Long-term mortality burden trends attributed to black carbon and $PM_{2.5}$ from wildfire emissions across the continental USA from 2000 to 2020: a deep learning modelling study

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Summary

Background Long-term improvements in air quality and public health in the continental USA were disrupted over the past decade by increased fire emissions that potentially offset the decrease in anthropogenic emissions. This study aims to estimate trends in black carbon and $PM_{2.5}$ concentrations and their attributable mortality burden across the USA.

Methods In this study, we derived daily concentrations of $PM_{2.5}$ and its highly toxic black carbon component at a 1-km resolution in the USA from 2000 to 2020 via deep learning that integrated big data from satellites, models, and surface observations. We estimated the annual $PM_{2.5}$-attributable and black carbon-attributable mortality burden at each 1-km² grid using concentration–response functions collected from a national cohort study and a meta-analysis study, respectively. We investigated the spatiotemporal linear-regressed trends in $PM_{2.5}$ and black carbon pollution and their associated premature deaths from 2000 to 2020, and the impact of wildfires on air quality and public health.

Findings Our results showed that $PM_{2.5}$ and black carbon estimates are reliable, with sample-based cross-validated coefficients of determination of 0·82 and 0·80, respectively, for daily estimates (0·97 and 0·95 for monthly estimates). Both $PM_{2.5}$ and black carbon in the USA showed significantly decreasing trends overall during 2000 to 2020 (22% decrease for $PM_{2.5}$ and 11% decrease for black carbon), leading to a reduction of around 4200 premature deaths per year (95% CI 2960–5050). However, since 2010, the decreasing trends of fine particles and premature deaths have reversed to increase in the western USA (55% increase in $PM_{2.5}$, 86% increase in black carbon, and increase of 670 premature deaths [460–810]), while remaining mostly unchanged in the eastern USA. The western USA showed large interannual fluctuations that were attributable to the increasing incidence of wildfires. Furthermore, the black carbon-to-$PM_{2.5}$ mass ratio increased annually by 2·4% across the USA, mainly due to increasing wildfire emissions in the western USA and more rapid reductions of other components in the eastern USA, suggesting a potential increase in the relative toxicity of $PM_{2.5}$. 100% of populated areas in the USA have experienced at least one day of $PM_{2.5}$ pollution exceeding the daily air quality guideline level of 15 μg/m³ during 2000–2020, with 99% experiencing at least 7 days and 85% experiencing at least 30 days. The recent widespread wildfires have greatly increased the daily exposure risks in the western USA, and have also impacted the midwestern USA due to the long-range transport of smoke.

Interpretation Wildfires have become increasingly intensive and frequent in the western USA, resulting in a significant increase in smoke-related emissions in populated areas. This increase is likely to have contributed to a decline in air quality and an increase in attributable mortality. Reducing fire risk via effective policies besides mitigation of climate warming, such as wildfire prevention and management, forest restoration, and new revenue generation, could substantially improve air quality and public health in the coming decades.

Funding National Aeronautics and Space Administration (NASA) Applied Science programme, NASA MODIS maintenance programme, NASA MAIA satellite mission programme, NASA GMAO core fund, National Oceanic and Atmospheric Administration (NOAA) GEO-XO project, NOAA Atmospheric Chemistry, Carbon Cycle, and Climate (AC4) programme, and NOAA Educational Partnership Program with Minority Serving Institutions.

Lancet Planet Health 2023; 7: e963–75

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Introduction

$PM_{2.5}$ has a substantial impact on air quality, climate change, and public health.[1] Understanding and estimating this impact requires knowledge of the spatiotemporal variations of the amount and composition of surface-level $PM_{2.5}$. Multiple factors make this challenging, including the change in and diversity of aerosol sources and aerosol processes, as well as the small number of surface observation stations. Anthropogenic emissions are being regulated in many countries, whereas wildfires show significant temporal variations. Both are significant contributors to the $PM_{2.5}$ mass and composition, which includes sulphate, nitrate, ammonium, organic carbon, and black carbon. Of particular importance is black

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Research in context

Evidence before this study

We searched Google Scholar for all publications related to the terms “wildfires” OR “black carbon” AND “public health” OR “mortality burden” in the USA in English from Jan 1, 2019 to Aug 1, 2023. Most studies focused only on several specific years with major fires (eg, 2020), and assessed the total $PM_{2·5}$ mass to study the impact on air quality, climate change, health benefits, and associations with cardiovascular or nervous system diseases by cohort data analysis at the state or regional scale (eg, California). Due to the scarcity of long-term and high-resolution $PM_{2·5}$ data, especially its chemical composition, to date, few, if any, nationwide studies have investigated the long-term variations in wildfire-related species (black carbon) of fine particulate pollution and its attributed mortality.

Added value of this study

Our results advance previous studies by examining the impact of fires on the long-term public health burden in the USA. We first estimated daily gapless (ie, spatially complete coverage) $PM_{2·5}$ and black carbon concentrations at 1-km resolutions and showed that their significant decreases alleviated the mortality burden in the USA from 2000 to 2020. However, the rates of decline have slowed down nationally, and have even reversed to increase in the western USA, due to increased fire emissions in the past decade. When the greater toxicity of black carbon is considered, $PM_{2·5}$ led to an increase of around 930 deaths per year in the western USA, compared with an increase of 670 deaths per year when black carbon is not considered. This is much higher than the number of casualties directly caused by wildfires (around 89 deaths per year in the USA). The health benefits from air quality improvement measures are significantly offset by wildfires.

Implications of all the available evidence

Increasing wildfires in number and intensity in the USA tend to offset or even overshadow the reduction in anthropogenic emissions, aggravating air pollution and increasing the risk of both morbidity and mortality. Reducing climate warming, and hence reduced fire occurrence, should be an integral part of environmental protection for the sake of public health.

carbon due to its strong absorption of solar radiation, which can worsen air quality due to its positive feedback with the planetary-boundary layer,[2] augmenting the effects on climate, as well as its potentially high toxicity and hence more severe impact on public health.[3,4] Despite well documented historical records of anthropogenic emissions in the USA, the national effects of reduced emissions from specific air pollution sources that influence $PM_{2·5}$ and black carbon concentrations on public health have not been studied on decadal scales. The public health outcomes of air pollution concentrations are further obscured by large annual fluctuations in fire emissions and associated uncertainties, which vary regionally and seasonally. The long-term and acute impacts of increased $PM_{2·5}$ from wildfires on many health outcomes are well known.[5,6] However, little is known about the specific health effects of black carbon from fire emissions.

Previous studies have integrated satellite-based aerosol optical depth (AOD) products with in-situ ground measurements to estimate surface $PM_{2·5}$ over the USA via approaches such as Kriging,[7] land-use regression and Bayesian maximum entropy modelling,[8] gradient-boosted trees,[9] random forest,[10] geographically weighted regression,[11] machine-learning ensemble-based modelling,[12,13] neural network,[14] and convolutional neural network.[15] Unlike $PM_{2·5}$, there are few studies focusing on black carbon estimates in the USA.[11,16] Although the $PM_{2·5}$ concentration in the USA has been postulated to be declining due to persistent regulations aimed at reducing anthropogenic emissions since the enactment of the Clean Air Act of the 1970s, this conjecture cannot be fully verified with surface observation alone because it does not provide full continental spatial coverage, especially when considering the increase in fires in the western USA in the past decade.[6,17] Fire emissions are the second largest source of black carbon in the USA and a significant contributor to $PM_{2·5}$ in fire-prone areas,[18,19] potentially increasing the amount and toxicity of ambient $PM_{2·5}$ and exacerbating its health impacts.[19–21] This leads to the hypothesis that the overall impact of $PM_{2·5}$ on public health might not have changed or could even have increased in the past two decades in the USA, at least in the western USA.

