Environ. Sci. Technol. **2008,** *42,* 2812–2818

Interannual Variations in PM2.5 due to Wildfires in the Western United States

DAN JAFFE,*,† WILLIAM HAFNER,†,‖ DULI CHAND,† ANTHONY WESTERLING,‡ AND DOMINICK SPRACKLEN§

University of Washington, Bothell, 18115 Campus Way NE, Bothell, Washington 98011, University of California, Merced, California, and University of Leeds, England U.K.

Received November 1, 2007. Revised manuscript received January 22, 2008. Accepted January 28, 2008.

In this study we have evaluated the role of wildfires on concentrations of fine particle ($d < 2.5$ μm) organic carbon (OC) and particulate mass (PM2.5) in the Western United States for the period 1988–2004. To do this, we examined the relationship between mean summer PM2.5 and OC concentrations at 39 IMPROVE sites with a database of fires developed from federal fire reports. The gridded database of area burned was used to generate a database of biomass fuel burned using ecosystem-specific fuel loads. The OC, PM2.5, and fire data were evaluated for five regions: Northern Rocky Mountains (Region 1), Central Rocky Mountains (Region 2), Southwest (Region 3), California (Region 4), and Pacific Northwest (Region 5). In Regions 1, 2, and 5, we found good correlations of seasonal mean PM2.5 concentrations among the sites within each region. This indicates that a common influence was important in determining the PM concentration at all sites across each region. In Regions 1 and 2, we found a significant correlation between PM2.5 and both the area burned and biomass fuel burned in each region. This relationship is statistically significant using either the area burned or fuel burned, but the correlations are stronger using the biomass fuel burned. In all five regions we found a statistically significant relationship between biomass burned and organic carbon. Using these relationships, we can estimate the amount of PM2.5 due to fires in each region during summer. For the Regions 1 through 5, the average summer-long enhancement of PM2.5 due to fires is 1.84, 1.09, 0.61, 0.81, and 1.21 μg/m^3, respectively, and approximately twice these values during large fire years.

Introduction

The hot, dry summers of the Western United States make the region susceptible to large forest fires. As a natural phenomenon, fire serves an important ecological role. In terms of air quality, fires are significant sources of various gases and aerosols. There are numerous examples in the literature of the role that fires play on short-term air quality (*1–5*). There are also multiple examples of a significant influence from fires on air quality at great distances from the source region for both particulate matter (e.g., refs *6 and 7*) and gases, such as CO and O_3 (e.g., refs *8–11*).

The contribution of fires to particulate matter less than 2.5 μm diameter (PM2.5) depends on the size and duration of the fire, as well as the burning conditions. Dramatic, short-term increases in PM2.5 concentration due to fire have been observed on a local scale, while large fire seasons are responsible for broader, season long increases in PM2.5 over much larger regions.

Regulated by the Clean Air Act for public health reasons (*12*), concentrations of PM2.5 have been decreasing at many sites in the United States over the last two decades. Most of this decrease is attributed to reduced industrial emissions (*13*). However, at many sites, natural sources, including fires, are a significant portion of the total PM2.5 concentration and these sources can vary dramatically from year to year. The importance of fires is further highlighted by recent studies showing that fires in the Western U.S. and Canada have been increasing, both in frequency and duration, over the last few decades. This trend is linked to global climate change, which has resulted in warmer temperatures, reduced winter snowpacks, and longer fire seasons (*14, 15*). If fires continue to increase in severity, the downward trend in PM2.5 concentrations may cease and concentrations in some regions will increase.

In 1999, the U.S. EPA issued the Regional Haze Rules (RHR) to address visibility impairment in Class I areas of the United States (National Parks and designated wilderness). Visibility is impacted by the aerosol loadings, chemistry, and humidity. These rules require that natural visibility conditions be achieved in these areas by 2064. Progress toward this goal must be demonstrated by 2018. In the RHR, natural visibility conditions are defined as "the long-term degree of visibility that is estimated to exist in a given mandatory Federal Class I area in the absence of human-caused impairment" (*16*). The EPA suggests that states utilize a uniform natural visibility of 9.6 deciviews in the Eastern U.S. and 5.3 deciviews in the Western U.S., which includes the natural component of fires. States may develop a different definition of natural conditions, based on more detailed data.

While forest fires are usually thought to be natural, this is not always the case. Forest fires can be ignited by lightning, campers, vehicles, or industrial activities. In addition, climate change may be increasing the frequency of large fires in the Western United States (*14*). Thus, it is becoming more difficult to distinguish whether a fire is natural or human-caused. Nonetheless, quantification of the PM2.5 influence from fires remains important to our overall understanding of air pollution in the Western U.S.

Organic carbon (OC) is a primary component of the PM emitted by fires. In one study the elemental carbon (EC) to organic carbon ratio was found to be a good tracer of smoke from fires (*17*). The EC/OC emission ratio from wildfires is in the ranges of 0.065–0.14 (*18, 19*). Using the GEOS-CHEM global model, Spracklen et al. (*10*) were able to reasonably reproduce the variability in annual mean OC and EC concentrations measured by the IMPROVE network (*21*) averaged over the Western U.S. Using the same model and satellite constrained fire emissions, Park et al. (*22*) estimate emissions of 0.6 Tg of OC for U.S. wildfires in 1998, which is larger than the fossil fuel emissions of OC. Park et al. (*21*) extended their analysis using PM data from the IMPROVE network of observations in the U.S. (*22*). Using the ratio of observed fine particle total carbon to "non-soil" potassium, Park et al. (*23*) were able to estimate the contribution of fires to total carbon concentrations at each site in the IMPROVE

* Corresponding author e-mail: djaffe@u.washington.edu.
† University of Washington, Bothell.
‡ University of California, Merced.
§ University of Leeds.
‖ Now at SAIC, Inc.

