

Changes in PM_{2.5}-Attributable Mortality in the US by Sector, 2002–2019

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ABSTRACT: Levels of fine particulate matter (PM_{2.5}) air pollution in the United States have declined substantially in recent decades, yielding substantial benefits to public health. This study evaluates emission reductions across five key economic sectors—electricity, industrial, transportation, agriculture, and residential—and their impact on air quality and health. We employ a recently developed sector-specific inventory that provides emissions and their spatial disaggregation across time in a self-consistent framework. Using a national source-receptor matrix, we estimate annual PM_{2.5}-attributable mortality and its variability spatiotemporally and by sector. We find that annual PM_{2.5}-attributable mortality decreased 51% between 2002 (197,000 deaths) and 2019 (96,000 deaths). The largest reductions were from electricity and transportation, especially secondary PM_{2.5} from NO_x, SO_x, and VOC emissions. Emissions reductions from industrial and residential sectors were more modest. In contrast, agricultural emissions, especially NH₃, increased over time; the importance of agriculture among the five sectors increased from second-smallest (2002) to the largest (2019). While the reductions in PM_{2.5}-attributable mortality have been large (approximately a factor of 2), future progress may need to focus greater attention on agricultural emissions, in addition to traditionally dominant sources such as transportation and industry.

KEYWORDS: PM_{2.5}, attributable mortality, anthropogenic emissions, sectoral contributions, emission trends

INTRODUCTION

In recent decades, levels of criteria air pollution have declined substantially in the United States (US), yielding large public health benefits. Between 1990 and 2017, emissions of PM_{2.5} (i.e., fine particles), NO_x, SO_x, and VOCs decreased 29%, 58%, 88%, and 40%, respectively.¹ US Environmental Protection Agency (USEPA) monitoring stations indicate that annual PM_{2.5} levels declined 42% during 2000–2017.¹ These improvements reflect local, regional, and national policy efforts, driven largely by the Clean Air Act.^{2–4}

Despite this progress, PM_{2.5}-attributable mortality is the largest environmental risk factor in the US.^{5–7} Designing effective interventions and policies requires identifying the emission sources and economic sectors that contribute most to this burden, as well as understanding how their impacts have evolved over time. Prior research has typically examined either temporal trends or sectoral contributions but rarely both.

Temporal studies, such as Zhang et al. (2018), Fann et al. (2017), and Cohen et al. (2017), documented how overall PM_{2.5}-related mortality has declined over the past decades.^{8–10} Sectoral studies, such as Caiazzo et al. (2013) and Thakrar et al. (2020), provided detailed breakdowns of the contributions of specific economic sectors but were restricted to single years.^{11,12} A smaller set of studies—such as Fann et al. (2013), Tschofen et al. (2019), and Dedoussi et al. (2020), combined

temporal and sectoral perspectives, but did so at only a few discrete time points.^{13–15} Because these analyses relied on the triennial National Emissions Inventory (NEI), they offered snapshots rather than continuous annual trends.

Here, we build on these foundations by providing annual estimates of anthropogenic PM_{2.5}-attributable mortality from 2002 to 2019, with results disaggregated into five major economic sectors (transportation, electricity, industry, agriculture, and residential) along with activities, processes, chemical species, and states. This annual resolution allows us to track both short-term fluctuations and long-term trends, offering a uniquely comprehensive view of the effectiveness of past air pollution controls and highlighting opportunities for future policy action.

MATERIALS AND METHODS

Emissions of primary PM_{2.5} and the four secondary precursors of PM_{2.5} (NO_x, SO_x, NH₃, and VOCs), for 18 years (2002–

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Table 1. Concentration–Response Functions (CRFs) Used in the Core Analysis and in Sensitivity Analyses, and Associated Mortality Estimates