The mortality burden associated with $PM_{2·5}$ exposure has been estimated in many studies, often assessed by integrating $PM_{2·5}$ mass concentrations and population density distributions with concentration–response functions (CRFs).[22] Apte and colleagues[23] showed that global targets for $PM_{2·5}$ set out by the WHO air quality guideline in 2006 (annual mean $PM_{2·5}$ of 10 μg/m³) have not been met. The emission reduction of global $PM_{2·5}$ needed to meet this standard could have averted 23% of population deaths attributable to ambient $PM_{2·5}$ in 2010. However, populations in cleaner areas are more sensitive to $PM_{2·5}$ at lower concentrations. Therefore, the increase in $PM_{2·5}$ by fire emissions in the USA might cause a higher burden on public health than in often more polluted countries such as China and India. Aguilera and colleagues[21] found that wildfire-related $PM_{2·5}$ posed a greater risk to the human respiratory system than other sources of $PM_{2·5}$ in southern California during 1999–2012. O’Dell and colleagues[19] showed that the majority of deaths linked to smoke-related $PM_{2·5}$ occurred outside the western USA between 2006 and 2018. By contrast, few have investigated the health impacts of black carbon in the USA, which is due in part to the scarcity of exposure data sources and the absence of a CRF for black carbon. Smith and colleagues[24] provided estimates of the mortality effects related to long-term black carbon exposure in 66 US cities through a large time series and cohort study; their findings showed a greater health impact of black carbon compared with undifferentiated $PM_{2·5}$. Wang and colleagues[25] and Pond and colleagues[26] found strong positive associations between both cardiopulmonary mortality and all-cause mortality and exposure to black carbon in the USA.[27] Li and colleagues[16] estimated that around 14 000 premature deaths were caused by ambient black carbon in 2010 in the USA.

In the current study, we investigate how the surface $PM_{2·5}$ mass and its fraction of black carbon changed in the past two decades in the continental USA, and how much change (if any) in mortality burden due to $PM_{2·5}$ exposure can be attributed to fires. We address both questions by building upon the advances enabled by deep learning and long-term aerosol measurements from both space and the surface over the USA. We first derive daily surface $PM_{2·5}$ and black carbon concentrations from 2000 to 2020 in the USA with full spatial coverage at each 1-km² grid in the continental USA. Our study integrates multiple sources of satellite-based data products, re-analysis datasets of aerosol composition, and datasets from surface monitoring stations in the USA. Our method mitigates the impact of missing data associated with the spatial gaps in satellite AOD retrievals due to clouds and surface snow or ice cover and considers the spatiotemporal heterogeneity in the AOD–$PM_{2·5}$ relationship. We also evaluate the long-term (2000 to 2020) mortality burden in terms of the number of premature deaths associated with the change in annual $PM_{2·5}$ and black carbon concentrations and investigate the role of fire emissions in changing the annual mortality burden since the start of the new millennium. For the mortality burden assessment, the CRF of $PM_{2·5}$ was collected from a recent cohort study in the USA.[27] A sensitivity study was also conducted by taking the CRF of black carbon from a meta-analysis[3] to consider the potentially greater toxicity of black carbon compared with other $PM_{2·5}$ components.

Methods

Data sources

We selected the continental USA as the study region and further divided it into the eastern USA, western USA, and central USA based on geographical areas (appendix p 9). Measurements of surface 24-h average $PM_{2·5}$ concentrations were collected daily from the Environmental Protection Agency (EPA) Air Quality System and every third day from the Interagency Monitoring of Protected Visual Environments (IMPROVE)[28] at approximately 2740 monitoring stations from 2000 to 2020 throughout the USA (appendix p 9). Spatial representation was optimised by integrating the EPA and IMPROVE networks, in which monitors are distributed mainly in urban and rural areas, respectively. To further enhance the modelling of $PM_{2·5}$ levels during wildfire smoke events, daily $PM_{2·5}$ AirFire monitoring data were incorporated into our model, collected from the US Forest Service AirSis and Western Regional Climate Center archives at approximately 2800 monitoring locations, primarily located in the western USA.[13] Daily black carbon concentrations spanning the period 2000 to 2020 were collected from the EPA Chemical Speciation Monitoring Network and the IMPROVE network at approximately 370 monitoring sites across the USA, which are used as the ground truth for black carbon modelling.

Daily 1-km resolution Multi-Angle Implementation of Atmospheric Correction (MAIAC) Collection 6 AOD (at 550 nm) products (MCD19A2) were employed, developed from Moderate Resolution Imaging Spectroradiometer (MODIS) instruments on the Terra (approximately 1030 h local time) and Aqua (approximately 1330 h local time) satellites since their respective inception dates (Feb 24, 2000, and July 4, 2002) to the end of 2020. Estimates of the surface black carbon also used the Multi-angle Imaging SpectroRadiometer (version 23 level 3) monthly absorbing AOD product (0·5°×0·5°). Total aerosol extinction AOD, absorbing AOD (calculated by subtracting scattering AOD from total AOD), black carbon extinction AOD, and the surface mass concentrations of different aerosol components, including black carbon, organic carbon, dust, sulphate, and sea salt, were collected from Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2) aerosol diagnostics at a horizontal resolution of 0·625°×0·5°. Monthly anthropogenic emissions, including black carbon, ammonia, nitrogen oxides, sulphur dioxide, and volatile organic compounds, were obtained from the Copernicus Atmosphere Monitoring Service global emission inventories (0·1°×0·1°). Additionally, monthly smoke emissions from the Fire Energetics and Emissions Research database (0·5°×0·5° before 2003 and 0·1°×0·1° after 2003) were obtained.

Meteorological fields were extracted from ERA5-Land (0·1°×0·1°), including the 2-m temperature, precipitation, 10-m u-components and v-components of wind, and surface pressure, as well as ERA5 global reanalysis (0·25°×0·25°), including boundary layer height and relative humidity. In addition, the 90-m Shuttle Radar Topography Mission digital elevation model, monthly 1-km MODIS normalised difference vegetation index, and annual 1-km LandScan global population distribution products were used as inputs in deep learning modelling. All auxiliary variables (appendix p 25) were aggregated or resampled to 0·01°×0·01° grids (approximately 1 km) to be compatible with the resolution of MAIAC AOD products.

See Online for appendix

Surface $PM_{2.5}$ and black carbon estimates with deep learning

A deep learning model was trained by using the aforementioned satellite data and model outputs as features and surface measurements of $PM_{2.5}$ and black carbon as targets. To improve the estimates of air pollutants, the spatiotemporal autocorrelation and difference in $PM_{2.5}$ and black carbon were considered in the deep learning (ie, deep forest), leading to an extended spatiotemporally weighted deep forest (SWDF) model (appendix pp 1–3).[29] It uses the cascade structure by including multiple random forests and extremely randomised trees in each middle layer. The final result was determined by integrating the results of all intermediate hidden layers using the Light Gradient Boosting Machine. This model exhibits a stronger data mining and integration capability than traditional models by harnessing the strengths of diverse ensemble tree-based models while requiring fewer hyperparameter settings.

Specifically, the model construction included two main steps: we first derived daily $PM_{2.5}$ by training the SWDF model between $PM_{2.5}$ measurements and AOD together with $PM_{2.5}$ components, meteorological fields, anthropogenic emissions of $PM_{2.5}$ precursors, and land-use and population variables. MAIAC AOD was the primary input to the deep learning model for $PM_{2.5}$ estimation. Terra and Aqua MODIS AOD retrievals were first integrated using a linear regression model to minimise the difference caused by different observation times and enlarge the spatial coverage. In conditions of cloud and snow or ice surfaces and places with satellite swath gaps where MAIAC AOD was missing, AOD values were provided using the MERRA-2 re-analysis. MERRA-2 AOD data are generated by assimilating a variety of satellite retrievals (including MODIS) and ground-based observations and have been shown to have similar accuracy to satellite AOD data in areas with high-density observation networks (eg, North America and Europe).[30] $PM_{2.5}$ estimates were subsequently used as the main predictor in the SWDF model to predict black carbon mass concentration. Additional factors highly associated with black carbon (eg, the absorbing AOD and black carbon AOD, and black carbon surface mass simulations and emissions) were also used as inputs in training (appendix p 10).

