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 for 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 53% between 2002 (142,300 deaths) and 2019 (66,300 deaths). The largest reductions were in secondary $PM_{2.5}$ from NO_x, SO_x, and VOC emissions from electricity and transportation. Emissions reductions from industrial and residential sectors have been more modest. In contrast, agricultural emissions, especially NH_3, 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 (more than 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.

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].

$PM_{2.5}$-attributable mortality is the largest environmental risk factor in the US [5-7]. In understanding $PM_{2.5}$ health impacts and how to reduce those impacts, one would want to know which sources and economic sectors contribute to attributable impacts and also what are the patterns, especially the temporal trends, underlying those impacts. Prior research has generally *either* investigated temporal trends *or* the economic sectors contributing to $PM_{2.5}$-attributable health impacts, but not both. Examples of the former (i.e., studies of temporal trends) include the following. Zhang et al. (2018) modeled $PM_{2.5}$ and attributable mortality for 1990-2010 [8]. Fann et al. (2017) used monitoring data to estimate $PM_{2.5}$-attributable mortality for 1980, 1990, 2000, and 2010 [9]. Cohen et al. (2017) combined air quality model simulations, satellite data, and ground observations to evaluate temporal trends in $PM_{2.5}$-attributable disease burden at five-year intervals from 1990 to 2015 [10].

Examples of the latter (studies of sector-specific impacts) include Caiazzo et al. (2013), who assessed the impacts of six major economic sectors on $PM_{2.5}$ and O_3 -attributable mortality for year-2005 [11]. Thakrar et al. (2020) provided a detailed breakdown of $PM_{2.5}$-attributable mortality for multiple sectors, activities, and processes for year-2014 [12].

Here, we estimate $PM_{2.5}$-attributable mortality annually for 2002-2019. We disaggregate the results into five sectors (transportation, electricity, industry, agricultural, and residential) and as well into activities, processes, and chemical species, and by US state. These multiple disaggregations, along with temporal trends, can shed important light on the effectiveness of past air pollution controls and help identify challenges and opportunities for future policy efforts.

Methods

Emissions of primary $PM_{2.5}$ and the four secondary precursors of $PM_{2.5}$ (NO_x, SO_x, NH_3, and VOCs), for 18 years (2002-2019), are from USEPA EQUATES [13,14]. 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 the research questions investigated here.

In EQUATES, emissions are provided by county or latitude/longitude; we gridded them to the InMAP domain using 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 pollutants [12]. See Supplementary Information (SI) for additional details.

EQUATES includes non-anthropogenic emissions (e.g., wildfires) and transboundary pollution (e.g., emissions in other countries), however, those sources are outside the scope of this analysis to maintain consistency between emissions inputs and model capabilities.

Changes in annual-average $PM_{2.5}$ concentrations in the contiguous US are modeled using the Intervention Model for Air Pollution (InMAP), an open-source reduced-complexity model [14]. Specifically, we use the InMAP source-receptor matrices (ISRM) to estimate changes in annual speciated $PM_{2.5}$ concentrations (five species: primary $PM_{2.5}$, particulate nitrate, particulate ammonium, particulate sulfate, secondary organic aerosol (SOA)) from each emission source. InMAP and the ISRM have been widely used in the literature. Reported model performance indicates a population-weighted mean bias of -3.1 µg/m³ and a mean fractional bias (MFB) of -38% against observations, and $R^2=0.90$, MFB=-17% against WRF-Chem. (Additional model–measurement comparisons are in the Discussion section).

We use ISRM-estimated $PM_{2.5}$ concentrations, population data from the US Census, and county-level baseline mortality rates from the US Centers for Disease Control to calculate attributable mortality [15,16]. To spatially align emissions with the ISRM variable-resolution grid (ranging from 1 km to 48 km), we spatially allocate EQUATES inventory emissions using USEPA spatial surrogates following recommended methodology from Spatial Allocator Tool [19]. To ensure temporal consistency, we use population and mortality data from the same year as the emissions—for example, 2002 emissions are paired with 2002 Census and mortality data. This step is done for each sector, activity, processes and pollutants, for each year. We employ the dose response relationships from Nasari et al (2016), Burnett et al. (2018), and Krewski et al. (2009), as parametrized in Tessum et al. (2017) [7,17,18].

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 NH_3 emissions (20%). (Figure 1 uses Imperial units: 1 ton = 10^{-6} Mt = 2,000 pounds.)