Citation	Cohort ^a	Population age	Use	RR [95% CI] ^b	PM _{2.5} -attributable deaths
Orellano et al., 2024	Meta-analysis (Global)	All ages	Main analysis	1.095 [1.064 – 1.127]	
Orellano et al., 2024	Meta-analysis (American)	All ages	Main analysis	1.075 [1.055 – 1.096]	
Wu et al., 2020	Medicare (General)	Older adults (≥65)	Sensitivity	1.066 [1.058 – 1.074]	
Pope et al., 2019	NHIS	Adults (≥18)	Sensitivity	1.120 [1.080 – 1.150]	
Lepeule et al., 2012	Harvard Six Cities	Adults (≥25–74)	Sensitivity	1.14 [1.068 – 1.217]	
Krewski et al., 2009	ACS CPS- II (extended)	Adults (≥30)	Sensitivity	1.040 [1.030 – 1.060]	
Burnett et al., 2018 ^c	Global GEMM	Adults (≥25)	Sensitivity	GEMM NCD+LRI	

^aNHIS = National Health Interview Survey; ACS CPS-II = American Cancer Society Cancer Prevention Study II; GEMM = Global Exposure Mortality Model. ^bCI = confidence interval. GEMM NCD+LRI = Global Exposure Mortality Model function for noncommunicable diseases plus lower respiratory infections (i.e., all nonaccidental mortality). The log–linear CRFs (Orellano, Wu, Pope, Lepeule, Krewski) were applied using all-cause mortality without age-specific variation in relative risks; the nonlinear GEMM CRF (Burnett) was applied using nonaccidental (“non-acc.”) mortality, with parameters varying by age group. ^cApplying the GEMM nonlinear CRF at a location requires knowing the total PM_{2.5} at that location. To estimate total PM_{2.5} concentrations (including background and biogenic PM_{2.5}), here we employed census tract-level PM_{2.5} exposures from the Center for Air, Climate, and Energy Solutions (CACES) empirical model (www.caces.us/data). We applied a counterfactual minimum concentration of 2.4 $\mu\text{g}/\text{m}^3$, consistent with Burnett et al. (2018).

2019), were obtained from the USEPA EQUATES.¹⁶ Often, emission inventories are carried out for a single year at a time; spatial surrogates often change across inventories, making temporal comparisons more difficult and less useful (i.e., more challenging to interpret). In contrast, the EQUATES approach is internally consistent across time and space and covers the entire US, making it well-suited for our analysis.

EQUATES provides emissions at the county or latitude/longitude level; we gridded these emissions to our model domain using the USEPA spatial surrogates. Annual anthropogenic emissions in EQUATES are characterized into 5,434 Source Classification Codes (SCCs). Following Thakrar et al. (2020), we classify emissions in multiple ways: into five broad sectors (electricity, industrial, transportation, agriculture, residential), 25 activities, 10 processes, and 5 chemical species.¹² Nonanthropogenic emissions (e.g., wildfires and natural dust) and transboundary pollution are included in EQUATES but excluded here to maintain consistency with the model capabilities.

Changes in annual-average PM_{2.5} concentrations in the contiguous US were modeled using the Intervention Model for Air Pollution (InMAP), an open-source reduced-complexity model with a variable-resolution grid ranging from 1 km to 48 km depending on population density.¹⁷ Specifically, we use the InMAP source-receptor matrices (ISRM) derived from InMAP v1.6.1 to estimate changes in annual speciated PM_{2.5} concentrations (five species: primary PM_{2.5}, particulate nitrate, particulate ammonium, particulate sulfate, and secondary organic aerosol (SOA)) from each emission source. InMAP and the ISRM have been widely used in the literature;^{12,14,18–28} reported model performance indicates a population-weighted mean bias of $-3.1 \mu\text{g}/\text{m}^3$ and a mean

fractional bias (MFB) of -38% against observations, and $R^2 = 0.90$, MFB = -17% against WRF-Chem.

We estimate attributable mortality using InMAP-modeled PM_{2.5} concentrations, U.S. Census population data, and county-level all-cause mortality rates stratified by age group from the CDC.^{29,30} Each year's emissions are paired with population and mortality data from the same year to maintain temporal consistency. Our main analysis applies a log–linear concentration–response function (RR = 1.095 per 10 $\mu\text{g}/\text{m}^3$; 95% CI: 1.064 – 1.127) from Orellano et al. (2024), a recent meta-analysis.³¹ Sensitivity analyses use the nonlinear GEMM model (Burnett et al., 2018) and log–linear CRFs from Pope et al. (2019), Wu et al. (2020), Krewski et al. (2009), and Lepeule et al. (2012) with population and mortality matched to align with the respective study populations^{32–36} (see Table 1, below).