To validate our model, we used the sample-based ten-fold cross-validation method to assess the overall accuracy of our air pollutant estimates and ensure comparability with previous studies.[31–33] This procedure underwent nine runs and had all data samples randomly divided into ten folds, where the model was trained using nine folds and tested on the left-out fold. Furthermore, we adopted two additional independent spatial and temporal cross-validation approaches to ensure that our model was not overfitted with the training data. Similar ten-fold cross-validation procedures were performed but the data samples were randomly divided in terms of monitoring stations and days of the year, ensuring that training and validation samples were taken from different locations and on different days, thus minimising the impact of spatial and temporal autocorrelations. These procedures were followed to evaluate the accuracy of the model when predicting air pollutant concentrations at new (left-out) locations and times in the absence of ground-based measurements.[31–33]

Mortality burden assessment

$PM_{2.5}$ exposure can contribute to the disease burden, including specific diseases such as non-lung cancers, with increasing evidence emerging for the impact of wildfire-related exposure.[34,35] However, estimating the specific burdens of these diseases poses challenges due to the absence of disease-specific CRFs. Furthermore, the relative risk (RR) associated with $PM_{2.5}$ exposure can vary significantly across countries or regions. To minimise these limitations, we assessed the mortality burden attributed to $PM_{2.5}$ with use of a CRF for all-cause mortality derived from a large, representative cohort study conducted in the USA: the RR per 10 μg/m³ increase in long-term exposure to $PM_{2.5}$ is 1·12 (95% CI 1·08–1·15) for all-cause mortality.[27]

The mortality risk of black carbon to public health is reported to be more harmful (up to ten times higher) than $PM_{2.5}$,[3,4,25,26] but no universal CRF for black carbon is available. Considering the uncertainty in the toxicity of black carbon, we used a pooled estimate of CRFs derived from a comprehensive meta-analysis study: the RR per 1 μg/m³ increase in long-term exposure to black carbon is 1·06 (95% CI 1·04–1·09) for all-cause mortality.[3] The pooled RR estimate accounts for the uncertainty or heterogeneity across multiple studies and represents the average effect of a unit increase in black carbon on mortality. Lastly, the mortality burden associated with long-term exposure to $PM_{2.5}$ and black carbon was calculated by integrating their respective CRFs with variations in annual mean $PM_{2.5}$ and black carbon concentrations, population density, and baseline mortality rates (appendix pp 4–5).

Spatiotemporal analysis approach

Monthly $PM_{2.5}$ and black carbon data were calculated by averaging all the available daily satellite retrievals in a month, and then were used for calculating annual means, while ensuring that at least 20% of the available values at each monitoring station were used when comparing satellite retrievals with ground measurements. For mortality burden, we calculated the total accumulation of premature deaths, as well as premature deaths per 10 000 people in each 1-km² grid, to provide insights into areas where air pollution has the largest effects on human health. The temporal trends of $PM_{2.5}$ and black carbon, along with their associated mortality burdens, were determined using the least-squares linear regression method, and the statistical significance of the estimated trends was

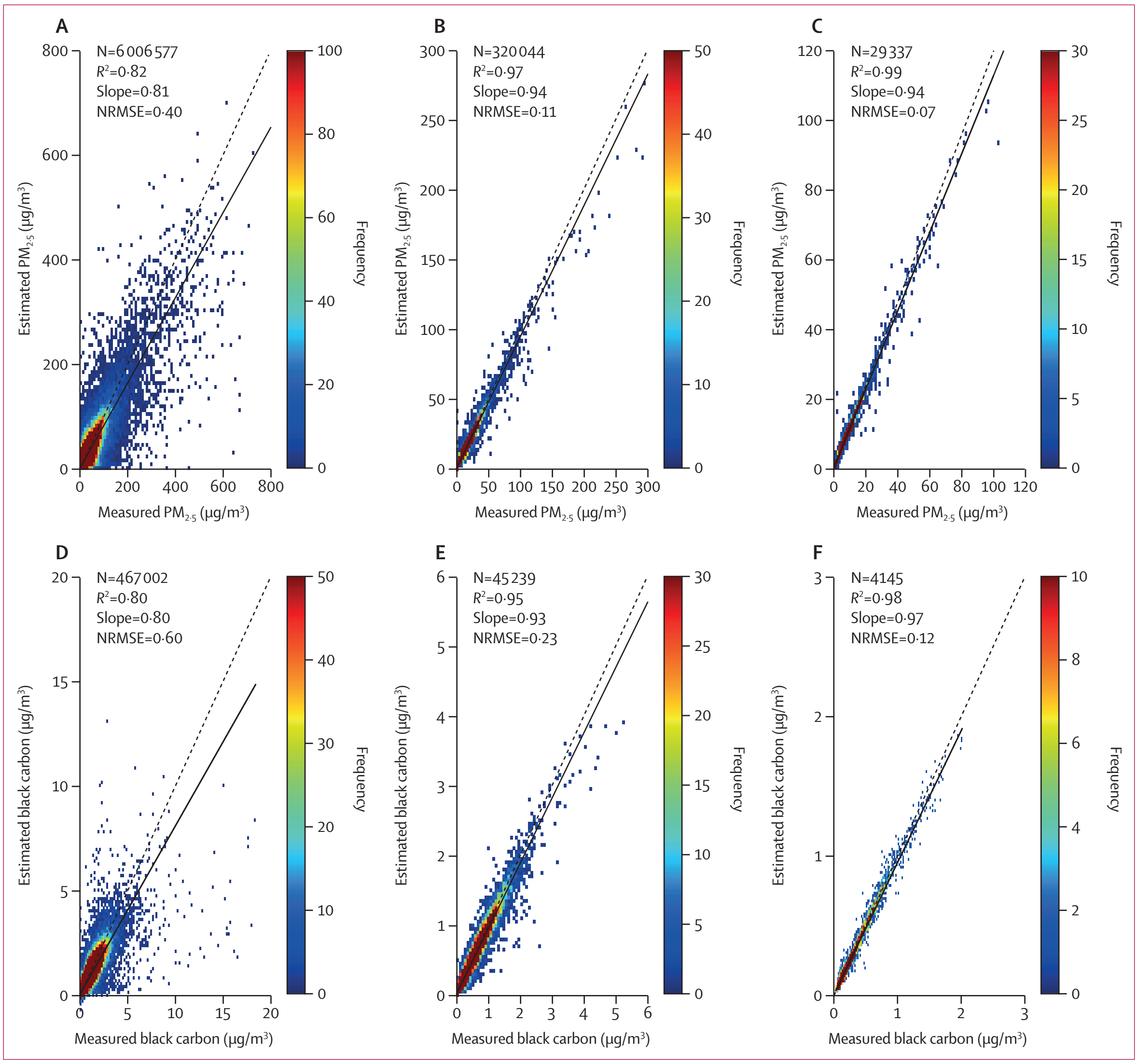

Figure 1: **Sample-based cross-validation of measured and estimated $PM_{2·5}$ and black carbon concentrations collected at all ground monitoring stations across the continental USA between 2000 and 2020**
Measured (x-axis) and estimated (y-axis) concentrations of $PM_{2·5}$ (A–C) and black carbon (D–F) are shown at daily (A, D), monthly (B, E), and annual (C, F) levels. The colours in the scatter plot represent the density of datapoints that fall within a given grid (frequency). The black dashed lines are 1:1 lines, and black solid lines are best-fit lines from linear regression between the retrievals and measurements. NRMSE=normalised root mean square error.

assessed via a two-sided test approach (significance levels of p<0·1, p<0·05, and p<0·01).[36] The breakpoint for long-term regional trends was identified with a combined approach that uses time-series analysis and sliding windows (appendix p 6). Furthermore, we present a two-step model by combining the Pauta criterion and the IQR method to remove outliers of potential wildfire-related $PM_{2·5}$ data from daily data (appendix p 7).

Role of the funding source

The funders of the study had no role in study design, data collection, data analysis, data interpretation, or writing of the report.