10.1021/es702755v CCC: $40.75
Published on Web 03/13/2008

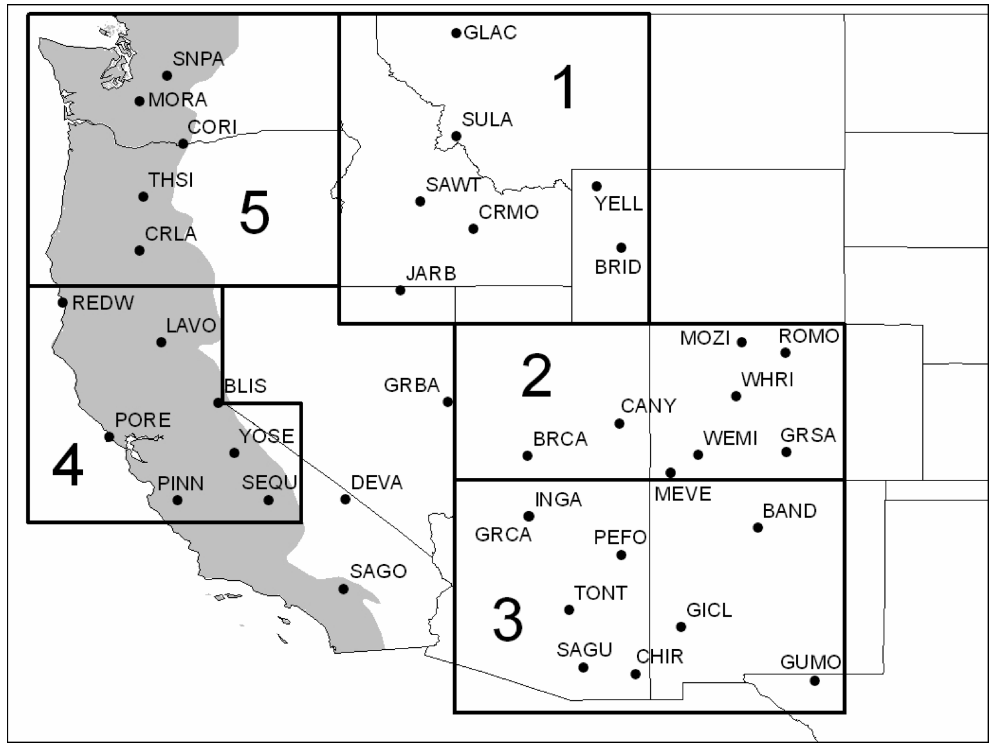

FIGURE 1. Map of the IMPROVE sites used in the analysis. Site names and coordinates can be found in Table S1 in the Supporting Information. Boundaries for the regions are included. Grey shading represents the humid temperate domain; white is the dry domain.

network for the years 2001–2004. As part of our analysis, we have derived a similar quantity and compare our results with those of Park et al. (*23*) in the Discussion section of this paper.

The goals of this paper are to (1) identify regions where multiple sites show a high degree of correlation in seasonal mean PM2.5; (2) quantify the extent to which these sites are influenced by fires; and (3) develop quantitative relationships that can be used to predict seasonal mean PM2.5 and OC enhancements due to fires.

To do this, PM2.5 and OC concentrations are correlated to the area burned and fire emissions for five regions in the Western United States. In a separate analysis, we have evaluated the role of fires on ozone (O_3) concentrations in the region as well (*25*).

Experimental Methods

Area Burned and Biomass Consumption. The forest wildland fire database was developed based on reports from multiple government agencies, including the U.S. Forest Service, Bureau of Land Management, National Park Service, and Bureau of Indian Affairs. These reports incorporate approximately 90% of all wildland area burned reported for the Western U.S. (*22*). The database spans from 1980 through 2004 with a 1° × 1° resolution ranging from 101°−125° West longitude and 31°−49° North latitude (*14*). In each grid cell, the number of acres burned was reported for the month of the fire start date. Fires which burned over the monthly divisions were only accounted for once. In other words, a fire that burned from July 29 through August 5 would be fully assigned only to the month of July. This results from the fact that the fire reports have only consistently reported start dates. Since much of the annual area burned comes from a number of large fires, this simplification can pose a problem if monthly PM data are used. For this reason, we combined fire and PM data for the summer months of June, July, and August. These three months are responsible for 70−93% of annual acres burned in the Western U.S., depending on the year.

One potential problem with the use of area burned for our analysis is that this does not consider variations in emissions from different biomass types: forest fires consume more biomass and emit more PM2.5 per acre burned than grass fires. Using ecosystem-specific fuel loadings and maps of the regional ecosystem type from the U.S. Forest Service (http://www.fs.fed.us/pnw/fera/fccs/maps.shtml), the area burned was converted to kilograms of biomass burned in each 1° × 1° grid cell. This conversion was done assuming that fires occur with 25% high, 25% medium, and 25% low severity. The final 25% was assumed to be unburned (*27*).

