Supporting Information for

Contribution of Wildland-Fire Smoke to US $PM_{2.5}$ and Its Influence on Recent Trends

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The following document contains suplemental information for "Contribution of Wildland-Fire Smoke to US $PM_{2.5}$ and Its Influence on Recent Trends" by Katelyn O'Dell, Bonne Ford, Emily V. Fischer, and Jeffrey R. Pierce. The document is 7 pages in length and contains the following information: (1) Alternative statistical approach to calculate $PM_{2.5}$ slopes presented in Figures 1 and 2, (2) a description of the process used to determine the kriging parameters, (3) Figure 2 trends for all other seasons using the observation-based approach, and (4) a discussion of interannual variability in two driving factors of smoke $PM_{2.5}$ concentrations in the PNW: smoke concentration intensity and smoke day frequency. The four sections and associated figures are listed in the table of contents below.

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Alternative statistical approach

For the alternative statistical approach, we follow a similar methodology outlined in the main text with the exception of the linear-least squares regression and associated significance test. Here we estimate slopes using the Theil-Sen estimator[1]which calculates a linear-least squares regression between each possible pair of points in the dataset (here, each available year) and takes the median of the resulting slopes. We use Kendall's tau to estimate the statistical significance of the slopes. The Theil-Sen estimator and Kendall's tau have been used recently in air quality trend studies[2,3]as they are less sensitive to outliers. We apply this alternative statistical approach to the datasets shown in Figures 1 and 2 of the main text. Results are presented in Figures S1 and S2. The two methods result it similar slopes for each dataset across the domain.

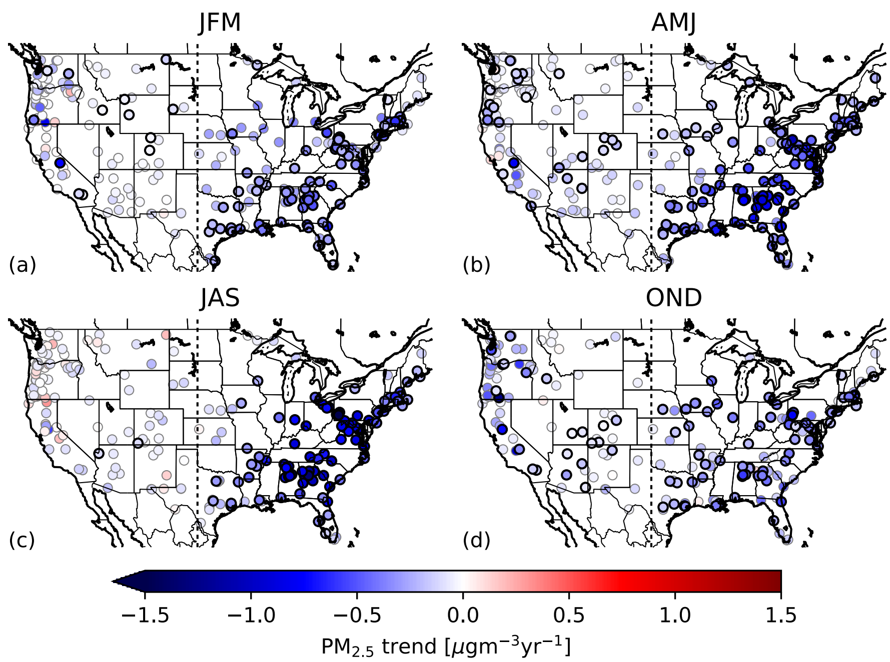


Figure S1. Thiel slopes of seasonal-mean $PM_{2.5}$ at EPA AQS sites from 2006-2016. Sites with significant correlations at the 95% confidence level according to Kendall's tau are outlined in black; sites with insignificant correlations are outlined in gray. Dashed line shown at 100° W.

