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# Understanding Reductions of $PM_{2.5}$ Concentration and Its Chemical Composition in the United States: Implications for Mitigation Strategies

Chi Li,* Randall V. Martin, and Aaron van Donkelaar

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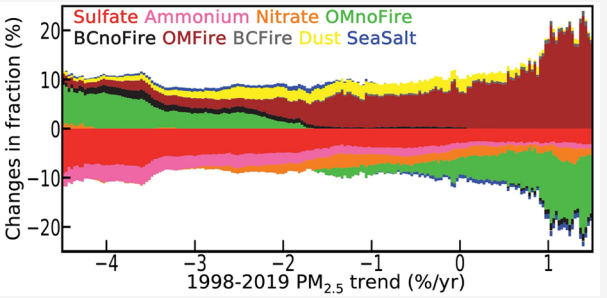
**ABSTRACT:** Motivated by the recent tightening of the US annual standard of fine particulate matter ($PM_{2.5}$) concentrations from 12 to 9 $\mu g/m^3$, there is a need to understand the spatial variation and drivers of historical $PM_{2.5}$ reductions. We evaluate and interpret the variability of $PM_{2.5}$ reductions across the contiguous US using high-resolution estimates of $PM_{2.5}$ and its chemical composition over 1998−2019, inferred from satellite observations, air quality modeling, and ground-based measurements. We separated the 3092 counties into four characteristic regions sorted by $PM_{2.5}$ trends. Region 1 (primarily Central Atlantic states, 25.9% population) exhibits the strongest population-weighted annual $PM_{2.5}$ reduction (−3.6 ± 0.4%/yr) versus Region 2 (primarily rest of the eastern US, −3.0 ± 0.3%/yr, 39.7% population), Region 3 (primarily western Midwest, −1.9 ± 0.3%/yr, 25.6% population), and Region 4 (primarily the Mountain West, −0.4 ± 0.5%/yr, 8.9% population). Decomposition of these changes by chemical composition elucidates that sulfate exhibits the fastest reductions among all components in 2720 counties (76% of population), mostly over Regions 1−3, with the 1998−2019 mean sulfate mass fraction in $PM_{2.5}$ decreasing from Region 1 (29.5%) to Region 4 (11.8%). Complete elimination of the remaining sulfate may be insufficient to meet the new standard for many regions in exceedance. Additional measures are needed to reduce other $PM_{2.5}$ sources and components for further progress.




**KEYWORDS:** *Air quality, Particulate matter, Population exposure, Chemical composition, Environmental regulation*

## INTRODUCTION

Fine particulate matter ($PM_{2.5}$) air pollution has adverse impacts on human health even at low levels[1,2] and is at present the second largest environmental risk factor to human health across the United States (US).[3,4] The Clean Air Act and subsequent amendments have led to sustained reductions of anthropogenic $PM_{2.5}$ sources in the US for over three decades.[5−8] The responses of total $PM_{2.5}$ are collectively determined by $PM_{2.5}$ chemical composition changes that are driven by changes in specific sources.[7−10] As the US Environmental Protection Agency (EPA) recently reduced the annual $PM_{2.5}$ standard from 12 to 9 $\mu g/m^3$, it is important to reflect upon prior reductions in $PM_{2.5}$ exposure to inform future mitigation strategies. Long-term dedicated monitoring of $PM_{2.5}$ chemical composition[11] has been essential to attribute reduced anthropogenic emissions as the main driver of recent $PM_{2.5}$ reductions across the US.[8−10,12−15] Characteristic spatial variation of these improvements, e.g., stronger reductions in $PM_{2.5}$ over the eastern than the western US,[16] has also been revealed. These existing studies primarily focused on absolute trends at regional and national scales that are not independent of local pollution level; there is need to interpret relative trends in population-weighted (PW) $PM_{2.5}$ concentrations and its chemical composition at fine spatial scales (e.g., county-level) across the contiguous US (CONUS) to better understand how changes in chemical composition affected population exposure.

Recent advances in satellite remote sensing and air quality modeling offer information to fill spatial gaps[17] of in situ monitoring and enable accurate estimation of high-resolution gapless $PM_{2.5}$ and chemical composition across the CONUS.[18−20] Such data enable investigation and interpretation of PW-$PM_{2.5}$ trends at the county-level. In this paper, we use timely and accurate estimates of $PM_{2.5}$ and chemical composition to quantify and interpret PW-$PM_{2.5}$ trends for 3092 counties across the CONUS during 1998−2019, focusing on relative $PM_{2.5}$ trends (i.e., $\Delta PM_{2.5}$ in %/yr) that are normalized to mass concentrations and independent of local pollution level. We find strong regional variation of PW-$PM_{2.5}$ reduction rates with an overall meridional gradient (i.e., weakening from the east to the west). Similar regional