PM2.5 and OC Data. The Interagency Monitoring of Protected Visual Environments (IMPROVE) network began making particulate matter measurements in 1988 at nearly 200 sites across the United States ((*27*); http://vista.cira.colostate.edu/improve/). Samples are collected for 24 h, 2–3 times per week, and analyzed for fine (particle diameter, $d < 2.5\ \mu$m) and coarse mass ($d = 2.5$–$10\ \mu$m), as well as an array of chemical species on the fine aerosol. PM2.5 is determined by filter weights and fine particle ($d < 2.5\ \mu$m) OC is determined by a multistep thermal oxidation to CO_2 (*27*). We used data from 39 of these sites that have more than 10 years of observations and fall within the spatial range of the fire database. Figure 1shows a map of the 39 sites and Table S1 (in the Supporting Information) provides the full names, coordinates, and other information.

Prior to 2001, an IMPROVE sample was collected at each site two days per week. Starting in 2001, this was changed to every three days. For each year, PM2.5 and OC mass concentration at each site were averaged for the summer months (June, July, and August). A total of up to 31 samples could be collected for the 3-month period, but at most sites a few samples were missed. We excluded the summer mean for a site if fewer than 11 samples were included in the average. Because of the 3−4 day delay between samples, some fires may be missed. In addition, even fires that are near a sampling site may be missed, depending on local winds. However, the largest fires, which make up the majority of biomass consumption in high fire years, can burn for several days or

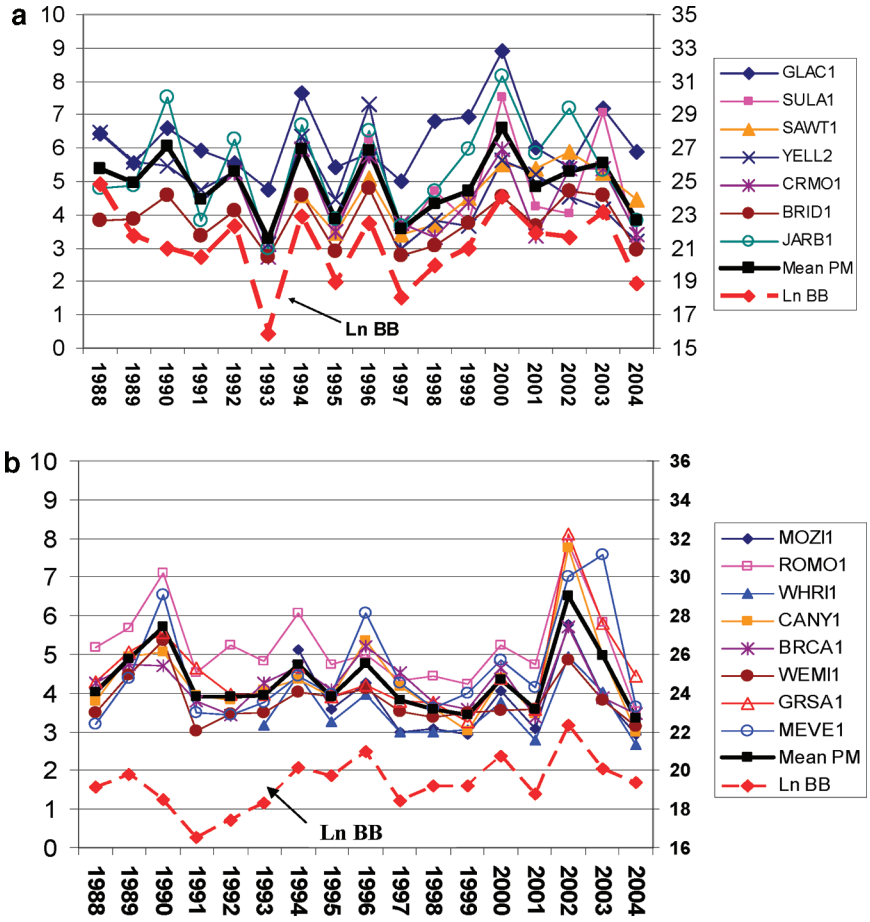

FIGURE 2. (a) Summer (June−August) PM2.5 in $\mu g/m^3$ for 7 sites in the Northern Rocky Mountains (Region 1), left axis. Also shown is the natural log of the biomass burned by fires (Ln BB) in this region for each summer (right axis). The overall summer mean PM2.5 in the region is 4.93 $\mu g/m^3$ with a range of 3.3–6.6 $\mu g/m^3$. (b) Summer (June−August) PM2.5 in $\mu g/m^3$ for 8 sites in the Central Rocky Mountains (Region 2), left axis. Also shown is the natural log of the biomass burned by fires (Ln BB) in this region for each summer (right axis). The overall summer mean PM2.5 in the region is 4.3 $\mu g/m^3$ with a range of 3.4–6.5 $\mu g/m^3$.

weeks (*3*), and therefore result in widespread PM enhancement. So we hypothesize that the regional averaged summer mean PM2.5 and OC concentrations should correlate with large fires in the Western United States.

