely populated (urban and suburban) areas (40 CFR Part 81).[75] Smoke is observed across the entire CONUS during the study period with potential exposure in the most densely populated areas peaking during the summer months (Figure 2). The plumes defined in the HMS database represent a conservative estimate of the extent of smoke due to thorough quality control efforts, including the requirement that the combustion source be identified, the nominal resolution of satellite data (1 km) potentially being too large to identify plumes associated with small agricultural and land management burns, the inability to spectrally identify smoke from underbrush burning (canopy masking), inability to distinguish smoke from anthropogenic haze, and cloud contamination preventing smoke detection from satellites. False identification of smoke is possible but expected to be minimal because of the strict quality measures, including the requirement to positively identify the source, and the infrared spectral properties of clouds compared to smoke aerosols.

Despite these limitations, at least 20.1% of the total daily NAAQS exceedances coincide with identifiable smoke plumes overhead (Table 1). On average, the $PM_{2.5}$ concentrations are higher for exceedance days during which smoke is present over the area. Of the exceedance days influenced by presence of smoke, 449 (12.3%) exceed the 99% confidence interval of the $PM_{2.5}$ PDF without smoke influences. That is concentrations observed on 12.3% of exceedance days potentially influenced by smoke are unlikely to be caused by nonsmoke related factors. The majority (96.4%) of these days occur during the summer and fall (61.2% and 35.2% respectively). Only two such days are found during the winter months despite winter having the highest percentage of exceedance days (44.7%).

Our study shows the utility of the HMS data set in identifying considerations for exceptional event demonstration caused by smoke from wildland fires. As demonstrated through the historical distribution of concentrations under smoke influenced and smoke free conditions, a clear and causal relationship satisfying tier 1 demonstration[76] requirements can be made between smoke from both wildfires and prescribed burning and extreme $PM_{2.5}$ enhancements. Probability density estimates of individual sites may provide the historical context needed for exceptional event identification and demonstration due to smoke. Nationwide summaries (Figure 4) reveal statistical significant differences between identifiable smoke and nonevent days. Calculated enhancements outside the region of maximum smoke frequency (Figure 5), particularly in the southeast, provide confidence that the HMS product can be combined with ground-level monitor data to establish evidence of spatial and temporal concentrations changes due to smoke. Furthermore, near-real time analysis can aid in mitigating smoke impacts especially considering increasing trends in controllable prescribed fire activity[77] (SI Figure S4).

With pollution standards becoming increasing stringent, it is to be expected that average urban/suburban pollutant concentrations will approach regional background (i.e., rural) concentrations. Such patterns are observed with respect to fine particulate matter across much of the southern U.S. (EPA Regions 4 and 6; Figure 3, SI Figure S3) largely in response to decreases in anthropogenic emissions (SI Figure S4). Similarly, pronounced decreases in $PM_{2.5}$ are evident in many regions

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(Regions 1–4) accompanied by minimal changes in areal coverage of smoke and increasing fire emissions during the 12 year period. In contrast, in regions where smoke areal coverage is high the pattern of decrease in the observed $PM_{2.5}$ is less evident. Our analysis does not account for factors such as regional time trends in emissions and transport from anthropogenic sources. However, when combined with continuous decreases in anthropogenic emissions ($PM_{2.5}$ emissions remain near constant while precursors decline) and increases in fire $PM_{2.5}$ and precursors (SI Figure S4), our results suggest that smoke will become a major contributor to air quality exceedances as regulations become more stringent. Concurrently, prescribed burning emissions are generally increasing with respect to total fire emissions (SI Figure S4) indicating the possibility for increased smoke mitigation practices. While continued NAAQS attainment under future regulations may not be possible in many areas without further anthropogenic cuts, scenarios exists in which compliance of future regulations will require nonanthropogenic reductions or, when mitigation efforts are exhausted, increased exceptional event data exclusion. Extrapolating from trends over rural stations, it is possible that smoke may ultimately be a contributor to as many as 40% of all $PM_{2.5}$ exceedances when emissions for other sources are minimized. This will result in federal standard attainment increasingly dependent on the nature of smoke influences and thus the need for tools and analysis techniques to demonstrate exceptional events resulting from such events.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b03292.

Four figures showing 1) the EPA regions and a survey of land cover across North America; 2) average cloud fractions for each season across the United States; 3) the time series of $PM_{2.5}$ distributions and smoke area for EPA regions not provided in Figures 3; and 4) trends in emissions of $PM_{2.5}$ and precursors from anthropogenic sources, wildfire and prescribed burning for each EPA region (PDF)

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Notes

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