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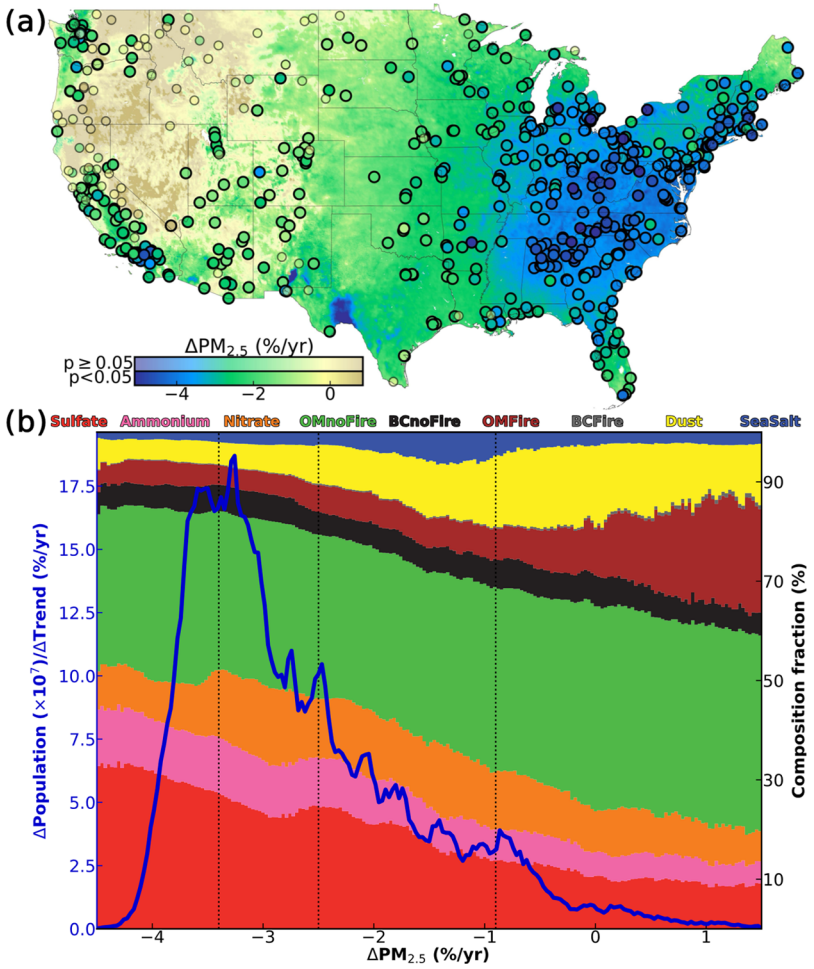


**Figure 1.** Strong regional variation of $PM_{2.5}$ relative trends ($\Delta PM_{2.5}$) across the CONUS. (a) Relative areal trends (%/yr) in annual mean $PM_{2.5}$ during 1998−2019 from the satellite-derived estimates (background) and in situ measurements (points). Insignificant trends with $p \geq 0.05$ are displayed with more transparent colors and smaller symbols. (b) Population (bin-width normalized distribution, blue line, left *Y*-axis) and 1998−2019 mean population-weighted (PW) $PM_{2.5}$ chemical composition (right *Y*-axis, color-coded on the top) as a function of these trends. All components are presented at 35% relative humidity (RH) for consistency with $PM_{2.5}$ mass concentration measurements. OM and BC are separated into contributions from open burning (Fire) and other sources (noFire). Vertical dotted lines are the thresholds to separate the four regions (Figure S4).

distributions are identified for the $PM_{2.5}$ mass fraction of one chemical component—sulfate, which also exhibited the strongest reductions among all components in 2720 counties (76% of the CONUS population). We conclude that policies that reduced sulfur dioxide emissions have been the most responsible for these PW-$PM_{2.5}$ reductions and their regional variation. We find that even complete elimination of sulfate would be insufficient to meet the new EPA standard; other measures are needed to sustain such progress in the future.