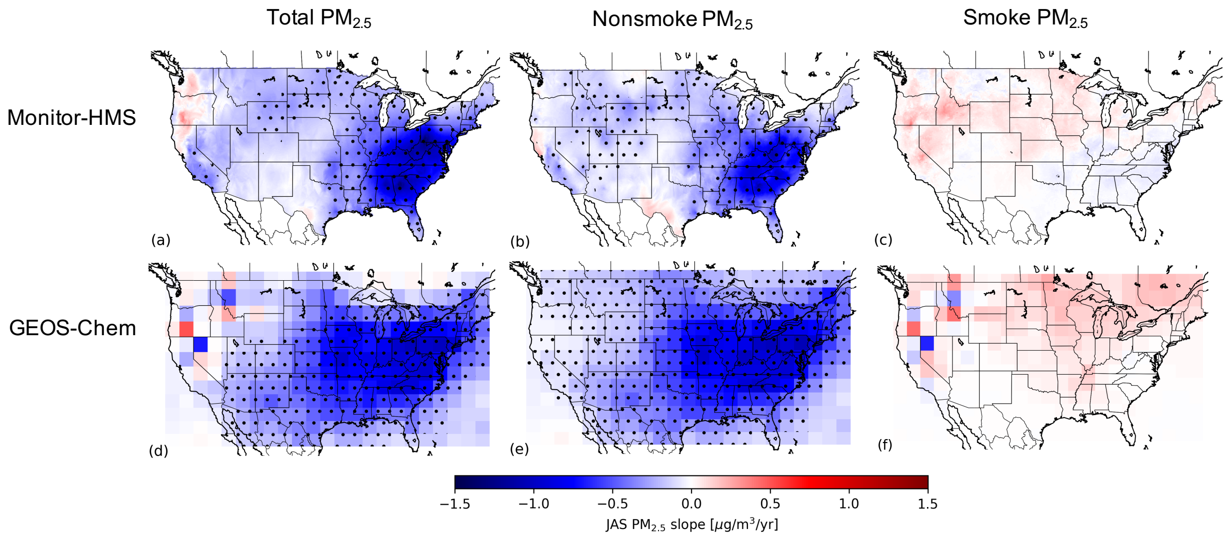
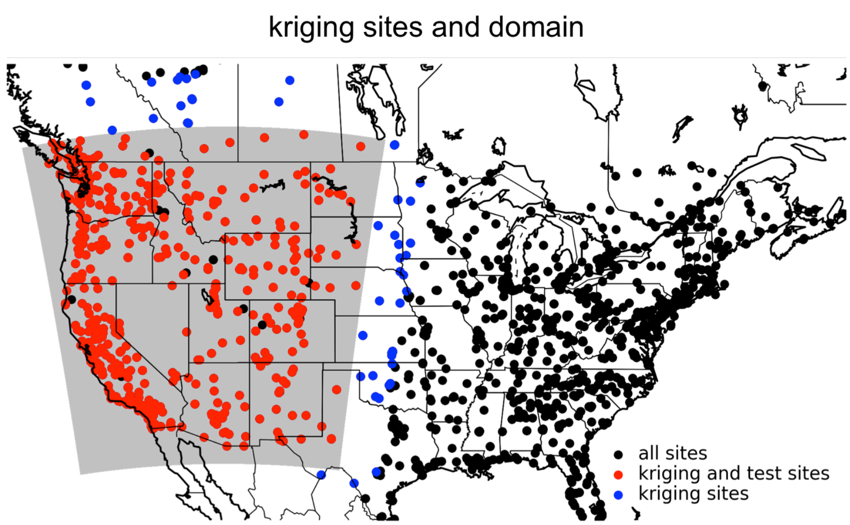


Figure S2. Thiel slopes of summer-mean (JAS) total $PM_{2.5}$, nonsmoke $PM_{2.5}$ (summer median for monitor-HMS method), and smoke $PM_{2.5}$ from 2006-2016 for the monitor-HMS (panels a, b, and c) and GEOS-Chem (panels d, e, and f) methods. Locations with significant correlations at the 95% confidence level according to Kendall's tau are dotted.

Determining kriging parameters

Kriging parameters (sill, range, and nugget) are determined using a k-fold cross validation with ten folds. 2,700 different combinations of parameters are tested. Values tested for each parameter are as follows: sill: 0.2, 0.4, 0.6, 0.8, 1.0, … 3.0; range: 0.5, 1.5, 2.0 … 10.0; nugget: 0.1, 0.2, 0.3 … 1.0. The parameters are evaluated over the western US with the sites shown in Figure S3 for May - October of 2015. For each set of parameters, the available monitoring sites were divided into ten unique groups (or 'folds') containing 101 monitors each (except the final group which contained 104 monitors). We remove one group of monitors and krige the remaining monitors to obtain a continuous estimate of $PM_{2.5}$ across the domain. We evaluate the krigged estimate against the $PM_{2.5}$ concentrations reported by the removed monitors for each day by calculating R^2, slope, mean bias, and mean absolute error. This process is repeated for each group of monitors. We then average the statistical parameters across the ten folds. We select the set of parameters we use in this study from the 15 sets of parameters that produce an R^2 in the highest 10%, slope in the highest 10%, mean bias in the lowest 10% absolute values, and mean absolute



error in the lowest 10%.

Figure S3. Kriging domain (shaded region) and sites used for selecting kriging parameters. Black points indicate locations of monitors available in the EPA AQS dataset. Blue points indicate locations of monitors used to constrain the edges of the kriging domain, but not used in the kriging evaluation. Red points indicate locations of monitors used in the interpolation and to evaluate the kriged surface. Note: black points within the kriging domain represent locations of monitors with no available data during the time period that we use to test the kriging parameters (May - October 2015).