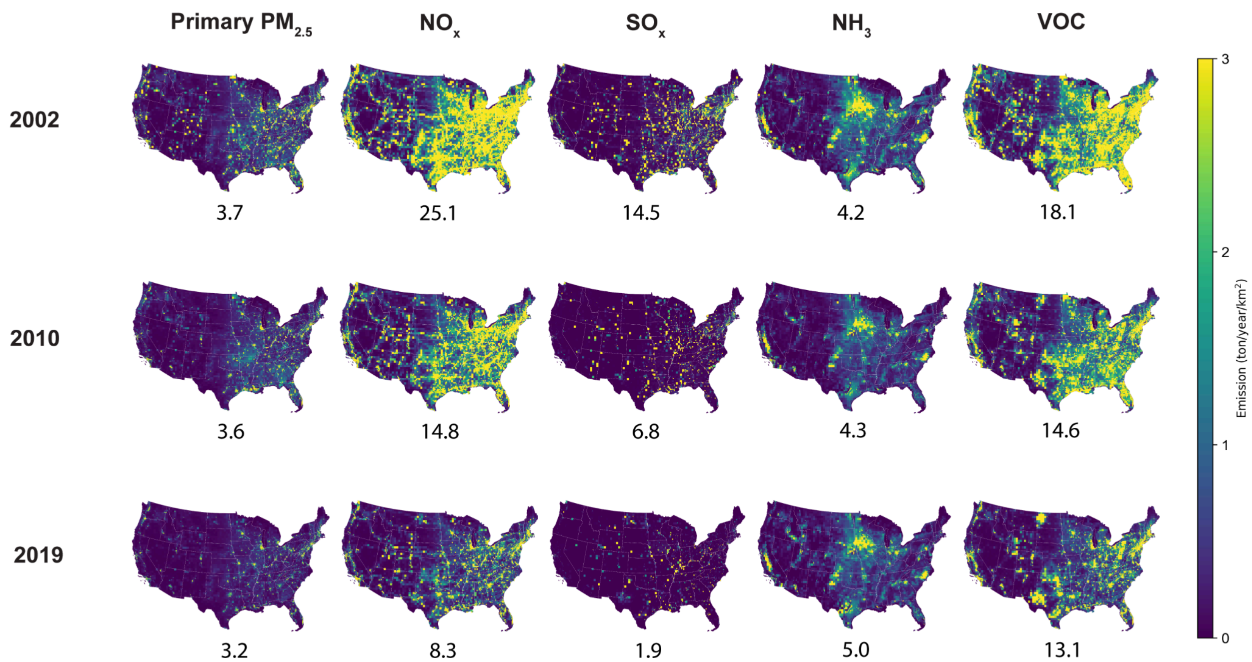

Figure 1. Annual emissions of primary $PM_{2.5}$ and the four secondary-$PM_{2.5}$ precursors (NO_x, SO_x, NH_3, VOCs), for three of the years. Numbers below each map indicate the total annual emissions (Mt).

ISRM-estimated speciated and total $PM_{2.5}$ 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), and 25% (primary $PM_{2.5}$) and increasing 8% (particulate ammonium). The net result is that population-weighted total $PM_{2.5}$ levels decreased 53%, from 9.6 µg/m³ (2002) to 4.5 µg/m³ (2019). Figures 1 and 2 show beginning, middle, and end years [2002, 2010, 2019]; additional years are in Table S1 and S2.

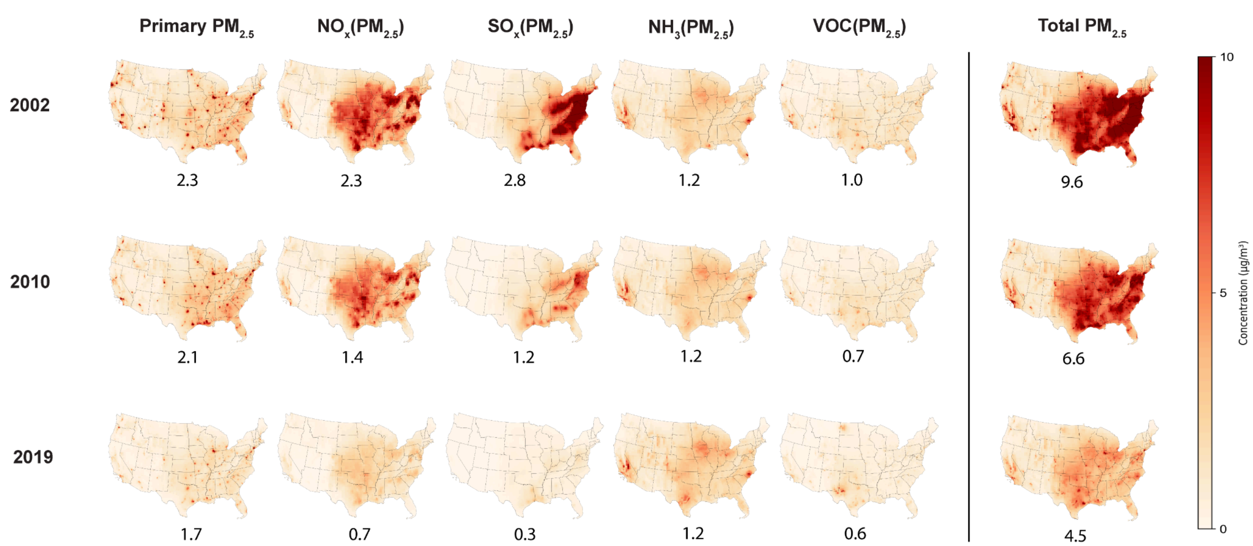

Figure 2. Annual-average concentrations of speciated and total $PM_{2.5}$ for three of the years. Numbers below each map indicate the US population-weighted concentration (µg/m³).

Annual attributable mortality decreased 53%, from 142,300 (in 2002) to 66,300 (2019), reflecting sector-specific reductions in attributable mortality from electricity (88% reduction, 2002-2019), industrial (47%), transportation (63%), and residential (41%), and increases for agriculture (23%) (Figure 3).