## MATERIALS AND METHODS

**$PM_{2.5}$ Data.** We use monthly estimates of $PM_{2.5}$ mass concentration and its seven major chemical components (sulfate, ammonium, nitrate, organic mass (OM), black carbon (BC), dust, and sea salt) across the CONUS based on a recent study.[18] The original data (2000−2016) are revised using more recent satellite retrievals of aerosol optical depth (AOD) and updated long-term simulations using the GEOS-Chem model of atmospheric composition in its high-performance configuration[21,22] (Text S1). We use the "hybrid" $PM_{2.5}$ and chemical composition during 1998−2019 that have merged information from satellite retrievals, modeling, and ground-based observations, achieving a high degree of consistency with observations (Text S1). To aid interpretation, we use the simulated contribution from open burning to further apportion the estimated OM and BC into concentrations from fire and other sources. Here we use the terms "OM" and "BC" to indicate their total mass concentrations, unless specifically referring to their fire or non-fire components. To focus on evaluation of long-term air quality regulations, we do not use years after 2019 to avoid impacts from COVID-19 lockdown[23,24] and extreme wildfire events in the western US in 2020 and 2021.[25−27]

**Ground-Based Measurements.** We use long-term monitoring data of $PM_{2.5}$ and its chemical composition to develop, evaluate, and interpret trends from the gapless estimates. Description of these observations is provided in Text S2. We require at least 10 (3) months of data available for an annual (seasonal) mean to be calculated, and at least 17 (out of 22) years available for a trend during 1998−2019 to be estimated (i.e., "long-term" criteria). These completeness criteria yield >660 $PM_{2.5}$ sites and >190 sites for chemical composition (e.g., Figures S1 and S2).


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**Table 1. Correlation Coefficients (r) between the 1998−2019 Mean Mass Fraction of Major $PM_{2.5}$ Chemical Compositions (Separated by Rows) and Relative $PM_{2.5}$ Trends at Pixel (Column 2) and County (Column 3, population-weighted) Level, from in Situ Observational Sites (Column 4), and from Co-located Estimates at Observational Sites (Column 5), across the CONUS[a]**

| Component | Correlation (1 km$^2$) | Correlation (county) | Correlation (in situ) | Correlation (co-located) |
|---|---|---|---|---|
| Sulfate | **−0.78** | **−0.79** | **−0.56** | **−0.62** |
| Ammonium | **−0.44** | **−0.38** | **−0.48** | **−0.49** |
| Nitrate | 0.08 | 0.25 | −0.07 (0.4) | −0.03 (0.7) |
| OM | 0.29 | 0.21 | 0.36 (0.001) | 0.34 (0.007) |

[a]Correlations with $p < 0.001$ are bold and otherwise are followed by the $p$-values in the brackets. For in situ observations, the nearest $PM_{2.5}$ measurement (within 20 km) is used for each component observation to derive the mass fraction. Only locations with significant ($p < 0.05$) $PM_{2.5}$ trends are investigated.

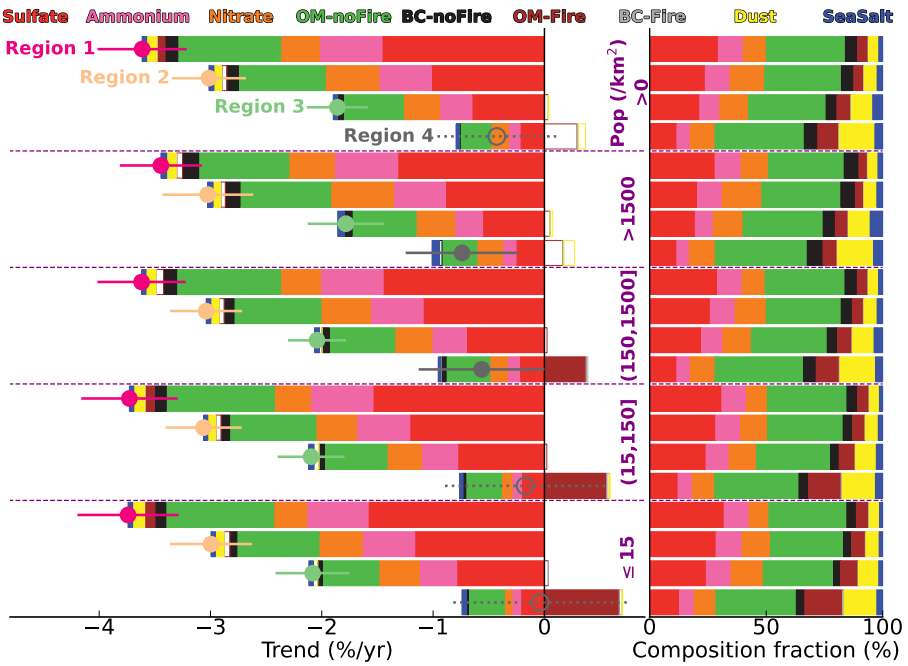


**Figure 2.** Sulfate dominates the $PM_{2.5}$ reductions and its regional variation. On the left, circles and error bars (color-coded for the four regions) represent relative trends and 95% confidence intervals in annual mean regional PW $PM_{2.5}$ (35% RH) over 1998−2019. Pixels in each region are further divided into four population bins (separated by purple lines and texts) following the overall PW-$PM_{2.5}$ cases (the top four rows). Stacked bars indicate compositional trends (consistently normalized to multi-year mean PW $PM_{2.5}$) to represent their contributions. Filled circles and bars are significant trends ($p < 0.05$) and empty ones are not. On the right, stacked bars represent compositional contribution to the multi-year mean PW $PM_{2.5}$ for each case. OM and BC are separated into contributions from open burning (Fire) and other sources (noFire). All components are presented at 35% RH for consistency with $PM_{2.5}$ mass concentration measurements.