Because the underlying atmospheric chemistry in InMAP is calibrated to 2005 conditions, following Thakrar et al. (2024), we perform a sensitivity analysis to reflect the changes in background air pollution.²⁸ Declining background concentrations can alter the marginal impacts of precursor emissions—for example, NH₃ becomes less effective at forming particulate matter when NO_x and SO_x levels are lower, while SO_x can become relatively more influential.³⁷ Based on Holt et al. (2015), we adjusted precursor sensitivities (NH₃: -38% , NO_x: -29% , SO_x: $+23\%$ by 2012, relative to 2005) and assumed a linear change through 2019.³⁸ This adjustment tested the robustness of our results to background concentration trends.

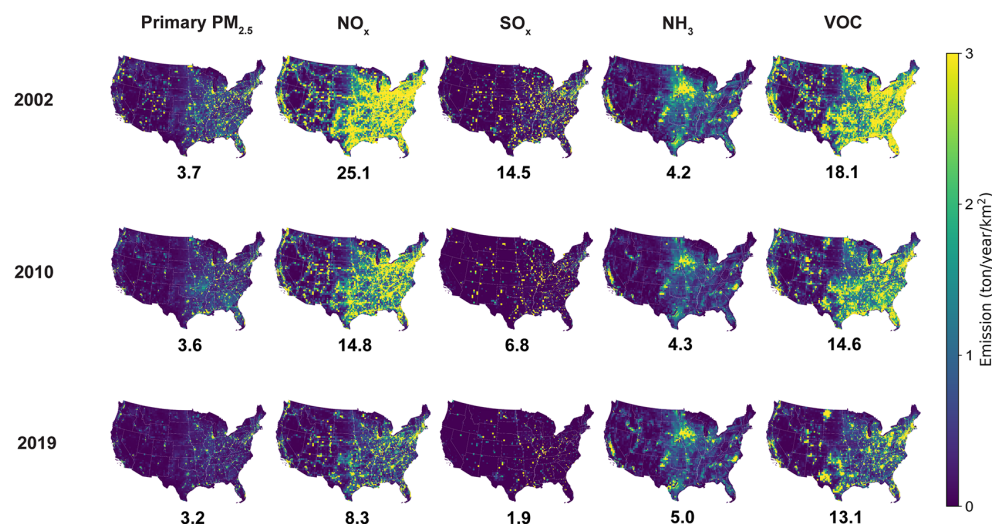


Figure 1. Annual emissions of primary PM_{2.5} and the four secondary-PM_{2.5} precursors (NO_x, SO_x, NH₃, VOCs), for three of the years. Values below each map indicate total anthropogenic annual emissions (Mt; 1 ton = 10^{−6} Mt = 2,000 pounds).