Two noticeable sectoral changes in Figure 3 are in the electricity sector, from relatively important (2002) to the lowest-impact sector (2019), and in agriculture, from relatively less important (2002) to the largest-impact sector (2019). Agriculture's increase was modest (23%), but is in contrast to large reductions in the remaining sectors (in aggregate, 65%).

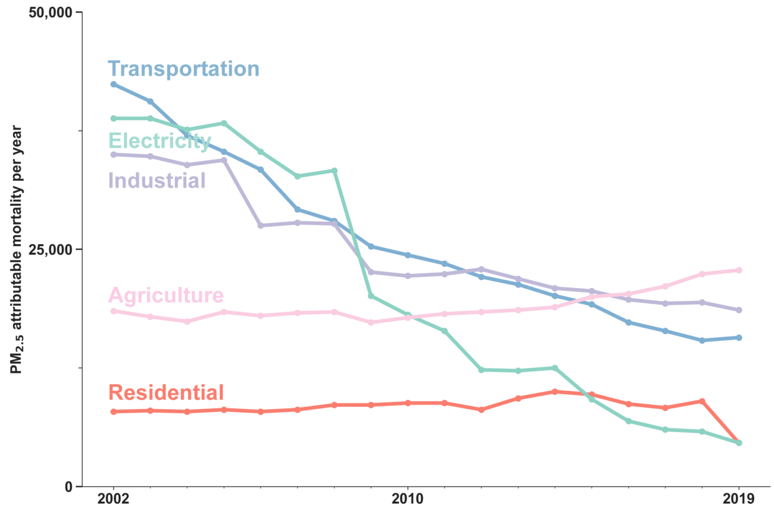

Figure 3. Annual attributable mortality by sector.

National trends in $PM_{2.5}$ pollution levels and attributable mortality reflect locally-varying combinations of policies, emissions, population density and 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). As mentioned above, and following Thakrar et al. (2020), we also disaggregated attributable mortality by activity, process, and pollutant [12]. Results (Figure 4, lower plots) document the dramatic reshuffling of importance of specific activities and processes. 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 have increased, and coal-electricity has decreased. SO_x shifted from most-important species (2002) to least-important (2019). See SI for additional details.