**Population Data.** We use population estimates at 1 km$^2$ resolution from the Gridded Population of the World (GPW v4) database.[28] GPW is available every five years for 2000−2020. For each year during 1998−2019, we scale the GPW population distribution in the closest year by a constant factor to match the annual CONUS total population.[29]

**Trend Analysis.** We estimate 1998−2019 linear trends in the time series of PW-$PM_{2.5}$ and chemical composition at county and regional levels based on a linear least-squares fitting approach. We investigate the linear slope (in $\mu g/m^3/yr$), its 95% confidence interval (CI), as well as the $p$-value (two-tailed student's t-test) at seasonal and annual scales. These multi-year slopes are less sensitive to abnormal years (i.e., versus differences between the beginning and ending years). We confirmed that the derived trends at county-level are highly consistent ($R^2 > 0.9$) with those from a non-parametric (Mann−Kendall) trend estimation approach.[30,31] The linear slopes of $PM_{2.5}$ are then normalized by multi-year mean $PM_{2.5}$ to represent relative $PM_{2.5}$ trends ($\Delta PM_{2.5}$) at each location. We tested this normalization for sensitivity to the choice of mean or median $PM_{2.5}$ and found negligible changes in population-weighted monthly concentrations. We find that the sum of PW trends from the seven components is highly consistent with the trends in total PW-$PM_{2.5}$ (typically within 5%) despite independent linear regressions, a feature that facilitates evaluation of contributions from each component to the derived relative $PM_{2.5}$ trends. For in situ measurements, we conduct similar evaluations of component contributions by comparing composition trends vs $PM_{2.5}$ trends from the nearest site (within 20 km). Not all sites provide simultaneous measurements of all seven components; e.g., the number of long-term sites with OM measurements are significantly fewer than sites with sulfate measurements (Figure S1). We therefore only perform the comparison among component trends over sites that have maintained long-term measurements for at least 4 main components (sulfate, ammonium, nitrate, and OM, contributing to >69% of annual $PM_{2.5}$ mass for 95% pixels of the CONUS).


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## RESULTS

**Spatially Varying $PM_{2.5}$ Reduction Rates across the CONUS.** Figure 1a shows the relative $PM_{2.5}$ trends (%/yr) from the estimates (background) and in situ observations (points). We find that 95% of the CONUS population experienced significantly ($p < 0.05$) decreasing $PM_{2.5}$ concentrations, while $PM_{2.5}$ concentrations across the Mountain West exhibit insignificant or slightly positive trends. Except for several discrete locations (e.g., Greater Los Angeles, GLA), pixel-level relative $PM_{2.5}$ trends exhibit an overall meridional gradient, increasing from near zero over the Mountain West to the strongest (<−3%/yr) reductions over the Central Atlantic states. This spatial distribution of relative $PM_{2.5}$ trends from the hybrid estimates is highly consistent ($R^2 = 0.71$) vs. observations. Absolute trends (in $\mu g/m^3/yr$) are also stronger over the eastern US plus GLA (e.g., Figure S1) than over most of the west, as is already well established in previous studies.[16,32−34] However, if there were no strong spatial differences in historical $PM_{2.5}$ mitigation effectiveness, relative $PM_{2.5}$ trends should be similar across the CONUS, while the significant regional variability of relative $PM_{2.5}$ trends in Figure 1a further illustrates that these absolute trends are not proportional to, and thus cannot be fully explained by, the spatially varying baseline pollution level (e.g., Figure S2).

