

Spatial-temporal patterns of anthropogenic and biomass burning contributions on air pollution and mortality burden changes in India from 1995 to 2014

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Abstract. Anthropogenic (ANTHRO) emissions and biomass burning (BB) are major contributors to ambient air pollution, with the latter playing a particularly dominant role in non-urban regions. India has experienced a dramatic deterioration in air quality over the past few decades, but no systematic assessment has been conducted to investigate the individual contributions of ANTHRO and BB emissions changes over the long term in India, particularly in non-urban areas. In this study, we conduct a comprehensive analysis of the long-term trends of particulate matter with aerodynamic diameters $< 2.5 \mu\text{m}$ ($\text{PM}_{2.5}$) and ozone (O_3) in India and their mortality burden changes from 1995 to 2014, using a state-of-the-art high-resolution global chemical transport model (CAM-chem). Our simulations reveal a substantial nationwide increase in annual mean $\text{PM}_{2.5}$ ($6.71 \mu\text{g m}^{-3} \text{decade}^{-1}$) and O_3 ($7.08 \text{ ppbv decade}^{-1}$), with the Indo-Gangetic Plain (IGP) and eastern central India serving as hotspots for $\text{PM}_{2.5}$ and O_3 trend changes, respectively. Noteworthy substantial O_3 decreases were observed in the northern IGP, potentially linked to NO titration due to a surge in NO_x emissions. Sensitivity analyses highlight ANTHRO emissions as primary contributors to rising $\text{PM}_{2.5}$ and O_3 , while BB played a prominent role in winter and spring. In years with high BB activity, the contributions from BB emissions on both $\text{PM}_{2.5}$ and O_3 changes were comparable with or even exceeding ANTHRO emissions in specific areas. We further estimate that the elevated air pollutants were associated with increased premature mortality attributable to $\text{PM}_{2.5}$ and O_3 , leading to 97.83 K and 73.91 K per decade. Despite a decrease in premature mortality per capita in the IGP region, the increased population offset its effectiveness.

1 Introduction

Air pollution is among the most detrimental environmental factors to human health. According to the World Health Organization (WHO) database, 99% of the global population lives in areas where air quality surpasses WHO guideline limits

(WHO: Air Pollution, World Health Organization, available at: https://www.who.int/health-topics/air-pollution#tab=tab_1, last access: 21 September 2024). The two most concerned pollutants, particulate matter with aerodynamic diameters $< 2.5 \mu\text{m}$ ($\text{PM}_{2.5}$) and ozone (O_3), can cause significant damage to the human heart and lungs (Hoek et al., 2013; Hystad et al., 2013; Villeneuve et al., 2015), potentially leading to premature death when exposed over extended periods (Dedoussi et al., 2020; Fuller et al., 2022). The latest Global Burden Disease (GBD2019) study, a comprehensive research initiative that quantifies health loss due to diseases, injuries, and risk factors worldwide, estimated that exposure to air pollution, including both household and ambient pollution, led to 6.7 million premature deaths (95% confidence interval [CI], 5.9 to 7.5 million) worldwide in 2019 (GBD 2019 Risk Factors Collaborators., 2020). Thus, the urgency of dealing with air pollution has become one of the most pressing global challenges.

It is well-known that surface air pollution is usually unequally distributed in space, with higher levels in developing countries than in developed countries (GBD 2015 Risk Factors Collaborators., 2016). For example, India was ranked as the most polluted country in the world in 2021, with 63 of the world's 100 most polluted cities (IQAir: 2021 World Air Quality Report, available at: https://lib.icimod.org/record/35767/files/HimalDoc2022_2021WorldAirQualityReport.pdf?type=primary, last access: 21 September 2024). Previous modelling studies indicated that districts exceeding India's annual ambient standard of $40 \mu\text{g m}^{-3}$ rose from 200 to 385 out of 640 from 1998 to 2020 (Guttikunda and Ka, 2022). The GBD2019 study estimated that premature deaths attributed to ambient $\text{PM}_{2.5}$ and O_3 pollution accounted for 10.4% (8.4-12.3) and 1.8% (0.9-2.7) of the total deaths in India in 2019, respectively, and the death rate per 100,000 people increased by 115.3% (28.3-344.4) and 139.2% (96.5-195.8) from 1990 to 2019, respectively (Pandey et al., 2021). However, the GBD2019 study did not separate the air quality changes due to various contribution factors, such as anthropogenic (ANTHRO) and biomass burning (BB). Meanwhile, the elevated chemical reaction rates in India, driven by intense sunlight and warm temperatures, create conditions conducive to ozone formation. Additionally, strong convection enhances the transport of ozone and its precursors, such as NO_y , to higher altitudes, where the prolonged ozone lifetime promotes accumulation. This phenomenon positions India as a hotspot for ozone pollution, significantly impacting air quality in downwind regions (Zhang et al., 2016, 2021a).