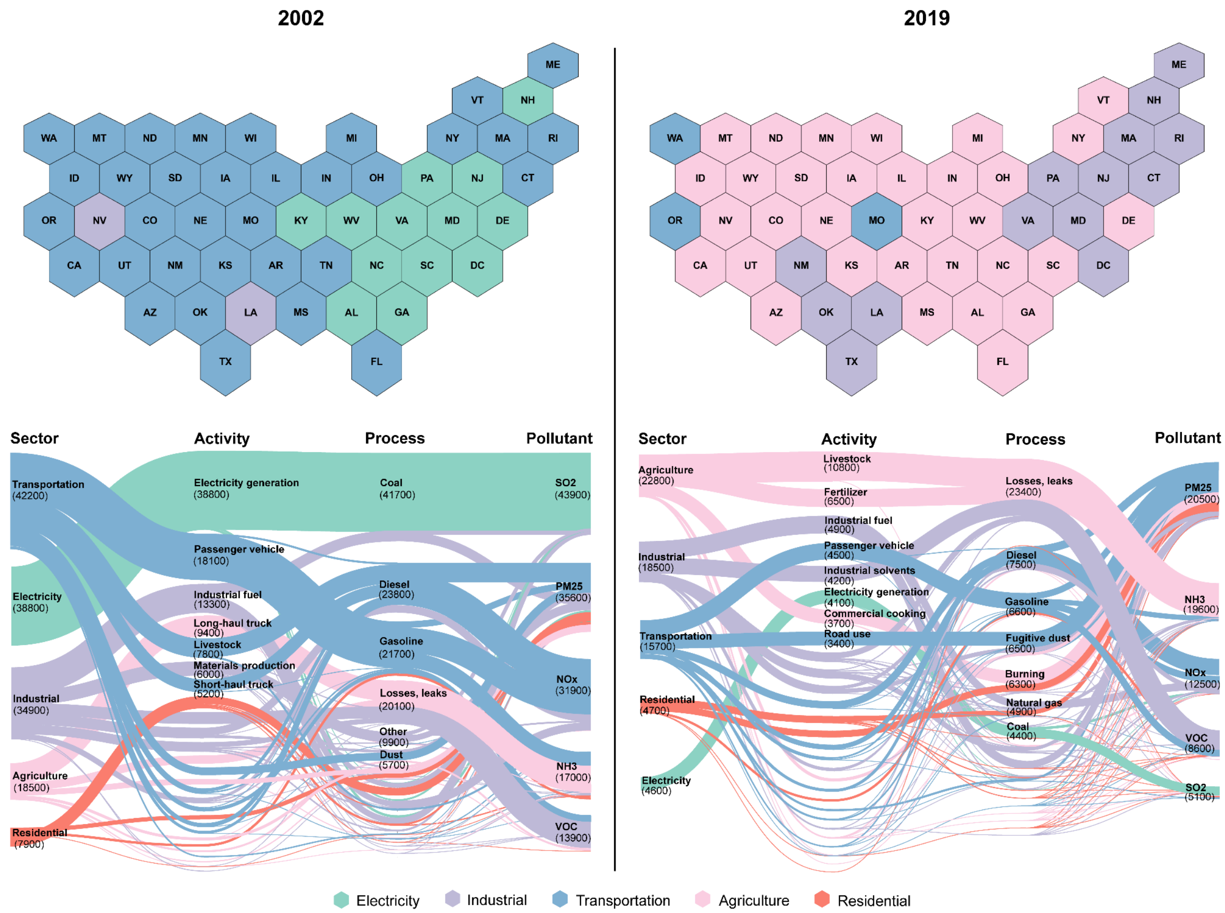

Figure 4. Economic sector contributing the most to air pollution-related mortality in each US state in years 2002 and 2019 (upper plots). Annual attributable mortality, by sector, activity, process, and pollutant emitted for years 2002 and 2019 (lower plots). The number of deaths for each category is provided in parentheses. Flows are color-coded by sector: electricity (green), industrial (purple), transportation (blue), agriculture (pink), and residential (red). Within each column, flows are ordered from largest (top) to smallest (bottom) deaths per year. Activities and processes responsible for more than 3,000 annual deaths are labeled.

The timing of emission-reduction matters; enacting emission reductions sooner or faster will result in greater total benefits to health than delaying the same reductions. To uncover this aspect, one can consider attributable mortality in cumulative, rather than annual, terms. Over the 18-year study period, in aggregate, approximately 1.8 million deaths in the US were attributed to $PM_{2.5}$ air pollution. If the observed emission-reductions had not happened (i.e., if emissions post-2002 were fixed at 2002 levels), we estimate that an additional 0.7 million attributable deaths (i.e., a 39% increase) would have occurred (Figure 5). Those avoided deaths reflect the public health improvements attributable to emission reductions during 2002-2019. Cumulatively during 2002-2019, transportation and industry were responsible for the highest number of deaths, followed by electricity; in recent years, contributions from these sectors have plateaued, while those from agriculture have continued to rise.

Of the 0.7 million avoided deaths, the electricity, transportation, and industrial sectors contributed the most. The cumulative (2002-2019) avoided mortality for agriculture is negative 9700 deaths (i.e., reflecting that agriculture emissions are increasing, counteracting the progress made in other sectors).

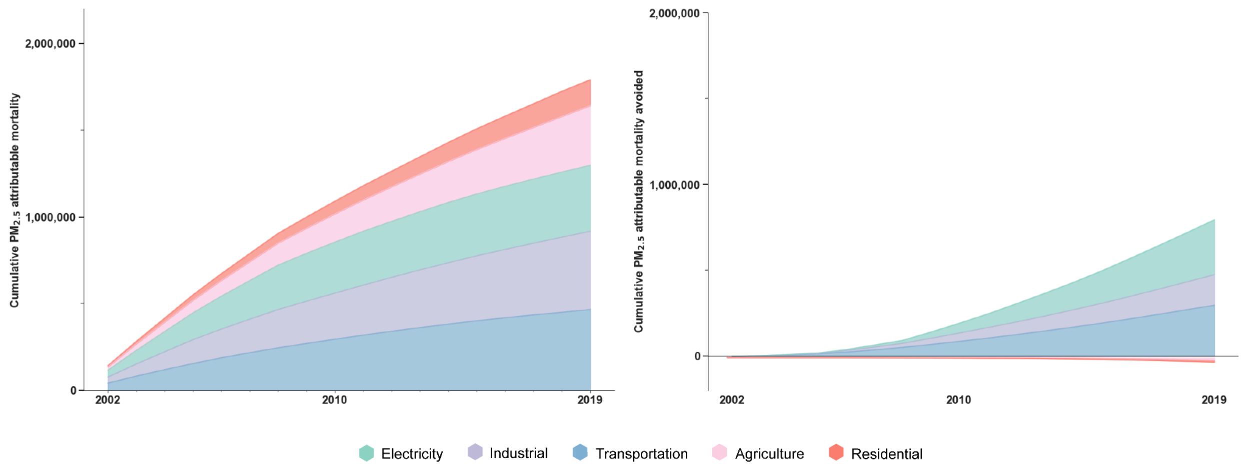

Figure 5. Cumulative attributable mortality (left) and avoided-mortality (right), 2002-2019. Left-plot represents cumulative attributable mortality for emissions as they actually happen in the data; right-plot reflects the cumulative attributable mortality difference between actual-emissions (i.e., the left plot) and if, counterfactually, emissions during 2002-2019 has stayed constant at year-2002 levels.

Discussion

We investigated $PM_{2.5}$ levels and attributable mortality by state, for 2002 to 2019 (annually), for each sector, activity, process, and chemical pollutant. $PM_{2.5}$ attributable mortality declined substantially (53%), from 142,300 deaths (in 2002) to 66,300 (2019).