Results

Figure 1 shows a map of the IMPROVE sites used in this analysis and the boundaries of the five regions used in our analysis. Figure 2a and b show the summer mean PM2.5 concentration in the Northern Rocky Mountain (Region 1) and Central Rocky Mountain regions (Region 2), respectively. Both also show the natural log of the biomass burned by fires within each region, which is discussed below. In Regions 1 and 2, the seasonal mean PM2.5 concentrations show excellent correspondence. Figure 3 is a correlation matrix that shows the significant correlations between summer PM2.5 at each site with all other sites. Significant correlations were defined as those with a P value less than 0.05, the correlation coefficient depends on the number of data points considered. As a point of comparison, summer mean PM2.5 data from Denali National Park in Alaska were included, and found not to be significantly correlated with any of the sites we examined in the Western United States (Denali data are not shown).

Within Regions 1, 2, and 5, most sites show a significant correlation ($P < 0.05$) with most other sites in the region. An exception is Crater Lake in Region 5, which shows a poor correlation with other sites in the Pacific Northwest and a better correlation with two sites in California. The significant correlation in seasonal mean PM2.5 within one region suggests that there are large-scale factors responsible for these interannual variations across the region. Regions 3 and 4 have fewer significant correlations between sites. This suggests that local influences play a greater role in explaining the interannual variations at these sites. There are also numerous significant correlations outside of one region. For example Craters of the Moon (CRMO1) and Bridger Wilderness (BRID1) show numerous significant correlations with sites in Colorado. This suggests that transport of PM from one region to the other is likely also important.

Our hypothesis is that fires play a primary role in explaining the interannual variations in PM2.5 concentrations. Therefore, it is important to group sites based on their geographic proximity as well as their correlation in seasonal mean PM2.5. For Regions 1, 2, and 5, the choice of regional boundaries is fairly straightforward. However, for the Southwest and California regions, the poor correlation between sites suggests that any regional boundaries will be somewhat arbitrary.

Comparison of PM2.5 and OC Concentrations with Fires by Region. The IMPROVE sites were grouped by region, as shown in Figure 1. To evaluate the role of fires, we calculated the number of acres burned and the quantity of biomass burned (kg) in each region for each summer period. Between 1988 and 2004, these five regions accounted for 88% of the biomass burned in the entire Western U.S. The lowest percentage occurred in 1997 (34%), a year with a very low amount of burning in the Western U.S. In all other years,

	Pacific NW					California								N. Rocky Mtns								Central Rocky Mtns								Southwest								
	SNPA1	MORA1	CORI1	THSI1	CRLA1	REDW1	LAVO1	BLLS1	PORE1	YOSE1	PINN1	SEQU1	SAGO1	DEVA1	GLAC1	SULA1	SAWT1	YELL2	CRMO1	BRID1	JARB1	GRBA1	MOZI1	ROMO1	WHRL1	CANY1	BRCA1	WEML1	GRSA1	MEVE1	GRCA2	INGA1	BAND1	PEFO1	TONT1	GICL1	SAGU1	CHIR1	GUMO1
SNPA1	1	1		1		1						1	1																									
MORA1	1	1	1	1								1	1																									
CORI1		1	1	1																																		
THSI1	1	1	1	1							1	1																										
CRLA1					1		1	1		1		1		1						1		1	1	1	1	1	1	1	1	1		1				1		
REDW1	1					1					1		1																		1							
LAVO1					1		1	1		1				1											1	1	1		1		1						1		
BLIS1					1		1	1						1																				1					
PORE1									1					1								1	1			1	1					1			1			
YOSE1					1		1			1		1						1	1	1		1					1				1	1						
PINN1				1		1					1													1				1										
SEQU1	1	1		1	1					1		1						1				1								1	1							
SAGO1	1	1				1							1																			1	1					
DEVA1					1		1	1	1					1			1			1		1	1	1	1	1	1	1	1									
GLAC1															1	1			1	1	1																	
SULA1															1	1			1	1	1														1	1		
SAWT1														1			1		1	1	1	1		1					1								
YELL2										1		1						1	1	1	1	1																
CRMO1										1					1	1	1	1	1	1	1	1	1	1	1					1					1			
BRID1					1					1				1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1				1		1	
JARB1															1	1	1	1	1	1	1	1	1	1	1			1		1								
GRBA1					1				1	1		1		1			1	1	1	1	1	1	1	1	1	1	1	1	1	1	1					1	1	
MOZI1					1				1					1				1	1	1	1	1	1	1	1	1	1	1	1					1			
ROMO1					1						1			1			1		1	1	1	1	1	1	1	1	1	1	1	1		1	1	1	1	1	1	1
WHRI1					1		1							1					1	1	1	1	1	1	1	1	1	1	1			1		1	1	1	1
CANY1					1		1		1					1						1	1	1	1	1	1	1	1	1	1			1	1	1	1	1	1
BRCA1					1		1		1	1				1						1	1	1	1	1	1	1	1	1	1	1		1		1	1	1	
WEMI1					1						1			1						1	1	1	1	1	1	1	1	1	1	1		1	1	1	1	1	1
GRSA1					1		1							1						1		1	1	1	1	1	1	1	1				1	1		1	1
MEVE1					1									1			1		1	1	1	1	1	1	1	1	1	1	1				1	1	1	1	1
GRCA2					1		1			1		1								1		1		1			1	1		1		1						
INGA1						1				1		1	1									1									1							
BAND1					1				1				1											1	1	1	1	1		1		1		1		1	1	
PEFO1								1																1		1		1	1				1				1	
TONT1																1				1	1		1	1	1	1	1	1	1	1			1		1	1	1	1
GICL1									1							1						1			1	1	1	1		1				1	1	1	1	1
SAGU1					1		1													1		1	1	1	1	1	1	1	1			1		1	1	1	1	1
CHIR1																							1	1	1		1	1	1			1		1	1	1	1	1
GUMO1																																						1