Results

Over the 21-year study period (2000 to 2020) in the USA, our model consistently produced highly accurate daily $PM_{2·5}$ and black carbon estimates with average sample-based cross-validation coefficient of determination (R^2) values of 0·82 and 0·80, respectively, and strong slopes of 0·81 and 0·80, showing average normalised root mean square error (NRMSE) values of 0·40 for $PM_{2·5}$ and 0·60 for black carbon, respectively (figure 1A, D). Additionally, we performed spatial cross-validation, in which monitoring stations were randomly left out. Our model performed well in predicting daily $PM_{2·5}$ (R^2=0·72, slope 0·73, and NRMSE 0·50) and black carbon (R^2=0·68, slope 0·72, and NRMSE 0·76) concentrations in regions

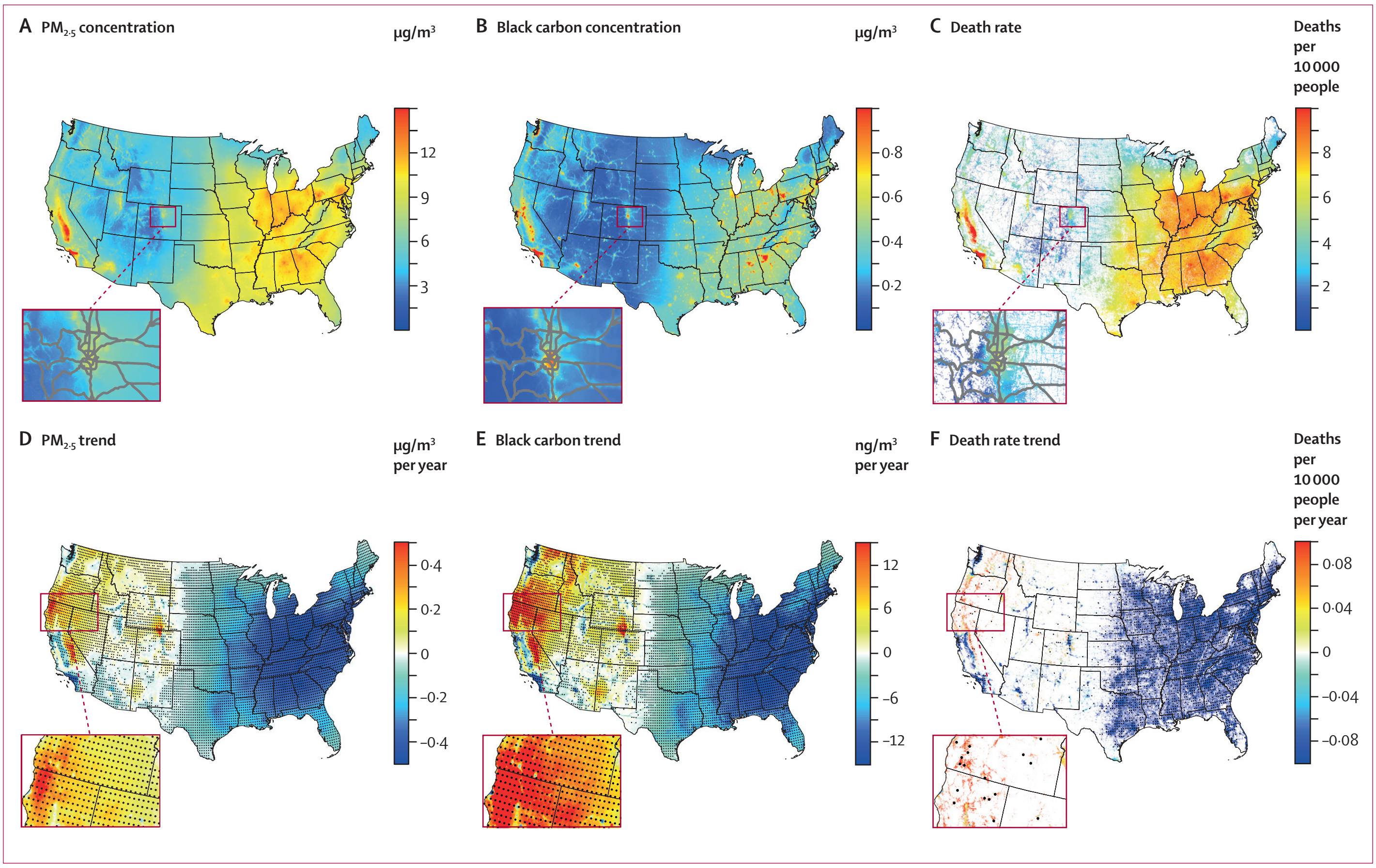

Figure 2: **Spatial distribution of $PM_{2·5}$ and black carbon concentrations and death rates between 2000 and 2020 across the USA**
Multiyear means are shown for $PM_{2·5}$ concentration (A), black carbon concentration (B), and $PM_{2·5}$-associated death rate (C) averaged over the period 2000 to 2020 at each 1-km² grid across the continental USA. Annual trends are shown for $PM_{2·5}$ concentration (D), black carbon concentration (E), and death rate (F) across the USA. $PM_{2·5}$ and black carbon trends were determined based on deseasonalised monthly anomalies. Each black dot in D–F denotes a 30-km² area where the trend is significant at the 95% (p<0·05) confidence level. The magnified sections show Denver, with grey lines representing major roads (A–C) and fire-prone areas encompassing California, Oregon, and Nevada (D–F).

without ground-based measurements (appendix p 11). Similarly, temporal cross-validation, in which randomly selected dates were left out, showed high reliability of our model in predicting daily $PM_{2·5}$ (R^2=0·74, slope 0·75, and NRMSE 0·48) and black carbon (R^2=0·78, slope 0·79, and NRMSE 0·63) concentrations on days without surface observations (appendix p 12).

Validation at the individual site scale showed that our model performs well in capturing daily surface $PM_{2·5}$ levels at most ground-based monitoring stations (eg, 74–86% of the monitors have sample-based, spatial-based, and temporal-based cross-validation R^2 values greater than 0·5, and 73–82% have NRMSE values less than 0·6; appendix p 13). The spatial pattern of surface black carbon retrievals was similar to that of $PM_{2·5}$, but the overall accuracy was poorer, with slightly smaller cross-validation R^2 values and larger NRMSE values when compared with ground-based observations, showing cross-validation R^2 of greater than 0·5 at 77–87% of the sites at the overall, spatial, and temporal scales, and NRMSE of less than 0·8 at approximately 74–82% of sites. Furthermore, our model showed reliability at the regional scale with varying sources of pollution (appendix p 26); for example, in the eastern USA, where anthropogenic emissions dominate, cross-validation R^2 was 0·75–0·85 for $PM_{2·5}$ and 0·65–0·81 for black carbon, and in the western USA, where natural sources are dominated by frequent wildfires, cross-validation R^2 was 0·70–0·81 and 0·67–0·77, respectively. These statistical agreements are consistently improved upon in the comparisons of monthly (cross-validation R^2 of 0·76–0·97 for $PM_{2·5}$ and 0·73–0·95 for black carbon) and annual (0·76–0·99 and 0·77–0·98, respectively) $PM_{2·5}$ and black carbon averages at both the national and regional scales (figure 1, appendix p 26).

The mean spatiotemporal distribution and the trends of $PM_{2·5}$, black carbon, and mortality burden in the USA during 2000 to 2020 are shown in figure 2 (maps for each year are provided in the appendix, pp 14–16). Annual $PM_{2·5}$ and black carbon concentrations had similar spatial distributions (figure 2A, B). Mean concentrations were around two times higher in the eastern USA

($PM_{2·5}$ 9·5 μg/m³ [SD 1·8] and black carbon 0·45 μg/m³ [0·14]) than in the western USA (5·1 μg/m³ [1·7] and 0·23 μg/m³ [0·13]), and 1·2–1·5 times higher than in the central USA (8·0 μg/m³ [2·0] and 0·31 μg/m³ [0·12]), reflecting the population distribution and anthropogenic emissions. At the individual state level, the highest persistent pollution levels were found in some areas in California, probably reflecting the wildfire smoke patterns and local sources of dust, especially in the Central Valley and nearby areas. Indeed, both $PM_{2·5}$ and black carbon increased by 49–52% in the hot seasons (summer and autumn) when compared with the cold seasons (winter and spring) in the western USA (appendix pp 17–18). The mortality burden per capita closely aligned with the distribution of air pollution, with higher death rates in the densely populated eastern region and the western California region, particularly in major cities such as Los Angeles (figure 2C). The cumulative number of premature deaths associated with exposure to $PM_{2·5}$ pollution in the continental USA was estimated to be approximately 3·2 million (95% CI 2·2–3·9) during the 21-year study period. Additionally, our fine-resolution data allowed us to study air pollution and its impact on public health at each 1-km² grid (see magnified sections in figure 2). Large differences were observed in the pollution levels of urban and rural regions. In particular, high black carbon concentrations were observed along highways due to traffic-related emissions (largely from diesel trucks). Contrasting distributions in the mortality burden in large cities and their surrounding areas could also be seen. These results highlight the advantages of high-resolution air pollution data.