These emission reductions occurred across nearly all major sources, including an 89% (37,300 fewer deaths/y) reduction in annual mortality attributable to coal-fired electricity, a 68% (16,300) reduction from diesel-powered transportation, and 69% (15,100) for gasoline-transportation. Annual attributable mortality for agriculture increased 23% (4,300 more deaths/y), including attributable mortality increases of 20% (1800) for livestock, 57% (2,300) for fertilizer use, and 54% (1,300) for commercial cooking. In aggregate during 2002-2019 (i.e., cumulative rather than annual), $PM_{2.5}$ pollution resulted in an estimated 1.8 million deaths; we estimate that that number would be 0.7 million deaths larger if emission-reductions had not happened (i.e., if emissions during 2002-2019 were constant at 2002 levels).

Additional examples of activities/processes with increasing rates for attributable mortality include garden equipment use (35%), burning industrial waste disposal (23%), fugitive dust from road use (22%) and tilling (11%); see SI ("Dataset") for complete list. This list can potentially be useful for framing emission-reduction efforts, including activities/sectors for which past emission-reduction efforts have been successful (or not) and for which future efforts should potentially focus additional attention.

Despite overall progress, annual air pollution-related mortality remains high at 66,300 deaths in 2019, reinforcing the fact that air pollution remains the leading environmental health risk in the United States. These findings underscore the need for continued targeted regulatory interventions to address $PM_{2.5}$ air pollution; this need includes agricultural emissions, which remain largely unregulated compared to other major pollution sources.

The overall reduction in attributable mortality (i.e., the 53% decline mentioned above) is primarily driven by decreases in average exposure, owing to emission reductions; indeed, the (emission-driven) changes in concentration, taken alone, account for a 50% drop in attributable mortality (i.e., nearly equal to the 53% decline in mortality). Over the same period, improvements in baseline mortality rates contributed an additional 25% reduction (i.e., baseline mortality [annual deaths per 100,000 people] was 845 in 2002, and 715 in 2019, a 15% decrease; this matters because $PM_{2.5}$-attributable mortality is calculated as a relative risk, i.e., a relative increase in the baseline mortality) [20,21]. However, population growth (288 million (2002), 328 million (2019), a 14% increase) led to a 22% increase in total attributable mortality [22,23]. (Here, attributable mortality nationally does not scale precisely linearly with national population because many variables, including population, baseline mortality, and concentration, vary in space and may be spatially correlated.) Thus, the attributable mortality *decrease* from improved health outcomes nearly offset the *increase* from population growth. (If we instead consider annual $PM_{2.5}$-attributable mortality per 100,000 people, that rate decreased 59%, from 49 in 2002 to 20 in 2019.)

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]. 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 $PM_{2.5}$ levels at USEPA monitoring stations that had data across the relevant years (2002 - 2019). The two trends (i.e., modeled, measured; 2002–2019) show remarkable similarity in temporal patterns (Figure S1), with a model-measurement correlation of 0.97 (Figure S2) and both indicating a ~5 μg/m³ reduction in population-weighted $PM_{2.5}$ 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 S3 and S4, patterns differ by US state. These insights highlight the shifting burden of air pollution across sectors, reinforcing the need for adaptive regulatory strategies at local, state, and national levels to address evolving sources of emissions.

Supplementary Information

The emissions groups used in this paper follow those used in the EPA NEI source categories, but with simplified aggregations that are more relevant for the analysis. All years in the EQUATES dataset were used, except that for year-2010 transportation NO_x emissions, owing to anomalies in the EQUATES data, we linearly interpolated those emissions between the year-2009 and -2011 emissions.

Below are definitions of some of the emissions groups as used here to clarify their scope and meaning by Thakrar et al. (2020) [12].

Sector. Broad aggregations of polluting activities largely following EPA NEI source categories.

Industrial & commercial. This sector largely overlaps with the EPA NEI industrial and commercial source categories, except for industrial and commercial sources that are within the scope of the other sectors, such as commercial cooking, which is included in the food & agriculture sector instead.

Transportation. This sector largely overlaps with the EPA NEI transportation source category, except for transportation sources that are within the scope of the other sectors, such as off-road agricultural machinery use, which is included in the food & agriculture sector instead.

Food & agriculture. This sector includes emissions that take place on farms and in agricultural operations, and is extended to include emissions from commercial food processing and cooking.

Activity. A human-caused action that directly results in pollution.

Crop burning. Crop burning is solely an agricultural activity. Land management for reducing wildfire risk, or burning logging residue, is not included within this sector nor within the scope of human- caused emissions.

Road use. Driving on roads causes emissions primarily by disturbing dust particles present on road surfaces, but also includes generating small particles from brake and tire wear. This activity also includes minor contributions from oil spills.

Commercial cooking & food processing. This activity includes food processing for retail as well as restaurant emissions.

Oil, gas & petroleum production. This activity largely overlaps with the EPA NEI source categories for the petroleum industry and the oil and gas industry. Emissions include flares and volatilization of products stored in tanks.

Solvent use. This includes only the use of solvents, such as applying paints or printer inks, and not the production or storage of solvents.

Waste disposal. This activity includes emissions from site remediation, and burning waste for the purpose of disposal. Waste combustion for use as a fuel is not included within the scope of this activity.

Materials production. This activity includes the production of any material used in goods production, such as glass, paper, and ceramic. Emissions from mining and smelting, for example, are often included in this activity.

Municipal vehicle use. This activity includes use of public buses and road cleaning vehicles.

Process. The means by which the pollution is generated, for any given activity.