Total, nonsmoke, and smoke $PM_{2.5}$ trends in winter, spring, and fall

Figure S4 shows the trends in total, nonsmoke, and smoke $PM_{2.5}$ for winter, spring, and fall seasons. Trends were calculated using the observation-based method described for the summer

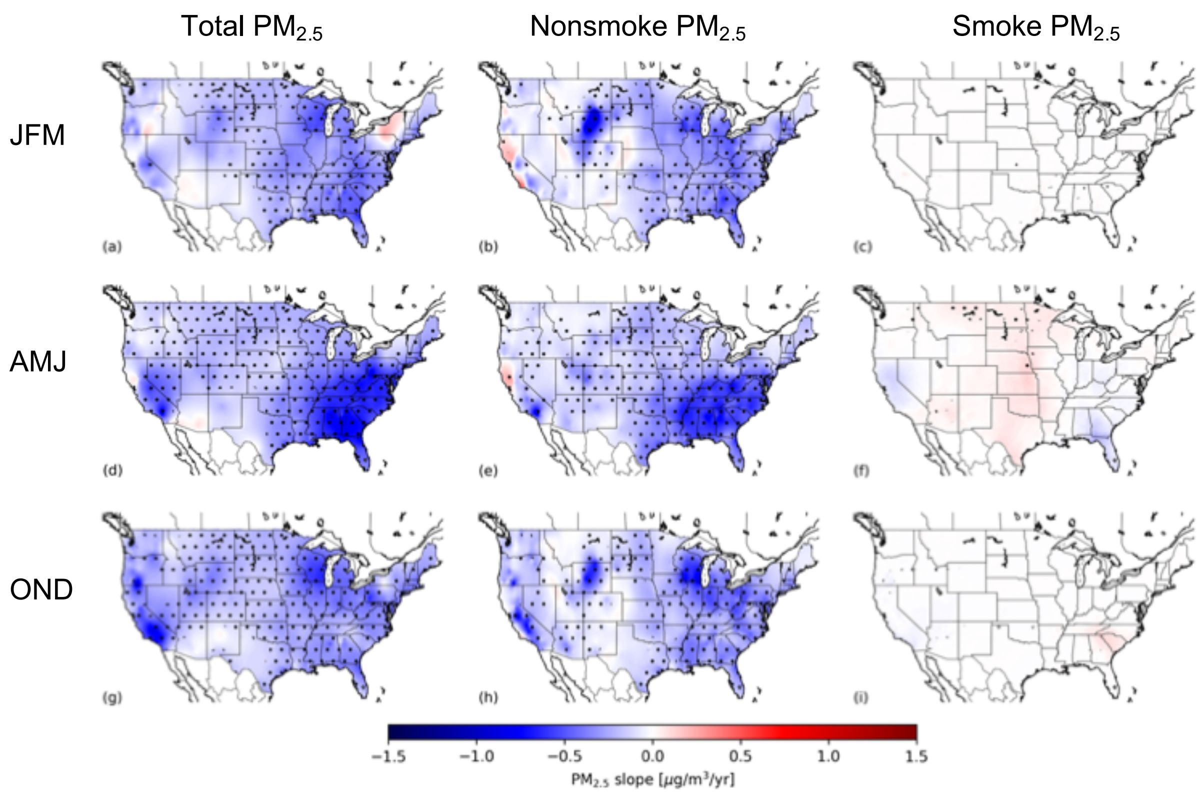


Figure S4. Trends in total, nonsmoke, and smoke $PM_{2.5}$ for winter (panels a, b, and c), spring (panels d, e, and f), and fall (panels g, h, and i).

season trends in the main text. Overall, the total and nonsmoke trends are decreasing in all seasons with a few exceptions in New York and northern California. Neither of these increases are reflected in the observations and thus could be an artifact of the kriging. In the case of the spring increases in California, the increasing trend could be due to separating out an apparent decreasing trend in smoke $PM_{2.5}$. The insignficant decreasing spring trend in smoke $PM_{2.5}$ in northern Califonia is likely due to the large fire season in June 2008 near the start of our study period. The spring trends in smoke $PM_{2.5}$ also show insignificant increases along the Mississippi River, where there is often a large amount of agricultural burning in spring. The overall smoke trend in both winter and fall is close to zero, due to the lack of smoke days in both seasons.

There are slight, insignificant increases in smoke $PM_{2.5}$ across South Carolina in the fall season, likely trend in both winter and fall is close to zero, due to the lack of smoke days in both seasons. For the fall and winter seasons with very few smoke days and near-zero trend in smoke $PM_{2.5}$, the difference between the nonsmoke and total $PM_{2.5}$ trends is likely not attributable to smoke and could arise from differences between the mean and median $PM_{2.5}$.