Temporally, the amounts of $PM_{2·5}$ and black carbon in 2000 to 2020 showed a significant decreasing trend in the eastern USA ($p<0·05$) but remained quite stable in the central USA (figure 2D, E). However, the trends varied considerably in direction (positive or negative) and magnitude across the western USA. Significant decreasing trends were observed in city clusters located in the southwest (eg, Los Angeles) and northwest (eg, Seattle) corners ($p<0·05$). By contrast, significant increasing trends were identified in central inter-mountainous and northwest areas ($p<0·05$; outlined by the red rectangles). Additionally, declining trends throughout the USA were found in winter and spring, whereas in summer and autumn, trends increased in the western USA and decreased in the eastern USA (appendix pp 17–18). These findings strongly suggest the potentially growing impact of wildfires on surface $PM_{2·5}$ and black carbon in the western USA.[6,17] Overall, the death rates associated with $PM_{2·5}$ exposure showed a significant reduction in the majority of densely populated areas across the USA in the past two decades ($p<0·05$), with particularly notable health benefits observed in the eastern USA (figure 2F). However, mortality rates increased markedly in specific regions, including eastern and northern California, as well as southwestern Oregon (outlined by the red rectangle), where elevated concentrations of $PM_{2·5}$ and black carbon are evident.

The time series of annual mean $PM_{2·5}$, black carbon, and total premature deaths during the years 2000 to 2020 were analysed for the continental USA, eastern USA, and western USA (figure 3). At the national level, $PM_{2·5}$ concentrations decreased by around 22% and black carbon concentrations decreased by around 11% over the entire period, with the highest levels in 2000 and the lowest levels in 2019. The decreasing trends were more prominent in the first decade and gradually slowed down in the second decade (figure 3A, E). The eastern USA showed larger and more consistent declining trends of around 50% for $PM_{2·5}$ concentration and 43% for black carbon concentration, with small fluctuations (figure 3B, F). By contrast, the western USA showed almost no notable trends throughout the entire period, primarily due to large interannual fluctuations (figure 3C, G). Of note, downward trends (slope $k<0$) were observed for $PM_{2·5}$ and black carbon concentrations before 2010, but these trends reversed (slope $k>0$) in subsequent years.

Similarly, the annual number of total premature deaths due to $PM_{2·5}$ pollution exposure across the continental USA significantly decreased from 198 000 (95% CI 137 000–240 000) in 2000 to 141 000 (97 000–172 000) in 2010 ($p<0·01$). The rate of decline then slowed by a factor of three until 2020, with small fluctuations (figure 3I). Overall, the decline rate was 4200 premature deaths per year (2960–5050) over the past two decades. In the eastern USA, the rate of deaths decreased continuously and significantly, with a decrease of 2650 deaths per year (1860–3190; $p<0·01$; figure 3J). By contrast, in the western USA, the annual death burden steadily decreased by 1280 deaths per year (910–1530; $p<0·01$) until 2010 (burden of 28 000 deaths [95% CI 19 000–34 000]), after which there was an opposite trend, with an increase of 670 deaths per year (460–810), with the peak burden in 2020 (burden of 42 000 deaths [29 000–51 000]; figure 3K). These findings probably indicate the effects of escalating fire emissions since 2010, which could contribute to a national slowdown and a regional reversal in the western USA of the decreasing trends of air quality and mortality burden.[18,32]

The fine spatial resolution used for mapping surface $PM_{2·5}$ and black carbon can be used to investigate the impact of the population movement at the neighbourhood scale on public health. A sensitivity analysis showed that downscaling the spatial resolution by five times or ten times could yield an overall underestimation of around 3% and 4%, respectively, in the total number of deaths attributed to $PM_{2·5}$ over the 21-year period in the continental USA. Corresponding values are slightly more pronounced or doubled for black carbon. However, despite these differences, the temporal trends of the disease burden showed a high level of agreement, with small absolute differences ranging from 20 to 80 premature deaths (appendix p 19).

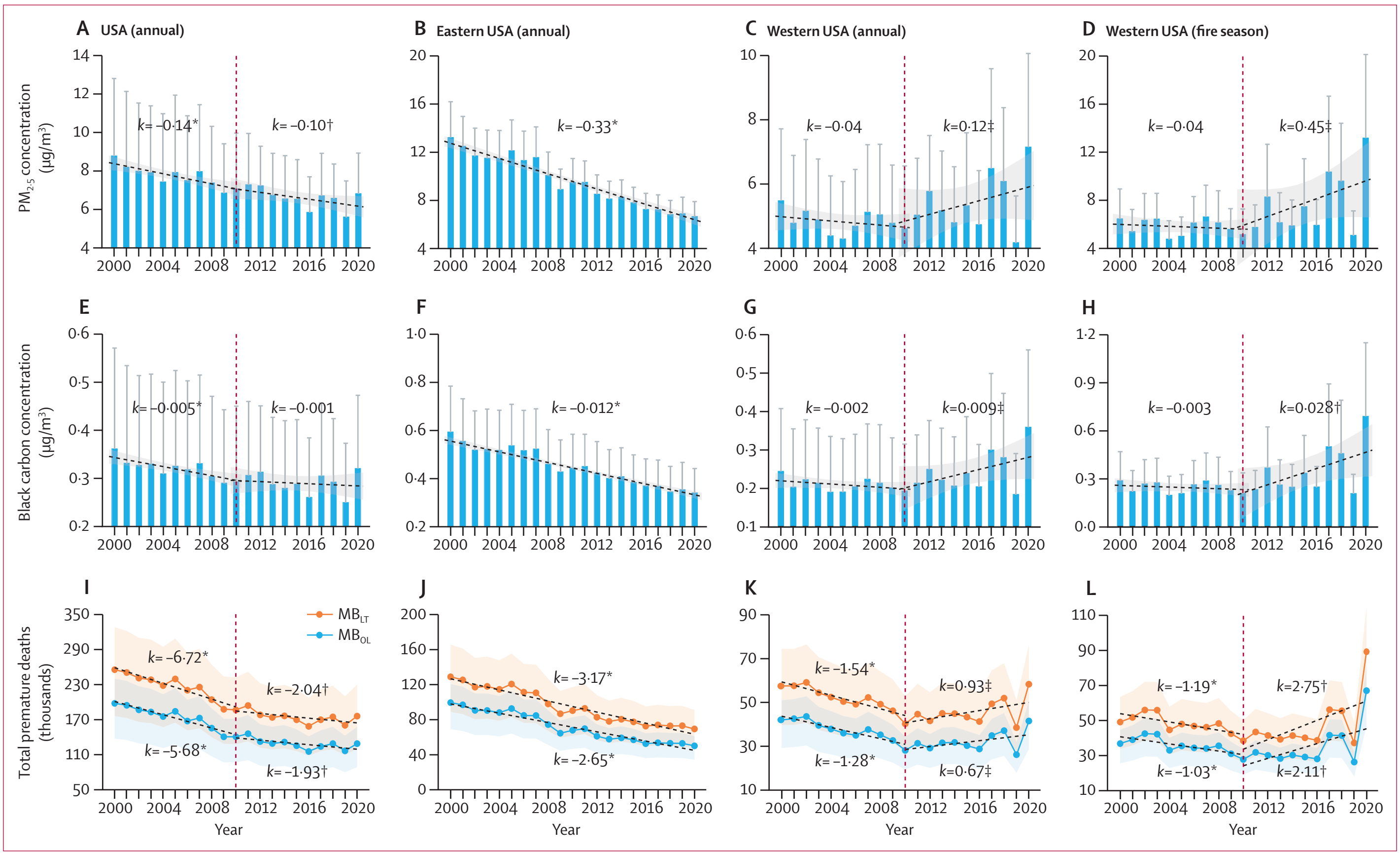

***Figure 3:* Time series of annual mean $PM_{2·5}$ and black carbon concentrations and total premature deaths associated with the total $PM_{2·5}$ pollution in the years 2000–2020 in the continental USA, eastern USA, and western USA**