FIGURE 3. Correlation of PM2.5 concentrations among sites. Correlation among annual summer mean (June−Aug) PM2.5 for 39 sites in the contiguous United States. A "1" indicates the correlation is significant with $P \leq 0.05$. All significant relationships had positive correlations. The regional groupings, shown in Figure 1, are highlighted with bold lines.

these five regions accounted for at least 70% of the total biomass burned in the Western U.S. For this time period, Regions 1 through 5 contained 50%, 4%, 2%, 10%, and 23%, respectively, of all biomass burned by fires each summer in the Western U.S.

Regional PM2.5 and OC concentrations were calculated as the average of the individual site averages for each summer. Area burned and biomass burned were calculated as the sum over each summer period. An analysis of the area burned and biomass burned data shows that for all regions the data are not normally distributed. This reflects the fact that a few years have a much larger amount of burning. The annual PM2.5 data for each region are very close to a normal distribution. For OC, the data are also normally distributed with the exception of one outlier (Region 4, 2002), which had very high OC concentrations at several sites in the region. In this year, the sites in Northern California appear to have been strongly influenced by the Biscuit fire in Southern Oregon/Northern California.

For this reason, all further calculations on both area burned and biomass burned are computed on the natural log of these quantities. Annual summer mean PM2.5 and OC concentrations along with annual fire data for each region are given in Table S2 in the Supporting Information.

Figure 2a and b show time series of the summer PM2.5 concentration and biomass burned by fires for the Northern Rocky Mountain and Central Rocky Mountain regions, respectively. Table 1 gives the regression parameters for PM2.5 and OC in each region with the natural log of biomass burned and area burned. These regressions were calculated using Ordinary Least Squares (OLS), which assumes that errors in the X values are small compared to errors in Y. We also computed regressions using the reduced major axis method (RMA) method (*30*) and found that the slopes were significantly (30%–50%) greater. However, consideration of the data suggests that the Y values (regional mean PM concentration) do have a much greater uncertainty. This is because the regional average PM or OC concentration is based on only a small sample of points in each region. In a later section we estimate the uncertainty in our PM values based on the regression slope uncertainty, and these uncertainties overlap with the PM values that would be calculated by using RMA regression. Note that because the correlations are calculated on the natural log of burned area or biomass consumed, the intercepts can not be used to give information on the background concentrations in the absence of fires.

Using OLS regressions, Regions 1, 2, and 4 show statistically significant relationships between PM2.5 and biomass burned. For Regions 3 and 5, a statistically significant relationship between fires and PM2.5 is not found. In Regions 1 and 2, statistically significant relationships are found using either biomass burned or area burned; however, in both cases the regressions have a greater R^2 and lower P value using biomass burned. This indicates that correction of the area burned by ecosystem type is a useful procedure to estimate the overall impact of fires in the Western U.S.

In Region 4 (California) the relationship between fires and PM2.5 was relatively weak (R^2 of 0.25). This is somewhat surprising given that, in some years, there are significant fires in the region. However, for the California sites the mean summer PM2.5 concentration over this period was 6.6 μg/m^3, which is almost 2 μg/m^3 higher than other regions. In

TABLE 1. Regression Parameters from Correlations of PM2.5 and OC vs Biomass Consumption and Area Burned (Only Significant Correlations ($P < 0.05$) are Shown)

	PM2.5 vs Ln biomass burned				PM2.5 vs Ln area burned			
	slope ($\mu g/m^3$/ln kg)	intercept ($\mu g/m^3$)	R^2	*P* value	slope ($\mu g/m^3$/ln acre)	intercept ($\mu g/m^3$)	R^2	*P* value
Region 1	0.391	−3.25	0.69	<0.01	0.453	−0.641	0.60	<0.01
Region 2	0.354	−2.53	0.32	0.018	0.342	0.462	0.24	0.047
Region 4	0.312	0.333	0.25	0.046		not significant		

	OC vs Ln biomass burned				OC vs Ln area burned			
	slope ($\mu g/m^3$/ln kg)	intercept ($\mu g/m^3$)	R^2	*P* value	slope ($\mu g/m^3$/ln acre)	intercept ($\mu g/m^3$)	R^2	*P* value
Region 1	0.154	−1.71	0.51	<0.01	0.170	−0.568	0.40	<0.01
Region 2	0.171	−2.31	0.62	<0.01	0.173	−0.952	0.50	<0.01
Region 3	0.141	−1.60	0.28	0.03		not significant		
Region 4	0.0986	−0.450	0.19	0.08		not significant		
	0.0752[a]	−0.435[a]	0.28[a]	0.03[a]				
Region 5	0.180	−2.11	0.26	0.04		not significant		

[a] One outlier removed (2002). Regions 1−5 are the Northern Rocky Mtns., Central Rocky Mtns., Southwest U.S., California, and the Pacific Northwest, respectively.

addition, the IMPROVE sites in California have the highest percent nitrate contribution to fine mass compared to other sites in the west. The calculated NH_4NO_3 contribution (as reported in the IMPROVE database) to total fine mass for summer at the California sites is 11%, whereas it is 3.7% for all other sites in the Western U.S. Thus, it is likely that regional anthropogenic sources are much more significant in California and this obscures the relationship with fire (*31*).