Fuel combustion. Burning fuel for the purpose of energy use (e.g., heat or mechanical work).

Burning. This includes burning not for the purpose of energy use. Examples include waste burning, cropland residue burning, and residential and vehicle fires.

Losses & leaks. Losses & leaks are the inadvertent loss to the atmosphere of a product that is in gaseous or aerosol form, and typically valuable, but are not dust emissions. NH_3 volatilization and NMVOC evaporation are the main constituents of this group.

Table S1. Annual emissions (units: Mt = 10^6 ton = 2 x 10^9 pounds), by year, for primary $PM_{2.5}$, NO_x, SO_x, NH_3 and VOC, during 2002-2019

Year	Primary $PM_{2.5}$ (Mt)	NO_x (Mt)	SO_x (Mt)	NH_3 (Mt)	VOC (Mt)
2002	3.7	25.1	14.5	4.2	18.1
2003	4.2	24.3	14.6	4.4	19.0
2004	3.9	22.1	14.3	4.2	17.5
2005	4.0	21.7	14.7	4.5	17.6
2006	4.1	20.0	12.3	4.5	17.6
2007	4.1	18.7	11.4	4.7	17.7
2008	3.8	17.9	11.2	4.5	16.4
2009	3.4	15.3	7.9	4.3	14.6
2010	3.6	14.8	6.8	4.3	14.6
2011	3.7	14.3	6.3	4.5	15.4
2012	3.5	13.5	5.0	4.5	15.6
2013	3.2	12.6	4.7	4.4	14.1
2014	3.3	11.9	4.5	4.3	14.2
2015	3.4	10.8	3.4	4.5	14.9
2016	3.2	9.7	2.6	4.5	13.5
2017	4.3	9.3	2.5	4.7	16.2
2018	4.1	8.8	2.3	5.0	15.7
2019	3.2	8.3	1.9	5.0	13.1

Table S2. Population-weighted average concentration, by year, of primary $PM_{2.5}$, NO_x, SO_x, NH_3,VOC and total $PM_{2.5}$, during 2002-2019

Year	Primary $PM_{2.5}$ ($\mu g/m^3$)	NO_x ($\mu g/m^3$)	SO_x ($\mu g/m^3$)	NH_3 ($\mu g/m^3$)	VOC ($\mu g/m^3$)	Total $PM_{2.5}$ ($\mu g/m^3$)
2002	2.3	2.3	2.8	1.2	1.0	9.6
2003	2.4	2.3	2.8	1.2	1.0	9.7
2004	2.3	2.1	2.7	1.2	1.0	9.3
2005	2.4	2.0	2.7	1.3	1.0	9.4
2006	2.3	1.8	2.4	1.3	0.9	8.7
2007	2.3	1.6	2.2	1.2	0.9	8.2
2008	2.2	1.6	2.0	1.2	0.8	7.8
2009	2.0	1.4	1.4	1.2	0.7	6.7
2010	2.1	1.4	1.2	1.2	0.7	6.6
2011	2.0	1.3	1.1	1.2	0.7	6.3
2012	1.8	1.2	0.8	1.2	0.7	5.7
2013	1.8	1.1	0.8	1.2	0.7	5.6
2014	1.8	1.1	0.8	1.2	0.7	5.6
2015	1.8	1.0	0.6	1.2	0.7	5.3
2016	1.7	0.9	0.5	1.2	0.7	5.0
2017	2.0	0.8	0.4	1.2	0.7	5.1
2018	2.0	0.8	0.4	1.2	0.7	5.1
2019	1.7	0.7	0.3	1.2	0.6	4.5

Table S3. Annual attributable mortality, by year and sector, during 2002-2019

Year	Electricity	Industrial	Transportation	Agriculture	Residential	Total
2002	38800	34900	42200	18500	7900	142300
2003	38800	34800	40600	17900	8000	140100
2004	37600	33900	37000	17400	7900	133800
2005	38300	34400	35300	18400	8100	134500
2006	35300	27500	33400	18000	7900	122100
2007	32700	27800	29200	18300	8100	116100
2008	33300	27700	28000	18400	8600	116000
2009	20100	22600	25300	17300	8600	93900
2010	18100	22200	35600	17800	8800	102500
2011	16400	22400	23500	18200	8800	89300
2012	12300	22900	22100	18400	8100	83800
2013	12200	21900	21300	18600	9300	83300
2014	12500	20900	20100	18900	10000	82400
2015	9200	20600	19200	20000	9700	78700
2016	6900	19700	17300	20300	8700	72900
2017	6000	19300	16400	21100	8300	71100
2018	5800	19400	15400	22400	9000	72000
2019	4600	18500	15700	22800	4700	66300