Annual mean $PM_{2·5}$ concentration (A–C) and black carbon concentrations (E–G) and total premature deaths (I–K) are shown for the continental USA, eastern USA, and western USA, with corresponding results during the fire season (July to October) for the western USA (D, H, L). The blue bars in panels A–D represent the mean concentrations of air pollutants, and the black lines represent SDs. Black dashed lines represent the regressions fitted to the blue bars. Orange and blue lines in I–L denote the estimates of $PM_{2·5}$-associated mortality burden with and without considering the larger toxicity of black carbon (MB_{LT} and MB_{OL}), respectively. Shaded areas represent confidence intervals ($p<0·05$) for the regression fits of air pollution (A–H) and estimated premature deaths (I–L). The regressed slope (*k*) values are also given. Vertical red dashed lines represent the year of the detected breakpoint. *Significant at the 99% ($p<0·01$) confidence level. †Significant at the 95% ($p<0·05$) confidence level. ‡Significant at the 90% ($p<0·1$) confidence level.

Daily gapless estimates of $PM_{2·5}$ and black carbon offer a distinct advantage over monthly or annual estimates in capturing the day-to-day variations of extreme pollution events, particularly those due to sporadic wildfires. They provide valuable insights into the life cycle of smoke particles, including their formation, their local spread and long-range transport to downstream regions, and eventual disappearance from the atmosphere (appendix pp 20–21). To further explore the impact of wildfires on air quality, we analysed the long-term spatiotemporal changes of $PM_{2·5}$ and black carbon, specifically focusing on the black carbon-to-$PM_{2·5}$ ratio, during the fire season (defined as July–October) from 2000 to 2020. The spatial patterns of fire season $PM_{2·5}$ and black carbon showed high similarity to the annual average, but the pollution levels were approximately 1·3 times higher in the western USA (figure 4A, B). High black carbon-to-$PM_{2·5}$ ratio values (>10%) were predominantly found in major metropolitan areas, such as Seattle, San Francisco, Denver, Atlanta, and New York City, among others, consistent with the expected patterns based on the black carbon contribution to $PM_{2·5}$ anthropogenic emissions (figure 4C). Although it varies, the black carbon mass fraction in fire emissions of $PM_{2·5}$ typically remains below 6%.

The decreasing trends of fire-season $PM_{2·5}$ and black carbon in the eastern USA were consistent over the past two decades, whereas notable changes were observed in the western USA. During the first decade (2000 to 2010), there was a significant overall reduction across the majority of western USA regions, particularly in the central and southern areas (figure 4G, H). However, a reversal occurred in the second decade (2010 to 2020), with most western USA regions experiencing a substantial increase in pollution levels (figure 4J, K), surpassing three to four times the annual average ($p<0·1$ for $PM_{2·5}$ and $p<0·05$ for black carbon; figure 3D, H). This reversal could be largely attributed to the increasing occurrence of severe wildfires in recent years,[17] resulting in a deterioration in air quality in the downwind areas. This attribution, although ultimately requiring process-level

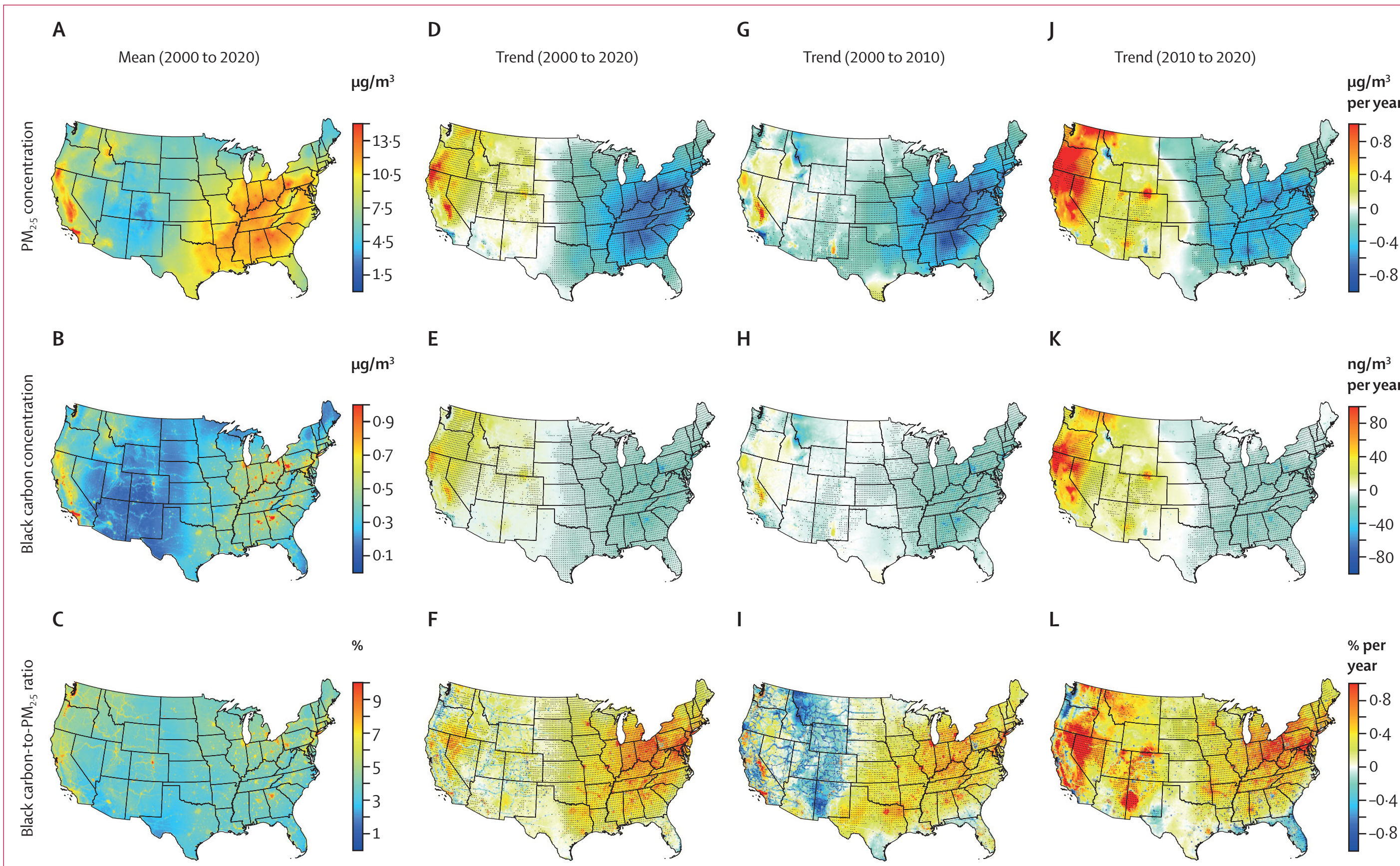

Figure 4: **Spatial distribution of $PM_{2·5}$ concentration, black carbon concentration, and black carbon-to-$PM_{2·5}$ ratio during the fire season in 2000 to 2020, 2000 to 2010, and 2010 to 2020**
Multi-year mean $PM_{2·5}$ concentration (A), black carbon concentration (B), and black carbon-to-$PM_{2·5}$ ratio (C) during the fire season (July–October) are shown, averaged over the period 2000 to 2020. Temporal trends of fire-season $PM_{2·5}$ concentration, black carbon concentration, and black carbon-to-$PM_{2·5}$ ratio are shown for 2000 to 2020 (D–F), 2000 to 2010 (G–I), and 2010 to 2020 (J–L), at each 1-km² grid across the continental USA. Each black dot in D–L denotes a 30-km² area where the trend is significant at the 95% ($p<0·05$) confidence level.

modelling, was further evidenced by the strong correlations ($R=0·89$ [$p<0·01$] for $PM_{2·5}$ and $R=0·88$ [$p<0·01$] for black carbon) and consistent changes between $PM_{2·5}$ and black carbon and their emissions from wildfires over the past two decades, particularly since 2010 at a statistically significant level ($p<0·1$; appendix p 22). Indeed, the time series of monthly $PM_{2·5}$ anomalies[36] in the western USA showed large fluctuations in certain years (eg, 2017, 2018, and 2020) associated with large wildfires (appendix p 23). However, when excluding the daily observations affected by intense wildfire events from the monthly averages, the original upward trend of $PM_{2·5}$ pollution is replaced by a significant downward trend (slope –0·06 [$p<0·01$]) in the western USA, particularly in California (slope –0·08 [$p<0·01$]).