As $PM_{2.5}$ chemical composition is insightful about responses of $PM_{2.5}$ to sources and has been important to interpret $PM_{2.5}$ trends,[9,10,19] we investigate how $PM_{2.5}$ speciation varies with relative $PM_{2.5}$ trends ($\Delta PM_{2.5}$) to unveil possible drivers of this regional variation. We sort the CONUS pixels by local relative $PM_{2.5}$ trends. Figure 1b (background) shows the 1998−2019 mean mass fraction for each $PM_{2.5}$ chemical component for 200 $\Delta PM_{2.5}$ bins. It is evident that the locations with stronger negative trends coincide with stronger sulfate mass fraction in $PM_{2.5}$ (red), which decreases from 33% to 9% as the relative $PM_{2.5}$ trends weaken in magnitude. This decrease is nearly monotonic across pixels with negative $PM_{2.5}$ trends, except for a transient reversal regime at $-3.4\%/yr < \Delta PM_{2.5} < -2.5\%/yr$ (dotted lines) where an enhanced nitrate (orange) contribution slightly disrupts this association of sulfate fraction versus relative $PM_{2.5}$ trends. Over the entire figure, the fraction of fire-associated OM (brown) plus dust (yellow) increases from 9% to 36%, as the relative $PM_{2.5}$ reduction weakens, and the fraction of the CONUS population (blue line) diminishes. Sulfate in the CONUS is dominantly formed by sulfur dioxide ($SO_2$) emissions from coal combustion in power plants,[12,35,36] which has been substantially reduced for five decades as a primary target of regulatory policies;[37−39] meanwhile fire-OM and dust are overall less readily regulatable due to their primarily natural origins and sensitivity to climate change.[32,40,41] The regional variation of relative $PM_{2.5}$ trends directly relates to such varying chemical composition and how effectively specific components have been reduced.

Table 1 summarizes the correlation coefficients between relative $PM_{2.5}$ trends and the long-term mass fraction of the main $PM_{2.5}$ components. Trends in the remaining components (BC, dust, and sea salt) do not make significant contributions to the total $PM_{2.5}$ trends (e.g., Figure 2). Sulfate mass fraction exhibits the strongest anticorrelation (e.g., $r = -0.78$ at pixel level) with relative $PM_{2.5}$ trends based on both the estimates and long-term in situ observations among all components, further indicating the driving role of $PM_{2.5}$ speciation in the regionally varying reduction rates. Figure S1 shows that sulfate exhibits CONUS-wide reductions that are the strongest among all components, supported by both the estimates and in situ observations (i.e., $R^2 > 0.45$ for co-located trends). CONUS-wide, the PW-$PM_{2.5}$ reduction (±95% confidence interval) of $-0.30 \pm 0.03$ $\mu g/m^3/yr$ is dominantly from PW decreases of sulfate ($-0.11 \pm 0.01$ $\mu g/m^3/yr$), OM ($-0.08 \pm 0.01$ $\mu g/m^3/yr$), ammonium ($-0.05 \pm 0.01$ $\mu g/m^3/yr$), and nitrate ($-0.04 \pm 0.01$ $\mu g/m^3/yr$).

Figure S3 shows the binned PW speciated component fraction (similar to Figure 1b) as a function of relative $PM_{2.5}$ trends ($\Delta PM_{2.5}$) for the start (a) and end (b) of the investigated period (1998−2019), as well as their corresponding changes (c). From 1998−2000 to 2017−2019, sulfate exhibits negative changes (by up to −9%) in its fraction in PW-$PM_{2.5}$ across all $\Delta PM_{2.5}$ levels, and stronger magnitudes of this fractional reduction are associated with more negative relative $PM_{2.5}$ trends. This reduced sulfate fraction is largely compensated by the OM fraction, the other main component across the CONUS with relatively slower reduction. The increase in OM fraction (by 3−9% across all bins) in PW-$PM_{2.5}$ is dominated by non-fire sources (by up to 8%) for pixels with relative $PM_{2.5}$ reductions stronger than −2%/yr, and by fire sources (by up to 23%) for locations with slower (e.g., >−1%/yr) relative reduction rates. Overall, Figure S3 implicates substantial changes in PW-$PM_{2.5}$ chemical composition (i.e., Figure S3a vs S3b) during 1998−2019 due to divergent changes in each component, with sulfate and OM exhibiting the strongest changes in their mass fractions. Consequently, the CONUS-wide PW sulfate/OM ratio of 0.70 in 1998 decreased to 0.47 in 2019.

The above evidence supports the conclusion that the effectiveness of sulfate regulation contributes the most to this US-wide regional diversity in relative $PM_{2.5}$ trends, followed by OM. Besides this overall CONUS pattern, regional component contributions to $PM_{2.5}$ and its trends warrants additional investigation to further understand and interpret such diversity of $PM_{2.5}$ reduction rates.