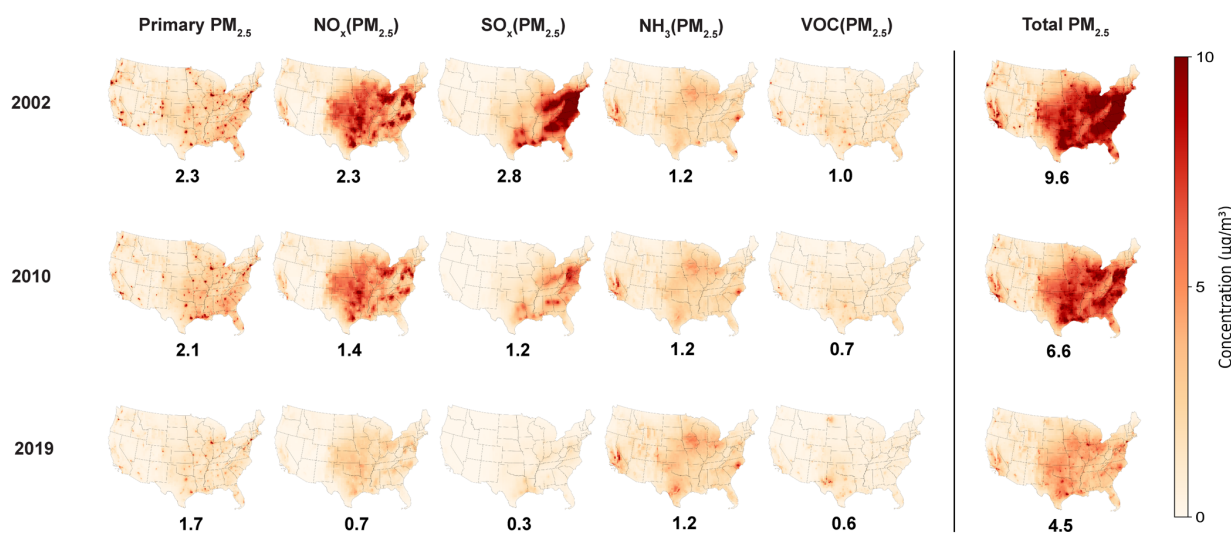


Figure 2. Annual-average concentrations of speciated and total PM_{2.5} for three of the years. Numbers below each map indicate the US population-weighted concentration (µg/m³) of total PM_{2.5} (rightmost column) or PM_{2.5} that can be attributed to the precursor shown (left five columns).

RESULTS

During 2002–2019, EQUATES emissions (primary PM_{2.5}, NO_x, SO_x, VOCs) declined significantly (Figure 1), with large reductions for SO_x (87%) and NO_x (66%), modest reductions for VOC (28%) and primary PM_{2.5} (14%), and an increase in NH₃ emissions (20%).

InMAP-estimated speciated and total anthropogenic PM_{2.5} concentrations (Figure 2) show corresponding changes during 2002–2019, with US population-weighted PM_{2.5} levels decreasing 90% (particulate sulfate), 70% (particulate nitrate), 40% (SOA), 25% (primary PM_{2.5}), and increasing 8% (particulate ammonium). The net result is that population-weighted total PM_{2.5} levels decreased 52%, from 9.6 µg/m³ (2002) to 4.5 µg/m³ (2019). Figures 1 and 2 show beginning, middle, and end years [2002, 2010, and 2019]; additional years are in Tables S1 and S2.

Annual anthropogenic PM_{2.5}-attributable mortality decreased by 51%, from 197,000 deaths (2002) to 96,000 deaths (2019). That result uses the Orellano CRF. Use of alternative CRFs (Table 1, Table S3) yielded attributable mortality estimates of ~107,000 (Wu) to ~283,000 (Lepeule) in 2002 and ~52,000 (Wu) to ~139,000 (Lepeule) in 2019. The temporal trend is consistent across CRFs (48% decline (Burnett); 51%–52% decline (remaining CRFs)).

Sectoral contributions to PM_{2.5}-attributable mortality shifted substantially between 2002 and 2019 (Figure 3, left). Attributable mortality declined for transportation (61% reduction), electricity (88%), industrial (45%), and residential (38%), but increased for agriculture (28%). In 2002, electricity generation and transportation were the leading contributors to PM_{2.5}-attributable mortality, whereas agriculture played a relatively minor role. By 2019, electricity had become one of the lowest-impact sectors, whereas agriculture had emerged as