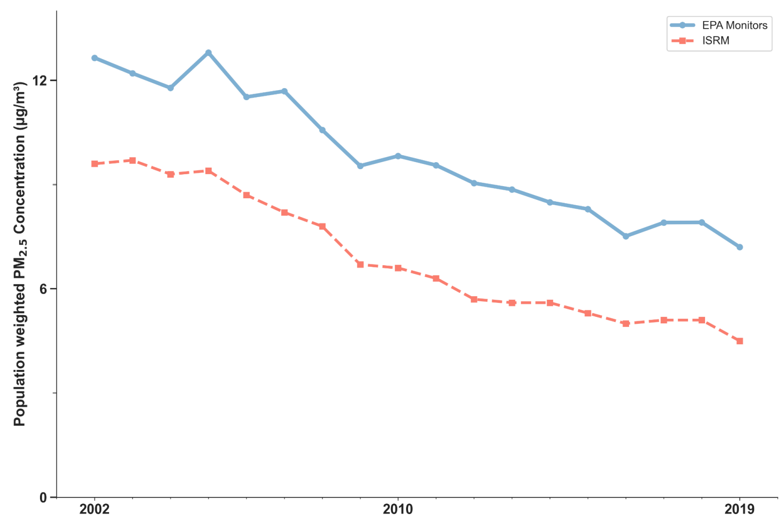

Figure S1. Trend in population-weighted annual-average concentrations of $PM_{2.5}$, 2002-2019, from EPA monitors and InMAP estimates. Estimates from EPA monitors are based on data from the n=72 monitoring stations with measurements reported for all 18 years of the study period.

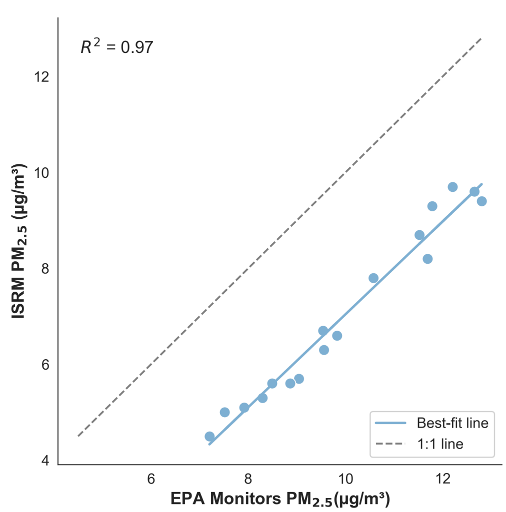

Figure S2. Comparison of population-weighted annual-average $PM_{2.5}$ concentrations from 2002 to 2019 between EPA monitors and ISRM (InMAP) estimates. InMAP estimates are consistently lower than EPA observations, likely due to the exclusion of non-anthropogenic emissions (e.g., wildfires, windblown dust) and transboundary pollution (e.g., emissions from other countries). Data in this figure are the same data as in Figure S3; each plotted point is one year in Figure S3. For more details on InMAP performance and limitations, see Tessum et al. (2017) [14].