**Regionally Varying Compositional Drivers of $PM_{2.5}$ Trends.** We further divide the CONUS counties into four characteristic regions (i.e., dotted lines in Figure 1b), with limits chosen based on the speciation of $PM_{2.5}$ as a function of relative $PM_{2.5}$ trends ($\Delta PM_{2.5}$). Region 1 ($\Delta PM_{2.5} < -3.4\%/yr$ & and $p < 0.05$) has the strongest relative $PM_{2.5}$ trends and 25.9% of the CONUS population, primarily located over the Central Atlantic states (Figure S4). Region 2 ($-3.4 \le \Delta PM_{2.5} < -2.5\%/yr$ and $p < 0.05$) hosts the largest CONUS population (39.7%), including the GLA and most of the eastern US apart from Region 1. Region 3 ($-2.5 \le \Delta PM_{2.5} < -0.9\%/yr$ and $p < 0.05$) broadly spans the western Midwest and parts of the Southwest, and includes 25.6% of the CONUS population. Region 4 ($\Delta PM_{2.5} \ge -0.9\%/yr$ or $p \ge 0.05$) includes the Mountain West (8.9% population) that is usually prone to sources of fire or dust. Using the above thresholds (i.e., dotted lines in Figure 1b), the division of four regions is almost identical as determined based on county- or pixel-level (Figure S4), again reflecting the strong regional rather than local division of variability in relative $PM_{2.5}$ trends. We use the county-level definitions throughout the discussion, which maintain better spatial separation for each region, and closer relevance with regulatory policies.

Figure 2 (top 4 rows) summarizes component contributions to annual mean PW-$PM_{2.5}$ and its trends for each region (Figure S5 shows more detailed results at the county level).


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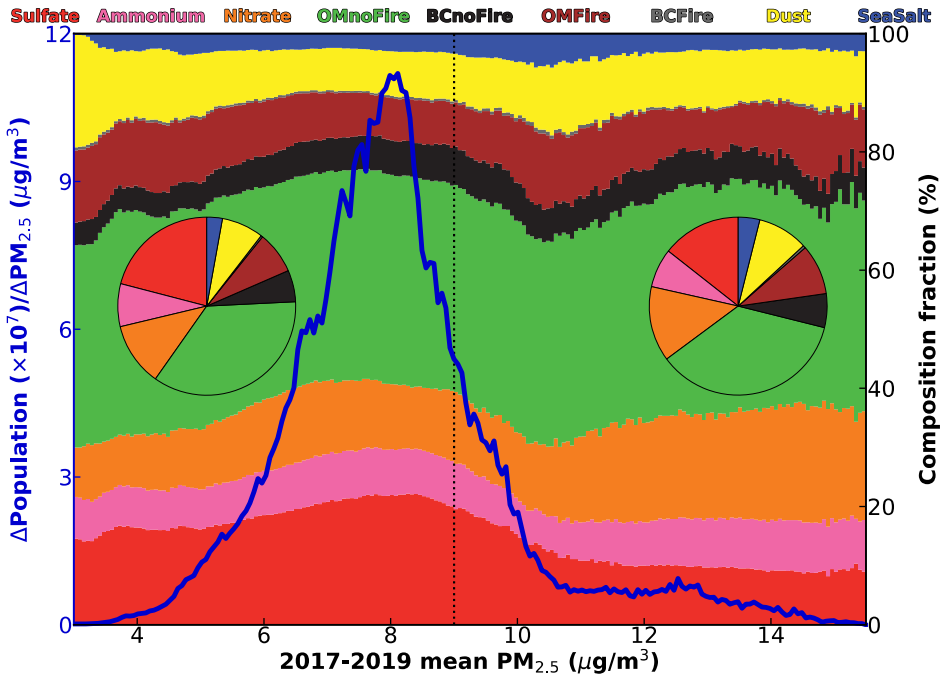


**Figure 3.** Future $PM_{2.5}$ mitigation over the CONUS will require stronger measures to reduce components in addition to sulfate. Population (bin-width normalized distribution, blue line, left *Y*-axis) and 2017−2019 mean population-weighted (PW) $PM_{2.5}$ chemical composition (right *Y*-axis, color-coded on the top) as a function of 2017−2019 mean $PM_{2.5}$ concentrations. The vertical dotted line represents the new EPA standard (9 $\mu g/m^3$). The two pie charts show the compositional fraction of PW-$PM_{2.5}$ for pixels attaining (left) and violating (right) this standard based on the 2017−2019 mean concentrations. All components are presented at 35% RH for consistency with $PM_{2.5}$ mass concentration measurements.