Significant increasing trends ($p<0·05$) in black carbon-to-$PM_{2·5}$ ratio were observed in fire-prone areas of the western USA between 2000 and 2020, particularly in California, Oregon, Nevada, and Arizona (figure 4F). These trends have become more prominent in the past decade (figure 4L), aligning closely with the significant increase in fire-season $PM_{2·5}$ and black carbon concentrations (figure 4J, K), probably attributable to emissions from local wildfires or the transport of smoke particles originating from nearby wildfires. However, no significant trend was found in the southern parts of the US Gulf states in recent years, which could be a result of fire emissions from prescribed and agricultural burns.[37] By contrast, during the first decade, the black carbon-to-$PM_{2·5}$ ratio in the western USA decreased considerably, especially in the central region along highways (figure 4I), probably attributable to the reduction in traffic-related emissions. Nonetheless, black carbon-to-$PM_{2·5}$ ratio values significantly increased throughout the USA, with an average annual growth rate of 2·4% ($p<0·01$). This increase was primarily originated from the sustained and significant increase in black carbon-to-$PM_{2·5}$ ratio in the eastern USA (slope 4·7% [$p<0·01$]), probably reflecting a faster decrease in other $PM_{2·5}$ components, such as sulphate and nitrate, as a result of significant reductions in emissions of nitrogen and sulphur oxides.[38] This finding highlights the importance of the combined effects of fire emissions and long-term reductions in anthropogenic emissions in regulating ambient $PM_{2·5}$ concentrations.

The toxicity of black carbon to human health remains uncertain in the literature. Many studies conclude

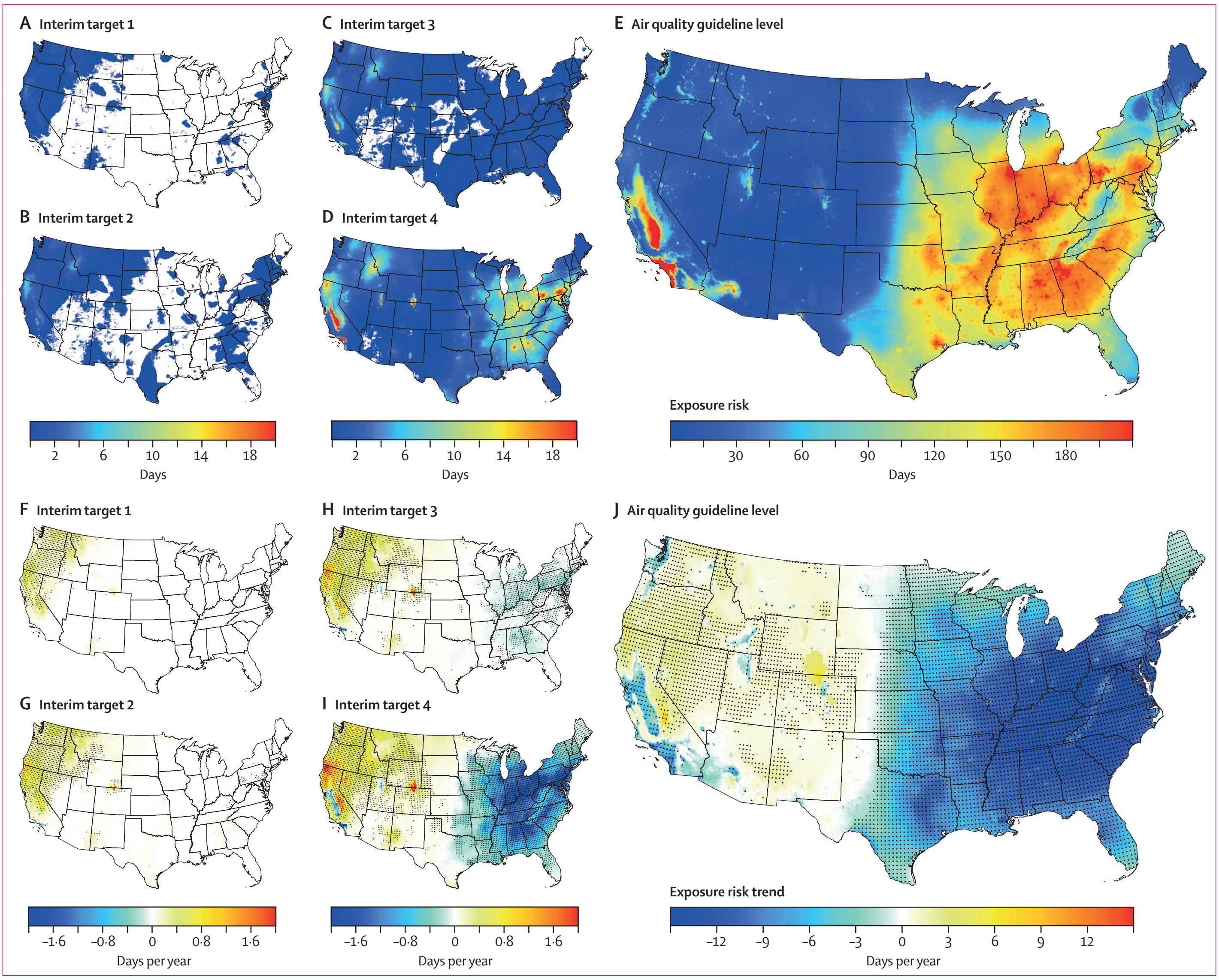

Figure 5: **Spatial distribution of days exceeding WHO-recommended short-term air quality standards between 2000 and 2020 across the continental USA**

Multi-year mean (A–E; unit days) and temporal trends (F–J; unit days per year) of days exceeding WHO-recommended short-term air quality standards, at interim target 1 (A, F; daily $PM_{2·5}$ of 75 μg/m³), interim target 2 (B, G; 50 μg/m³), interim target 3 (C, H; 37·5 μg/m³), interim target 4 (D, I; 25 μg/m³), and air quality guideline level (E, J; 15 μg/m³), during the period 2000 to 2020 at each 1-km² grid across the continental USA. Each black dot in F–J denotes a 30-km² area where the trend is significant at the 95% (p<0·05) confidence level.

that black carbon poses a greater relative risk and, consequently, a larger impact on mortality than other $PM_{2·5}$ components,[3,4,25,26] but some others suggest low confidence.[39] In a sensitivity study, we compared estimates of premature deaths under two assumptions: black carbon is no more toxic than other components and has a similar impact on health (blue lines in figure 3I–L); and black carbon has a greater toxicity (orange lines in figure 3I–L). We found that the mortality burden attributed to total $PM_{2·5}$ could increase by 28–38% when considering the greater toxicity of black carbon, and their trends could have accelerated much more in recent years. This acceleration was particularly distinct in the western USA (figure 3K), resulting in extra loss of life (due to the higher toxicity) at an increased rate of 260 deaths per year (95% CI 180–400) since 2010. Furthermore, when assuming fire-season pollution as the baseline level, the greater toxicity of black carbon could triple the increasing trend of mortality burden (figure 3L). These findings emphasise the increasing significance of wildfire-generated black carbon in the analysis of mortality burden. Additionally, the sensitivity analysis underscores the need for future research efforts in refining our understanding of the CRF for black carbon, enabling more informed public health interventions.