For the Pacific Northwest (Region 5), we speculate that the locations of the IMPROVE sites may miss emissions from some of the largest fires in the region. For example, in 2001 large fires burned in the North Cascades of Washington State and in south-central Washington State. During 2002, large fires occurred in the North Cascades as well as in Southern Oregon (the Biscuit fire). While PM from the Biscuit fire in 2002 fires was clearly seen at Crater Lake, most of the other sites in the Pacific Northwest were not as strongly influenced. The smoke from the fires in 2001 and 2002 appears to have missed most of the IMPROVE sites in the Pacific Northwest, which are located near the crest of the Cascade Mountain range in Washington and Oregon. These locations tend to be less directly influenced by fires in the North Cascades. In addition, some of these sites, such as Snoqualmie Pass and Mt. Rainier, are directly downstream from large urban areas. The Pacific Northwest region can also be influenced by fires in Canada, Alaska, and even Siberia (*10, 11, 32*).

In contrast, using OC we found a statistically significant relationship with fire biomass burned in all regions. This reflects the fact that OC is a better tracer of fires than PM2.5, which has a greater array of sources. As with PM2.5, we found the OC correlations better in all regions using biomass burned compared to area burned. By comparison of the slopes in Regions 1 and 2, we found that OC makes up, on average, 43.8% of PM2.5.

The average and maximum summer enhancement in PM2.5 from fires across each region can be estimated from the regression parameters. To do this, we assume the lowest fire year in each region represents a near-zero contribution to PM2.5, and compare this with the PM2.5 values calculated from the regression equation for the maximum and mean fire years. We used the regressions between OC and biomass burned, since in general, these had the highest R^2 values. This was combined with the fact that OC makes up 43.8% of PM2.5 in fires. Results are shown in Table 2. Clearly, this procedure has greater uncertainty for Regions 3, 4, and 5 where the regression parameters were weaker and vegetation types differ. As mentioned above, the weaker regression statistics can be explained by influence from urban pollution or fires outside of our domain. We estimate the uncertainty from the standard error in the regression slopes. These values are also given in Table 2.

TABLE 2. Mean and Maximum Summer-Long Enhancement in PM2.5 from Fires[a]

	mean enhancement in PM2.5 ($\mu g/m^3$)	max enhancement in PM2.5 ($\mu g/m^3$)	uncertainty (1 sigma)
Region 1	1.84	3.14 (1988)	25%
Region 2	1.09	2.26 (2002)	20%
Region 3	0.61	1.24 (2003)	41%
Region 4	0.81	1.28 (1999)	54%
Region 5	1.21	2.44 (2002)	44%
mean	1.11	2.07	

[a] Values are calculated from OC−Ln biomass burned regressions, shown in Table 1, the minimum, mean, and maximum fire year for each region, and using the observed OC/PM2.5 ratio of 0.438 (from the slopes in Regions 1 and 2). Regions are the same as those in Table 1 and shown in Figure 1. The mean is the average of all years and the maximum year is noted in parentheses.

In Regions 1 and 2 (Northern Rocky Mountains and Central Rocky Mountains), the mean PM2.5 enhancements are 1.84 and 1.09 $\mu g/m^3$, respectively, and approximately twice this amount during the largest fire years. The good regression between sites in both regions indicates that summer mean PM2.5 concentrations around the region (outside of urban areas) are relatively uniform. The uncertainty in the fire contribution to PM2.5 is 25% and 20%, respectively. In Region 3 (Southwest), the regression with regional fires is only significant using OC and the R^2value is only 0.28. The calculated PM2.5 concentration in this region is significantly lower and the uncertainty is greater (41%). In Region 4 (California) the correlation between sites is not as strong. Thus, the calculated PM values have greater uncertainty. Finally, in Region 5 (Pacific Northwest) four of the five sites show excellent correspondence between summer mean concentrations; however, the R^2 for the regression with regional fires and PM2.5 is not significant. The correlation between regional fires and OC is significant with $P = 0.04$. We believe the most likely explanation is that fires outside of our domain play a significant role in accounting for the interannual variations. Thus, our calculated PM2.5 contribution in this region due to fires is likely an underestimate.