PW-$PM_{2.5}$ over Regions 1−4 exhibit variable relative $PM_{2.5}$ trends of −3.6 ± 0.4%/yr, −3.0 ± 0.3%/yr, −1.9 ± 0.3%/yr, and −0.4 ± 0.5%/yr (statistically insignificant), respectively, with the 1998−2019 regional mean sulfate mass fraction decreasing from 29.5% in Region 1 to 11.8% in Region 4. Regional mean PW-sulfate roughly accounts for 40%, 34%, and 35% of relative $PM_{2.5}$ trends from Region 1 to Region 3, respectively (see also Figures S5 and S6), stronger than contributions from the other $PM_{2.5}$ components. OM is the second largest contributor to PW-$PM_{2.5}$ reductions in Regions 1−3 (27−28%, Figure S6), where the OM reductions are negligibly affected by fire (Figure 2). In contrast, the sulfate mass fraction and its reduction (−0.02 $\mu g/m^3/yr$) are less dominant in Region 4, where the decreasing components are counteracted by (statistically insignificant) increases in OM from fire (0.02 $\mu g/m^3/yr$) and dust (0.001 $\mu g/m^3/yr$) to yield an insignificant PW-$PM_{2.5}$ trend. At the county scale (Figure S5), the association of more sulfate contribution with higher PW-$PM_{2.5}$ reduction remains strong (e.g., Table 1), and sulfate leads with the fastest absolute reductions among all components in 2720 counties (76% population), which are located mostly over Regions 1−3 (Figure S7a, background). This dominance of sulfate as the most effectively mitigated component is also consistently indicated by the in situ observations (Figure S7a, dots). OM exhibits the second strongest reductions in most of the CONUS which can surpass sulfate decreases at certain locations, as also consistently indicated by both the estimates and ground-based measurements (Figure S7b).

Figure 2(other rows) further divides pixels in each region into four bins of population density (Figure S8). Variation of relative PW-$PM_{2.5}$ trends and their component contributions is consistently pronounced across regions, while differences between population bins within the same region are relatively smaller. This invariability among population bins is consistent with the sulfate dominance of these trends, since sulfate is a regional pollutant that can be secondarily formed at >1000 km downwind of $SO_2$ sources[42] and exhibits the weakest urban/rural differences among $PM_{2.5}$ components.[43] Only Region 4 exhibits a weakly systematic relation of relative $PM_{2.5}$ trends with population bins, with reduced impacts from fire OM and dust as well as stronger reductions in PW-$PM_{2.5}$ with increasing population density. Only in the relatively more populated places (e.g., >150/$km^2$) is PW-$PM_{2.5}$ significantly reduced in Region 4.

Figure S9 investigates the component contributions to the PW-$PM_{2.5}$ mass and relative $PM_{2.5}$ trends in winter (DJF) and summer (JJA). In winter, OM reductions become stronger than sulfate in all regions, reflecting both effective policies of reducing residential emissions of primary OM[44] and reduced photochemical formation of sulfate.[45] Nitrate also exhibits more pronounced reductions in winter due to the enhanced formation,[45,46] especially over Regions 2−4 where nitrate reductions compete with or in some cases surpass the sulfate reductions. In Regions 3 and 4, OM from fire and dust have enhanced impacts on $PM_{2.5}$ mitigation in summer, leading to weaker relative $PM_{2.5}$ trends than in winter. Meanwhile in the more populous Regions 1 and 2, PW-$PM_{2.5}$ reduction rates are stronger in summer than winter, consistent with the seasonality of stronger sulfate contribution in summer, and with the overall leading role of sulfate reductions in driving the relative $PM_{2.5}$ trends.

**Importance of Reducing Other Components to Meet the New EPA Standard.** As sulfate has already been substantially reduced, measures to reduce other components (e.g., OM and nitrate) will become increasingly important for further mitigation of $PM_{2.5}$ air pollution to meet a stricter annual mean standard. Figure 3 illustrates this argument with a


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focus on the new EPA standard (9 $\mu g/m^3$). Based on contemporary (2017−2019) $PM_{2.5}$ concentrations, 21.9% of the CONUS population lives where annual mean $PM_{2.5}$ concentrations exceed 9 $\mu g/m^3$, with low sulfate mass fractions especially at high PW-$PM_{2.5}$. The locations exceeding the new standard are mainly urban centers and fire-prone regions (Figure S10a). The PW-$PM_{2.5}$ and PW-sulfate for these regions are 10.5 and 1.5 $\mu g/m^3$ (Figure 3, pie chart on the right), respectively, indicating that even eliminating CONUS-wide anthropogenic sulfate sources (with interannual variability and remaining background sulfate) may be insufficient to meet the standard in many of these regions. In contrast, PW-OM (4.7 $\mu g/m^3$, 1.0 $\mu g/m^3$ from open burning for $PM_{2.5}$ > 9 $\mu g/m^3$) is more dominant over these nonattainment regions. Figure S10b indicates that reducing OM is especially critical over the fire-prone states of California, Arizona, Oregon, Washington, Idaho, Montana, and several metropolitan areas in the southeastern US (e.g., Atlanta). Furthermore, PW-nitrate (1.4 $\mu g/m^3$ for $PM_{2.5}$>9 $\mu g/m^3$ and 1.9 $\mu g/m^3$ for $PM_{2.5}$>10 $\mu g/m^3$) has greater contribution to PW-$PM_{2.5}$ at higher $PM_{2.5}$ levels (Figure 3), with particular relevance to achieve the new guideline over central and southern California, as well as cities near the Great Lakes (e.g., Chicago, Detroit, and Pittsburgh). Achieving the new standard will depend on enhanced measures to reduce these components in the future.