Interannual variability in drivers of smoke $PM_{2.5}$ concentations

In Figure S5, we are investigating two potential driving factors of interannual variability in smoke $PM_{2.5}$ in the Pacific Northwest (PNW) region defined by the grey boxes in Figure 2a of the main text. Interannual variability of smoke $PM_{2.5}$ in the PNW (orange bars in Figure S5a) may be driven by two factors:(1) the frequency of smoke days (purple bars in Figure S5b) and

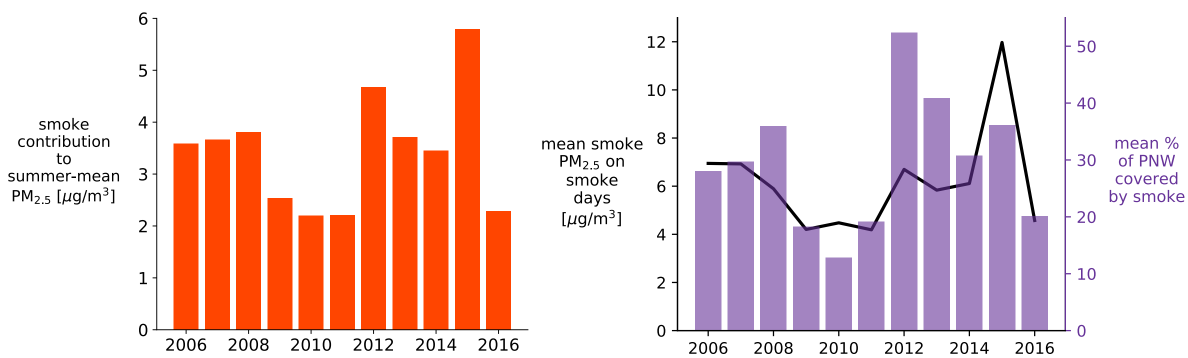


Figure S5. Panel (a): Smoke contribution to summer-mean $PM_{2.5}$ from the monitor-HMS method in the PNW (the orange portion of Figure 3a in the main text). Panel (b): Left axis: Black line shows the area-averaged summer (JAS) mean smoke $PM_{2.5}$ estimated using the monitor-HMS method on smoke-influenced days across the Pacific Northwest (PNW). Right axis: The average fraction of PNW area covered by a smoke plume during the summer is represented by the purple bars (i.e., the fraction of days that are smoke days).

(2) smoke $PM_{2.5}$ concentrations on smoke days (black line in Figure S5b); i.e., is smoke occurring more/less frequently, or are smoke concentrations higher/lower when they occur. The product of the smoke day frequency and smoke $PM_{2.5}$ concentrations on smoke days gives an estimate of summer-mean smoke $PM_{2.5}$. This value differs from the smoke $PM_{2.5}$ contribution shown in Figure S5a due to our method of defining nonsmoke $PM_{2.5}$ as a seasonal median rather than mean on nonsmoke days.

The summer-mean fraction of total area in the PNW with smoke somewhere in the column (i.e., the summer mean smoke-day frequency) ranges from 10 - 15% in low fire years (e.g. 2010) to >50% in extreme years (e.g. 2012). The summer-mean smoke $PM_{2.5}$ concentrations on smoke days range from ~2 μg m^{-3} in low fire years to ~6 μg m^{-3} in high fire years according to the monitor-HMS method. There is a moderate correlation between the two cases (R^2 = 0.36); big fire years tend to have higher values for both metrics. However, some years with high smoke contributions to the summer-mean total $PM_{2.5}$ (Figure S5a) are more driven by extensive smoke spatial/temporal coverage (e.g. 2012) while others are more driven by higher smoke $PM_{2.5}$ concentrations on smoke days (e.g. 2015). Overall, 2012 and 2015 are the two years with the highest smoke contributions to the summer-mean total $PM_{2.5}$ (Figure S5a), yet we estimate that they achieved this for different reasons. It is unclear from our estimates alone what is driving the independent variability in smoke area/temporal coverage versus smoke $PM_{2.5}$ concentrations. Large interannual variability in both the area/frequency of smoke days and smoke $PM_{2.5}$ on smoke days obscures decreasing trends in nonsmoke summer $PM_{2.5}$ in this region (Figure 3 in the main text and Figure S5 in the supplement).

References

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(3) Malm, W. C.; Schichtel, B. A.; Hand, J. L.; Collett, J. L. Concurrent Temporal and Spatial Trends in Sulfate and Organic Mass Concentrations Measured in the IMPROVE Monitoring Program. *J. Geophys. Res. Atmospheres* 2017, *122* (19), 2017JD026865. https://doi.org/10.1002/2017JD026865.