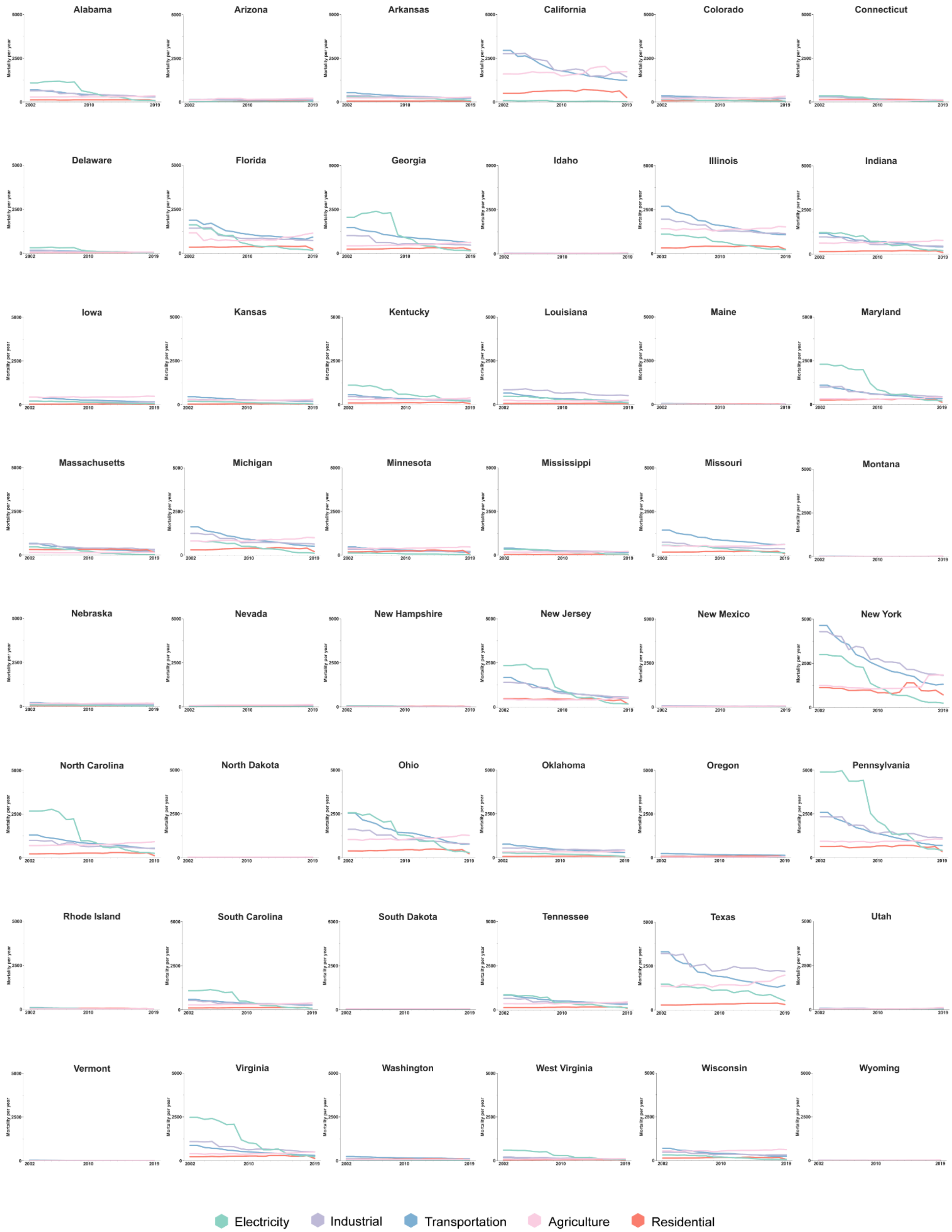

Figure S3. Trend in total $PM_{2.5}$-attributable mortality by sector across the 48 contiguous U.S. states. (This figure is similar to Figure 3, but disaggregated by state.)

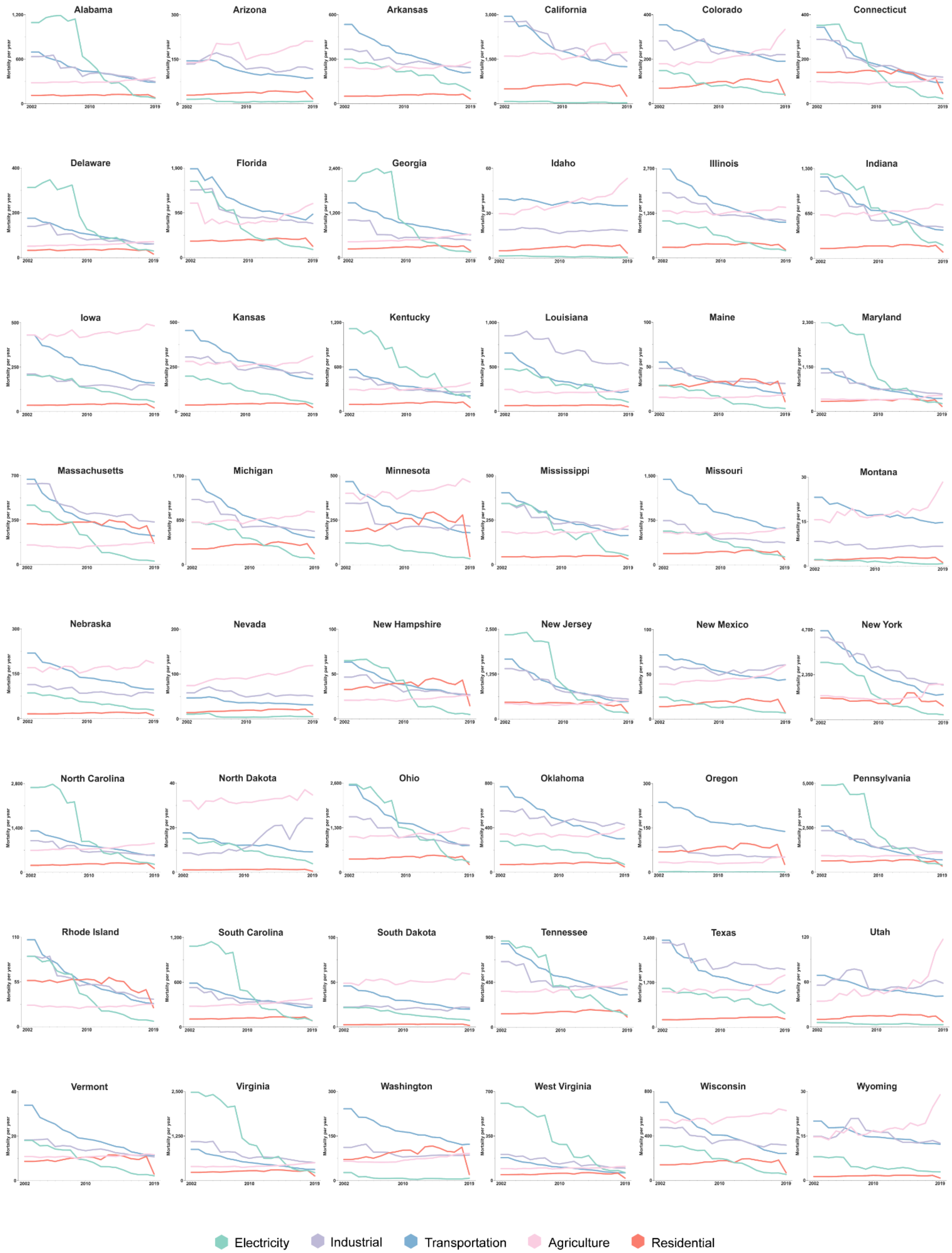

Figure S4. Trend in total $PM_{2.5}$-attributable mortality by sector across the 48 contiguous U.S. states. This figure is similar to Figure S3, but with state-specific y-axis scaling to magnify and illustrate trends in states with lower mortality levels.

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