## DISCUSSION

Motivated by the tightening of the annual $PM_{2.5}$ standard by the US EPA, we report a timely examination of the relative and absolute reductions of population exposure to $PM_{2.5}$ and its chemical composition over the CONUS over the past two decades. We employed high-resolution estimates developed from a combination of satellite remote sensing, modeling of atmospheric composition, and in situ measurements, with complete coverage across the CONUS that enabled examination of population-weighted quantities. Interpretation of these high-resolution estimates together with in situ observations reveals that regional reductions have a clear connection with sulfate contributions to $PM_{2.5}$, as sulfate has been the most successfully regulated $PM_{2.5}$ component during 1998−2019. Locations more dominated by sulfate thus experienced stronger relative $PM_{2.5}$ trends, an almost uniform CONUS-wide feature during the past two decades (e.g., Figures 1b and S4). This historical benefit from reduced sulfate becomes more prominent if jointly considering its partial contribution to mitigating OM during warm seasons by modulating secondary organic aerosol formation yields from biogenic volatile organic compounds (VOC) emissions.[47,48]

Our findings also highlight the necessity to improve the understanding and regulation of emissions and formation of the other $PM_{2.5}$ components, especially OM and nitrate. Even complete elimination of sulfate may be insufficient for many regions to achieve the new annual mean $PM_{2.5}$ standard of 9 $\mu g/m^3$ (Figure 3). We find that OM has the second strongest reduction (Figures S6 and S7) broadly over the CONUS and is the driving component of $PM_{2.5}$ reduction rates over certain locations (Figure S7a) and seasons (Figure S9a), with increasing dominance of PW-$PM_{2.5}$ in recent years (Figure S3). The latter finding implies an increasing urgency to sustain and strengthen previous OM reductions. Regulation of anthropogenic emissions of primary OM has been attributed to 2/3 of these reductions.[44] However, VOCs from volatile chemical products (VCP) have recently become more pervasive than vehicle VOCs and the dominant source of secondary OM in the urban US.[49,50] Moreover, fire over the CONUS (especially in the west) has been and is predicted to become stronger, more frequent, and spatially broader,[51−53] as reflected by its uniformly increasing contribution to PW-$PM_{2.5}$ (Figure S3c). Strategies are needed to deal with these emerging OM sources.[54−57] Locally, mitigation of nitrate, which comprises 13.3% of PW-$PM_{2.5}$ in locations violating the new standard, has been critical to the successful air quality improvement over Region 2 (e.g., GLA and the Great Lakes, Figures 1b, S2, and S10c). Sustaining nitrate mitigation requires more stringent efforts to reduce traditionally unregulated ammonia emissions.[46,58,59] In addition, measures to better characterize and regulate sources of other components including ammonium, dust, and BC can further aid the mitigation success[60−62] and possibly meet the more rigorous WHO guideline (5 $\mu g/m^3$).[63] Ongoing development of the combined ground- and satellite-based monitoring systems, together with advanced modeling capabilities, will be needed to assess and guide progress (Text S3). Following the growing recognition of health impacts of $PM_{2.5}$ at low concentrations (e.g., < 10 $\mu g/m^3$),[1] activities to sustain the air quality improvement in the US are increasingly crucial, calling upon improved understanding and mitigation actions of both combustion-related and traditionally unregulated sources.

## ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestair.4c00004.

Complementary descriptions of the used data sets and uncertainties, figures, and table to support interpretation (PDF)

## AUTHOR INFORMATION


### Corresponding Author

**Chi Li** − *Department of Energy, Environmental & Chemical Engineering, Washington University in St. Louis, St. Louis, Missouri 63130, United States;* orcid.org/0000-0002-8992-7026; Email: lynchlee90@gmail.com

### Authors

**Randall V. Martin** − *Department of Energy, Environmental & Chemical Engineering, Washington University in St. Louis, St. Louis, Missouri 63130, United States*

**Aaron van Donkelaar** − *Department of Energy, Environmental & Chemical Engineering, Washington University in St. Louis, St. Louis, Missouri 63130, United States*

Complete contact information is available at: https://pubs.acs.org/10.1021/acsestair.4c00004


### Author Contributions

The manuscript was written through contributions of all authors. The conceptualization was initialized by CL and RVM. The data and methodology were developed by CL and AVD. CL performed the visualization of the results, which were analyzed by all the authors. CL wrote the original draft. All authors have reviewed, edited, and given approval to the final version of the manuscript.

### Notes

The authors declare no competing financial interest.



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