Discussion

Using the ratio of nonsoil potassium to total carbon, Park et al. (*23*) calculated the contribution from fires at IMPROVE sites throughout the United States. In their analysis, they separately identify the influence from summer wildfires, prescribed fires, and biofuel. In the Western U.S. during summer, they conclude that wildfire is the dominant fire type and contributes 0.26 $\mu g/m^3$ to total carbon, averaged over the entire year. Correcting this value for the fact that these fires only occur in summer results in a contribution of 1.04 $\mu g/m^3$. Adjusting for the fact that total carbon is approximately half of the PM2.5 mass emitted by fires could bring this value up to $\sim$2 $\mu g/m^3$, a value which is higher than the average of the five regions (1.11 $\mu g/m^3$) that we calculated. However, the analysis by Park et al. (*23*) only covers the years 2001–2004, a time period that had relatively high burning. Our analysis covers the period from 1988 to 2004. For the entire western United States, annual biomass consumed averaged 55% higher during 2001–2004 compared to the 1988–2000 time period. If instead, we use only the data from 2001 to 2004, our calculated PM across the five regions increases by 30% to 1.43 $\mu g/m^3$. In addition, our analysis only considers fires within the domain of our database (101°−125° W, 31°−49° N). PM from fires that are outside this region will not be included in our calculated values.

The summer enhancements in PM2.5 due to fires are a significant fraction of the annual National Ambient Air Quality Standards (15 $\mu g/m^3$) and contribute significantly to regional haze in the Western United States. During short periods, the impacts can be much greater and can be associated with significant health impacts. In addition, fires in the Western United States and Canada are likely to increase under the influence of global warming. For these reasons, it is important to further quantify and understand the role that fires play on air quality and haze in the Western United States. In a separate analysis, we have also considered how fires influence O_3 concentrations in the Western United States (*25*).

Acknowledgments

This report was sponsored by the Coordinating Research Council (CRC) under Project A-54. The data used were collected as part of the IMPROVE network. We acknowledge the efforts of the many individuals who made these observations.

Supporting Information Available

Tables of full names and locations of the IMPROVE sites used in this analysis, and information on fires and PM in each region (June, July, August only). This information is available free of charge via the Internet at http://pubs.acs.org.

Literature Cited

(1) Carrico, C. M.; Kreidenweis, S. M.; Malm, W. C.; Day, D. E.; Lee, T.; Carrillo, J.; McMeeking, G. R. Hygroscopic growth behavior of a carbon-dominated aerosol in Yosemite National Park. *Atmos. Environ.* **2005**, *39*, 1393–1404.

(2) McMeeking, G. R.; Kreidenweis, S. M.; Carrico, C. M.; Lee, T.; Collett, J. L.; Malm, W. C. Observations of smoke-influenced aerosol during the Yosemite Aerosol Characterization Study: Size distributions and chemical composition. *J. Geophys. Res.* **2005**, *110*, D09206.; doi:10.1029/2004JD005389.

(3) Westerling, A. L.; Cayan, D. R.; Brown, T. J.; Hall, B. L.; Riddle, L. G. Climate, Santa Ana winds and autumn wildfires in Southern California. *Eos, Trans., Am. Geophys. Union* **2004**, *85* (31), 289–300.; doi:10.1029/2004EO310001.

(4) Robinson, M. S.; Chavez, J.; Velazquez, S.; Jayanty, R. K. M. Chemical speciation of PM2.5 collected during prescribed fires of the Coconino National Forest near Flagstaff, Arizona. *J. Air Waste Manage. Assoc.* **2004**, *54*, 1112–1123.

(5) Ward, T. J.; Hamilton, R. F.; Smith, G. C. The Missoula, Montana PM2.5 speciation study-seasonal average concentrations. *Atmos. Environ.* **2004**, *38*, 6371–6379.

(6) Colarco, P. R.; Schoeberl, M. R.; Doddridge, B. G.; Marufu, L. T.; Torres, O.; Welton, E. J. Transport of smoke from Canadian forest fires to the surface near Washington, DC: Injection height, entrainment, and optical properties. *J. Geophys. Res.* **2004**, *109*, D06203.; doi:10.1029/2003JD004248.

(7) DeBell, L. J.; Talbot, R. W.; Dibb, J. E.; Munger, J. W.; Fischer, E. V.; Frolking, S. E. A major regional air pollution event in the northeastern United States caused by extensive forest fires in Quebec, Canada. *J. Geophys. Res.* **2004**, *109*(D19), doi:10.1029/2004JD004840.

(8) Wotawa, G.; Trainer, M. The influence of Canadian forest fires on pollutant concentrations in the United States. *Science* **2000**, *288*, 324–328.

(9) McKeen, S. A.; Wotawa, G.; Parrish, D. D.; Holloway, J. S.; Buhr, M. P.; Hubler, G.; Fehsenfeld, F. C.; Meagher, J. R. Ozone production from Canadian wildfires during June and July of 1995. *J. Geophys. Res.* **2002**, *107*(D14), doi:10.1029/2001JD000697.

(10) Jaffe, D.; Bertschi, I.; Jaeglé, L.; Novelli, P.; Reid, J. S.; Tanimoto, H.; Vingarzan, R.; Westphal, D. L. Long-range transport of Siberian biomass burning emissions and impact on surface ozone in western North America. *Geophys. Res. Lett.* **2004**, *31*, L16106.

(11) Bertschi, I. T.; Jaffe, D. A. Long-range transport of ozone, carbon monoxide and aerosols to the NE Pacific troposphere during the summer of 2003: Observations of smoke plumes from Asian boreal fires. *J. Geophys. Res.* **2005**, *110* (D5), D05303.; doi: 10.10292004JD005135.

(12) Pope, C. A.; Dockery, D. W. Health effects of fine particulate air pollution: Lines that connect. *J. Air Waste Manage. Assoc.* **2006**, *56*, 709–742.