Daily gapless surface $PM_{2·5}$ mapping provides an opportunity to assess the risk of population exposure to short-term $PM_{2·5}$ pollution in relation to the updated WHO air quality guidelines (2021).[40] Over the 21-year average, severe daily exposure risk (daily $PM_{2·5}$ >75 μg/m³) occurred (at least once) only in the northwest region and a few eastern areas like New York City (figure 5A). However, both the area and frequency of daily $PM_{2·5}$ exposure risk occurrence could be considerably increased as more stringent standards for daily air quality are applied (figure 5B–D), following the WHO interim targets (daily $PM_{2·5}$ of 50 μg/m³ for interim target 2, 37·5 μg/m³ for interim target 3, and 25 μg/m³ for interim target 4). If the most stringent recommended air quality guideline level (daily $PM_{2·5}$ of 15 μg/m³) were enacted (figure 5E), 100% of populated areas would have been defined as having unhealthy air exposure risk for at least one day during the period 2000 to 2020, 99% for at least 7 days, and 86% for at least 30 days. The daily exposure risk was particularly pronounced in densely populated regions of the eastern USA and California (eg, Central Valley and Los Angeles), where the period exceeded 7 months during 2000 to 2020.

Emissions from intensifying wildfires could have played a crucial role in the substantial increase of daily exposure risk in the northwestern USA and California between 2000 and 2020 in terms of the number of high-pollution days (figure 5F, G). When more stringent air quality standards were adopted, this increasing trend in daily risk exposure due to fire emissions was more evident in the western USA (figure 5H, I), whereas the eastern USA showed an opposite (decreasing) trend. At the air quality guideline level (figure 5J), the spatial contrast in the direction of the trends was most pronounced, indicating a significant increase in polluted days in the western parts and a decrease in the eastern parts of the USA. Of note, an increase in polluted days in the midwestern USA was also observed, possibly due to the long-range wildfire smoke-particle transport from the western USA (figure 5H–J). We found that the national average number of polluted days has continuously diminished since 2000, and by around 2010, daily $PM_{2·5}$ levels successfully complied with the air quality guideline level. This cleaner air trend continued in the eastern USA during the period 2010 to 2020, but in the western USA, this trend was reversed, followed by a decrease in recent years (appendix p 24). Of note, a significant surge in high-pollution days (daily $PM_{2·5}$ >50 μg/m³) occurred in 2020, pinpointed from the daily time series of $PM_{2·5}$ data.

Discussion

Previous studies estimating $PM_{2·5}$ in the USA (appendix p 27) have often focused on individual years or shorter periods than in the current study (<15 years). Few have considered the role of widespread fires during recent years in the long-term US air quality trend.[7–11,13–15] In addition, the overall accuracies of their daily (cross-validation R^2 0·65–0·80)[9,10,13] or monthly (0·76–0·86)[7,8,11] $PM_{2·5}$ estimates could be limited by traditional statistical regression and machine-learning models that offer insufficient data-mining capabilities. In particular, most studies have used $PM_{2·5}$ observations from only the urban-dominated EPA Air Quality System network,[7–10,12,14,15] and few studies have considered the IMPROVE network in rural areas.[11,13] Almost no studies have explicitly considered smoke contributions to highly polluted conditions.[13] These omissions would inevitably introduce considerable uncertainties in the predictions, particularly in the western, intermountain, and central USA, where rural areas dominate and are susceptible to widespread smoke particles from fires. These uncertainties could explain why several studies with less extreme $PM_{2·5}$ observations or more regional focuses have slightly higher cross-validation R^2 values than ours.[12,14,15] Unlike $PM_{2·5}$ estimates, black carbon estimates have been made by only a handful of studies using chemical transport models, statistics, and the early generation of machine-learning methods. Although progress has been made, these studies have limitations in terms of available periods of data, overall accuracy (cross-validation R^2 0·74–0·75), and spatial (≥12 km²) or temporal (monthly or annual averages) resolutions in the USA.[11,16,41] By contrast, we have developed a robust model that uses advanced deep learning and integrates $PM_{2·5}$ and black carbon data from more networks, covering a wide range of conditions from often clean and rural areas in the central USA to areas severely polluted by wildfires. We have reconstructed, to date, the longest 1-km-resolution daily seamless $PM_{2·5}$ and black carbon data records (2000 to 2020) across the continental USA, with similar or superior accuracy compared with previous studies.

Using this new dataset, we have conducted an assessment of the long-term changes in mortality burden attributed to fine particle pollution (especially black carbon) during the past two decades in the USA. Most previous studies have primarily focused on the specific-cause mortality of $PM_{2·5}$ for different populations,[42–44] the impact and contribution of smoke-$PM_{2·5}$ on the death burden,[6,19] and the health benefits of reducing anthropogenic emissions for limited periods,[45,46] using black carbon data for individual years or specific cities obtained from either model simulations or ground-based measurements.[16,47] Our main focus has been to identify any disruptions in the otherwise long-term decreasing trends that could be attributed to emissions from wildfires. Subsequent analysis of the health impact highlights the importance of further quantifying the toxicity of black carbon, a key component of wildfires. Our study differs from previous studies in that we have provided a publicly available, long-term dataset of $PM_{2·5}$ and black carbon concentrations across the continental USA, specifically including data from rural areas; and analysis of this dataset shows the increasing role of fires in shaping long-term trends in air quality and the associated public health burden, including the trend of particle toxicity in terms of the black carbon-to-$PM_{2·5}$ ratio. Our study emphasises the need for implementing effective policies, such as wildfire prevention and management, forest restoration, and the generation of new revenue, to reduce the risk of fires. These measures will also contribute considerably to future benefits, enhancing both air quality and public health.

Our daily $PM_{2·5}$ retrievals under high-pollution conditions tend to be underestimated, which is a common problem among all models and is caused mainly by large uncertainties in AOD observations and the smaller number of data samples under highly polluted conditions.[48] The black carbon retrievals are less accurate than those of $PM_{2·5}$, mainly due to their lower concentrations and more varying relationship with the total mass of $PM_{2·5}$. The diminished accuracy is also affected by a smaller volume of black carbon observations and the relatively large uncertainty of black carbon measurements (100% compared with 10% for $PM_{2·5}$).[49] Although there are associations between frequent wildfire emissions and the subsequent deterioration of air quality and the potential increase in associated health burden in the western USA, these observation-based findings are primarily statistical. Both observation and modelling studies are required to gain deeper insights into the processes and understand the causal relationships between fire emissions and changes in surface-level air pollutants, as well as specific health effect studies of acute exposure to fresh and aged smoke.

Contributors

JWe and JWa conceived and designed the study. JWe developed the methods, performed the research, and wrote the initial draft of this manuscript. JWe, JWa, and ZL accessed and verified the data. Other authors provided the satellite and ground data, contributed to the interpretation of the results, and helped revise the manuscript. All authors had full access to all the data in the study and had final responsibility for the decision to submit for publication.

Declaration of interests

We declare no competing interests.

Data sharing

Web links to access all study data are included in the appendix (p 8). The generated 1-km $PM_{2·5}$ and black carbon datasets and codes are publicly available at https://doi.org/10.5281/zenodo.7884640 and https://doi.org/10.5281/zenodo.7971584, respectively.

Acknowledgments

This work is supported in part by the National Aeronautics and Space Administration (NASA) Applied Science programme (80NSSC19K0191, 80NSSC21K0428, 80NSSC21K1980, and 80NSSC20K0131), the NASA MODIS maintenance programme (to AL), the NASA MAIA satellite mission programme (managed by Jet Propulsion Laboratory with contract number 1583456 to the University of Iowa), the NASA GMO core fund (to AdS), the National Oceanic and Atmospheric Administration (NOAA) GEO-XO project (NA21OAR4310249 and NA21OAR4310250), the NOAA Atmospheric Chemistry, Carbon Cycle, and Climate (AC4) programme (NA19OAR4310178), and the NOAA Educational Partnership Program with Minority Serving Institutions (NA16SEC4810006 to CI). The contributions of DD were carried out at the Jet Propulsion Laboratory, California Institute of Technology, under contract with NASA. The scientific results and conclusions, as well as any views or opinions expressed herein, are those of the authors and do not necessarily reflect those of NOAA or the US Department of Commerce.

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