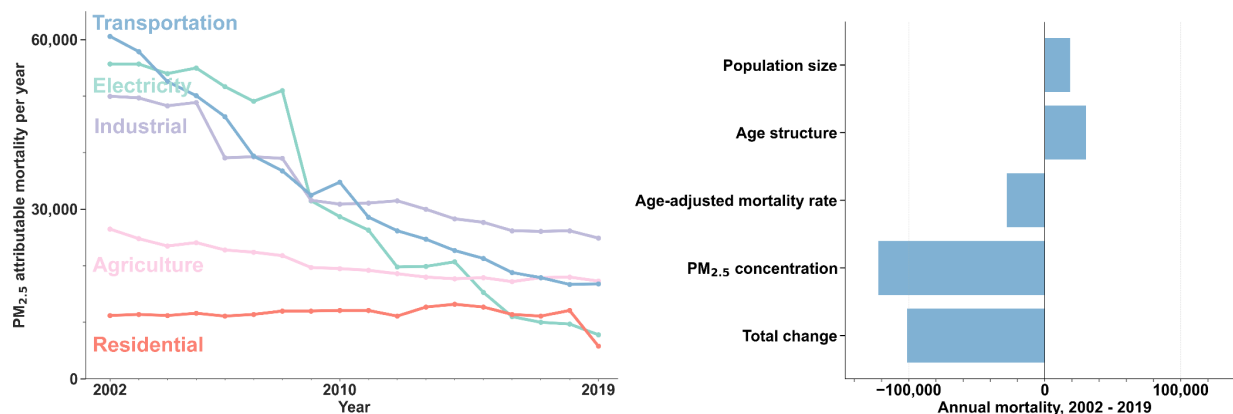


Figure 3. Disaggregation of changes in $\text{PM}_{2.5}$ -attributable mortality, 2002–2019: annually by sector (left plot) and attributable to changes in $\text{PM}_{2.5}$ concentration versus demographic attributes (right plot). Each bar (right plot) shows what the annual $\text{PM}_{2.5}$ -attributable deaths would be if only that attribute changed over time, while all others were held constant: Population size, age structure, age-adjusted baseline mortality rate, and $\text{PM}_{2.5}$ concentration (exposure). The final bar shows the total change.