(13) DeBell, L. J.; Gebhart, K. A.; Hand, J. L.; Malm, W. C.; Pitchford, M. L.; Schichtel, B. A.; White, W. H. Spatial and Seasonal Patterns and Temporal Variability of Haze and its Constituents in the United States, Report IV; Cooperative Institute for Research in the Atmosphere, Nov. 2006;ISSN 0737-5352-74.

(14) Westerling, A. L.; Hidalgo, H. G.; Cayan, D. R.; Swetnam, T. W. Warming and earlier spring increase Western U.S. forest fire activity. *Science,* **2006**, *313*, 940–943.

(15) Gillett, N. P.; Weaver, A. J.; Zwiers, F. W.; Flannigan, M. D. Detecting the effect of climate change on Canadian forest fires. *Geophys. Res. Lett.* **2004**, *31*, L18211.

(16) U.S. EPA. Part II, 40 CFR Part 51, Regional Haze Regulations; Final Rule. *Fed. Regist.* 1999, *64* 126; available at www.epa.gov/ttncaaa1/t1/fr_notices/rhfedreg.pdf.

(17) Ames, R.; Fox, D. G.; Malm, W. C.; Schichtel, B. A. Preliminary apportionments of carbonaceous aerosols to wild fire smoke using observations from the IMPROVE network;Paper #76, Conference on Regional Haze, AWMA, Asheville, NC, 2004.

(18) McDonald, J. D.; Zielinska, B.; Fujita, E. M.; Sagebiel, J. C.; Chow, J. C.; Watson, J. G. Fine particle and gaseous emission rates from residential wood combustion. *Environ. Sci. Technol.* **2000**, *34* (11), 2080–2091.

(19) Andreae, M. O.; Merlet, P. Emission of trace gases and aerosols from biomass burning. *Global Biogeochem. Cycle* **2001**, *15* (4), 955–966.

(20) Spracklen, D. V.; Logan, J. A.; Mickley, L. J.; Park, R. J.; Yevich, R.; Westerling, A. L.; Jaffe, D. Fires drive interannual variability of organic carbon aerosol in the western U.S. in summer. *Geophys. Res. Lett.* **2007**, *34*, L16816.; doi:10.1029/2007GL030037.

(21) Malm, W. D.; Sisler, J. F.; Huffman, D.; Eldred, R. A.; Cahill, T. A. Spatial and seasonal trends in particle concentration and optical extinction in the United States. *J. Geophys. Res.* **1994**, *99* (D1), 1347–1370.

(22) Park, R. J.; Jacob, D. J.; Chin, M.; Martin, R. V. Sources of carbonaceous aerosols over the United States and implications for natural visibility. *J. Geophys. Res.* **2003**, *108* (D12), 4355.

(23) Rokjin, J. P.; Jacob, D. J.; Logan, J. A. Fire and biofuel contributions to annual mean aerosol mass concentrations in the United States. *Atmos. Environ.* **2007**, *41* (35), 7389–7400.

(24) Malm, W. C.; Schichtel, B. A.; Pitchford, M. L.; Ashbaugh, L. L.; Eldred, R. A. Spatial and monthly trends in speciated fine particle concentration in the United States. *J. Geophys. Res.* **2004**, *109* (D3), D03306.

(25) Jaffe, D. A.; Hafner, W.; Chand, D.; Westerling, A.; Spracklen, D. V. Influence of Fires on O_3 concentrations in the Western U.S. Manuscript in preparation for submission to Environ. Sci. Technol.

(26) Westerling, A. L.; Gershunov, A.; Brown, T. J.; Cayan, D. R.; Dettinger, M. D. Climate and fire in the Western United States. *Am. Meteorol. Soc.* **2003**, 595–604.
(27) Randall, D. *Strawman-approach for technical refinement of large fires and fire. 197 complexes for WRAP Phase 1 fire emissions inventory*;Technical Memorandum, Project 198 No. 178-6; Air Sciences Inc.: Portland, OR, 2004.
(28) Malm, W. C.; Schichtel, B. A.; Ames, R. B.; Gebhart, K. A. A 10-year spatial and temporal trend of sulfate across the United States. *J. Geophys. Res.* **2002**, *107* (D22), 4627.
(29) Chow, J. C.; Watson, J. G.; Crow, D.; Lowenthal, D. H.; Merrifield, T. Comparison of IMPROVE and NIOSH carbon measurements. *Aerosol Sci. Technol.* **2001**, *34*, 23–34.
(30) Hirsch, R. M.; Gilroy, E. J. Methods of fitting a straight line to data: Examples in water resources. *Water Res. Bull.* **1984**, *20*, 705–711.
(31) Rinehart, L. R.; Fujita, E. M.; Chow, J. C.; Magliano, K.; Zielinska, B. Spatial distribution of PM2.5 associated organic compounds in central California. *Atmos. Environ.* **2006**, *40* (2), 290–303.
(32) Weiss-Penzias, P.; Jaffe, D.; Swartzendruber, P.; Hafner, W.; Chand, D.; Prestbo, E. Quantifying Asian and biomass burning sources of mercury using the Hg/CO ratio in pollution plumes observed at the Mount Bachelor Observatory. *Atmos. Environ.* **2007**, *41*, 4366–4379.

ES702755V