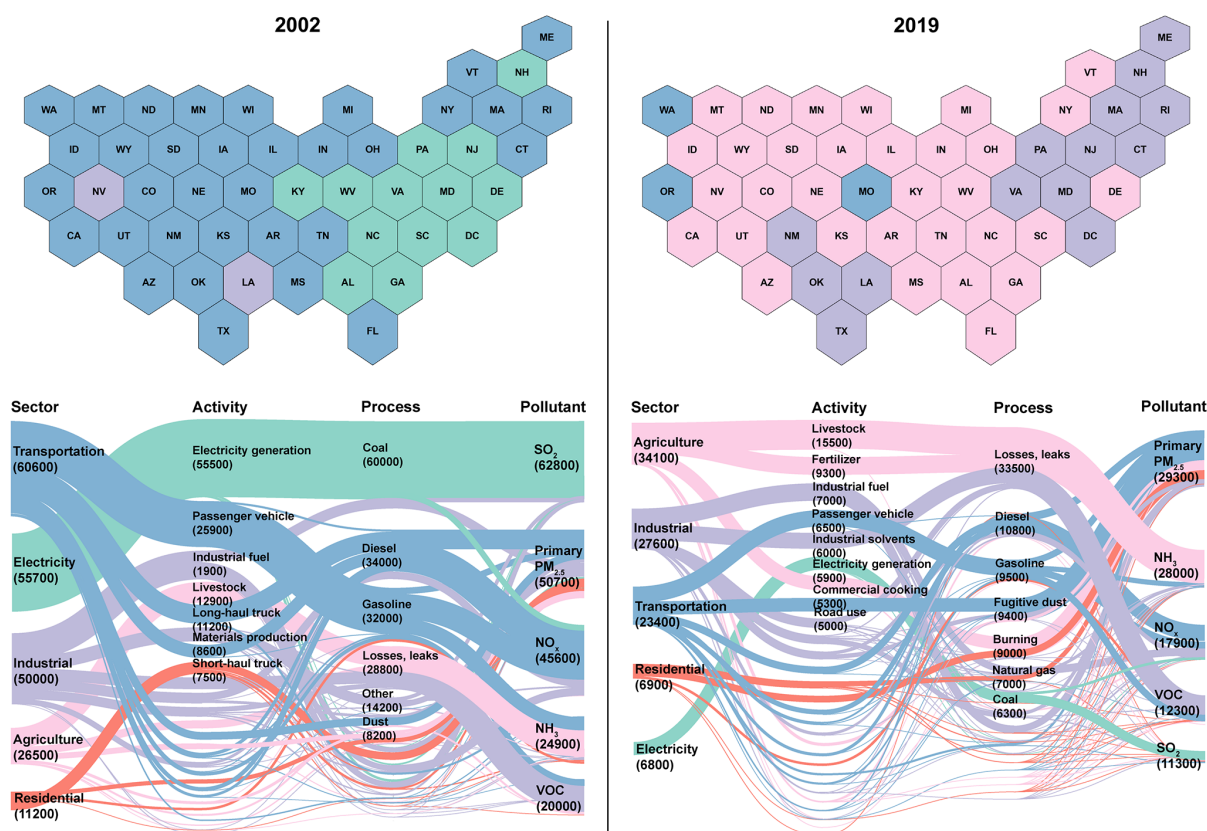


Figure 4. Economic sector (regardless of emission-location) that contributed the most to each state's anthropogenic $\text{PM}_{2.5}$ -related mortality in years 2002 and 2019 (upper plots). Annual attributable mortality, by sector, activity, process, and pollutant ($\text{PM}_{2.5}$ or its precursor), emitted for years 2002 and 2019 (lower plots). The number of deaths for each category is provided in parentheses. Within each column, flows are ordered from largest (top) to smallest (bottom) deaths per year. Activities and processes responsible for more than 5,000 annual deaths are labeled.

the largest contributor. This shift reflects in part the success of regulatory controls on fossil fuel combustion sources and the relative lack of comparable controls on agricultural emissions. Reductions in $\text{PM}_{2.5}$ -attributable mortality are driven by reductions in $\text{PM}_{2.5}$ levels; demographic changes, such as population aging, play a lesser role (Figure 3, right).

In a sensitivity analysis, when precursor sensitivities were adjusted for changing background concentrations (i.e., following Thakrar et al. (2024) and Holt et al. (2015); Figure S1), the overall temporal trends remained broadly consistent, but agricultural impacts decreased by 20% rather than increasing. This decrease was not driven by reductions in

agricultural emissions themselves (they still increase) but instead reflects declining background concentrations—owing to substantial emission reductions in other sectors, which reduced the marginal effectiveness of NH_3 in forming secondary $\text{PM}_{2.5}$. Under this sensitivity analysis, in 2019, agriculture ranked second (after industrial), rather than first. Together, these results similarly highlight how sectoral contributions have evolved, with strong progress in reducing combustion-related emissions but limited progress in addressing agricultural sources.

National trends in $\text{PM}_{2.5}$ pollution levels and attributable mortality reflect locally varying combinations of policies, emissions, population density, land use, and other factors. The largest economic sector contributing to attributable mortality by state has shifted over time (Figure 4, upper plots), from usually transportation and electricity (2002) to usually agriculture and industry (2019). A Sankey diagram, following Thakrar et al. (2020),¹² reveals the reshuffling of importance of specific activities, processes, and pollutants (Figure 4, lower plots). In 2002, leading activities and processes include coal-electricity, diesel-trucks, and gasoline-passenger vehicles. In 2019, the relative importance of livestock and fertilizer use increased, and coal-electricity has decreased. SO_x shifted from most-important species (2002) to least-important (2019). See SI for additional details.

The timing of emission reductions matters; enacting reductions sooner or more aggressively yields greater health benefits than delaying them. To illustrate this, we summed mortality attributable to emissions throughout 2002–2019. In total, 2.5 million deaths in the U.S. were attributable to anthropogenic $\text{PM}_{2.5}$ air pollution during 2002–2019. Had emissions remained at 2002 levels throughout, an additional 1.0 million deaths (a 40% increase) would have occurred (Figure S2). These avoided deaths highlight the substantial public health benefits of reductions in emission during this period.

DISCUSSION

We investigated $\text{PM}_{2.5}$ levels and attributable mortality by state, for 2002 to 2019 (annually), for each sector, activity, process, and chemical pollutant. $\text{PM}_{2.5}$ attributable mortality from anthropogenic US emissions declined substantially (51%), from 197,000 deaths (in 2002) to 96,000 (2019).

These anthropogenic emission reductions occurred across nearly all major sources, including an 89% (48,000 fewer deaths/y) reduction in annual mortality attributable to coal-fired electricity, a 68% (22,000) reduction from diesel-powered transportation, and 69% (23,000) for gasoline-transportation. Annual attributable mortality for agriculture increased 23% (7,000 more deaths/year), including attributable mortality increases of 20% (3,000) for livestock, 57% (3,000) for fertilizer use, and 54% (2,000) for commercial cooking. In aggregate during 2002–2019 (i.e., cumulative rather than annual), $\text{PM}_{2.5}$ pollution resulted in an estimated 2.5 million deaths; we estimate that that number would be 1.0 million deaths larger if emission-reductions had not happened (i.e., if emissions during 2002–2019 were constant at 2002 levels); see Figure S2.

Despite overall progress, annual air pollution-related mortality remains high at 96,000 deaths in 2019 (one death per ~6 min). Air pollution remains the leading environmental health risk in the United States.⁵ Emissions from agriculture (e.g., from fertilizer use; crop burning) remain largely unregulated

compared with other major pollution sources. Wildland fires (which were not modeled here) are increasing in frequency and intensity; recently, $\text{PM}_{2.5}$ levels might not have continued their downward trend.^{39,40} Emission-reduction efforts will need to consider both traditionally regulated sectors and emerging sources such as wildfires, in order to sustain and extend public health gains.

Our findings align with previous research on long-term air pollution-related mortality trends, which documented a decline in air pollution-related mortality over time in the United States^{8–10} (though acknowledging that recent trends may differ from longer-term trends).^{39,40} Additionally, our results are consistent with studies examining sectoral contributions to air pollution,^{11,12} which identified the electricity and transportation sectors as the dominant contributors in 2005, and the industrial sector as the leading source in 2014. Lastly, the concentration declines predicted here are strongly consistent with measurements at USEPA monitoring stations. Specifically, we compared our results against measured annual-average $\text{PM}_{2.5}$ levels at USEPA monitoring stations that had data across the relevant years (2002–2019) (Figure S3). The two trends (i.e., modeled, measured; 2002–2019) show remarkable similarity in temporal patterns (Figure S4), with a model-measurement correlation of 0.97 (Figure S5) and both indicating a ~5 $\mu\text{g}/\text{m}^3$ reduction in population-weighted $\text{PM}_{2.5}$ concentration during 2002–2019.

Our study expands upon these findings by integrating both mortality trends and sectoral contributions, providing a comprehensive, time-evolving analysis of air pollution sources. Our 18-year sector-specific analysis reveals the emerging dominance of agricultural emissions and the declining role of electricity generation in air pollution-related mortality. As shown above and in Figures S6 and S7, patterns differ by US state. These insights highlight the shifting burden of air pollution across sectors, reinforcing the need for adaptive regulatory strategies at the local, state, and national levels to address evolving sources of emissions.

Several limitations warrant acknowledgment. As stated above, the reported mean fractional bias for InMAP is –38% against observations, suggesting that predicted concentrations and associated mortality may be underestimated. Our modeling excludes long-range transport from outside the domain. The emission inventory omits biogenic and biomass sources, such as wildfires. While these limitations constrain our ability to quantify mortality from excluded sources (e.g., wildfires; outside-domain emissions), they might not strongly affect our main conclusions about relative temporal trends, sectoral reshuffling, or geographic shifts. For several core conclusions here (e.g., the growing importance of agricultural emissions), we believe that those findings would hold even if the model had zero bias.

Results also depend on the choice of concentration–response function, as shown above in sensitivity analyses (Table 1). $\text{PM}_{2.5}$ toxicities may vary by species, source, and size (Özkaynak et al., 1985; Zheng et al., 2025),^{41,42} yet our estimates assume uniform mass-based toxicity. Future work could investigate how differential toxicities would refine sector- and source-specific estimates.

■ ASSOCIATED CONTENT

Data Availability Statement

The analysis code is available at <https://github.com/bujinb/Equates> and the ISRM data set is available at [10.5281/zenodo.2589760](https://doi.org/10.5281/zenodo.2589760).

■ Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.estlett.5c00901>.

Detailed description of sector classifications: Table S1–S4 and Figure S1–S7 (PDF)

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Notes

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The authors declare no competing financial interest.

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