As seen from the Community Emissions Data System (CEDS) inventory (Hoesly et al., 2018), the increasing trends of ANTHRO emissions of major air pollutants, such as nitrogen oxides (NO_x), carbon monoxide (CO) and non-methane volatile organic compound (NMVOC), are significantly higher in India than those in other regions (Wang et al., 2022). Meanwhile, crop yields in India have significantly enhanced since the mid-1960s after the Green Revolution, contributing to increased BB emissions (Huang et al., 2022). The study showed that from 1950–51 to 2017–18, the crop residue burning in India increased from 18 million tonnes to 116 million tonnes in terms of total biomass burned (Venkatramanan et al., 2021). The frequency and intensity of forest fires in India have also increased in recent years due to persistent warmer temperatures and climate extremes (Vadrevu et al., 2019; Jain et al., 2021). These in turn could pose significant threats to ambient air quality and human health because large amounts of compounds are emitted into the atmosphere, namely carbon dioxide

65 (CO₂), NO_x, particulate matter (PM), and other chemical species (Crutzen and Andreae, 1990; Carvalho et al., 2011; Lan et al., 2022; Miranda et al., 2005). Previous studies have utilized observational and satellite data to assess the impacts of ANTHRO and BB sources on air quality trends in some Indian cities (Gurjar et al., 2016; Vohra et al., 2022). Additionally, model simulations have been employed to analyse source contributions to air pollution (Conibear et al., 2018a, b). However, there remains a lack of comprehensive assessments regarding the impacts of long-term ANTHRO and BB emissions changes on air quality, particularly in non-urban areas.

In this study, we aim to improve our understanding of the spatial-temporal distribution of major air pollutants, mainly surface PM_{2.5} and O₃, and the related mortality burden in India from 1995-2014 using a state-of-the-art global chemistry transport model. In addition, the individual contributions of changes in ANTHRO and BB emissions were further separated to better understand the causes of worsening air quality and escalating health risks in India. The selected period encompasses a dynamic phase of rapid changes in both ANTHRO and BB activities in India, thereby providing an ideal context for investigating their respective contributions to air pollution.

2 Methods

2.1 CAM-chem model configuration

We simulate surface PM_{2.5} and O₃ concentrations over India between 1995 and 2014 using the global chemistry model CAM-chem, which is based on version 6 of the Community Atmosphere Model (CAM6), the atmospheric component of the Community Earth System Model (CESM2), as detailed by Danabasoglu et al. (2020) and Emmons et al. (2020). Following Emmons et al. (2020), the original model is run at 1.25° (longitude) × 0.9° (latitude) horizontal resolution with 32 vertical levels reaching ~45 km. We configure the Model of Ozone and Related Chemical Tracers Tropospheric and Stratospheric (MOZART-TS1) chemistry mechanism with various complexity choices for tropospheric and stratospheric chemistry (Emmons et al., 2020). The aerosol module adopts the four-mode version of the Modal Aerosol Model (MAM4), including sulfate, black carbon, primary organic matter, secondary organic aerosols, sea salt, and mineral dust. The first level of the model outputs is considered the surface level, and all the model outputs are then regridded to a finer resolution of 0.5° × 0.5° to match the grid-cell population and baseline mortality rates datasets in performing the health impact assessment.

Global historical ANTHRO emissions are adopted from CEDS (version 2017-05-18), which provides monthly emissions of ANTHRO aerosol and precursor compounds at 0.5° × 0.5° from 1750 to 2014 and were used in the Coupled Model Intercomparison Project Phase 6 (CMIP6) experiments (Emmons et al., 2020; Hoesly et al., 2018). The ANTHRO emissions include eight sectors: agriculture; energy; industrial; transportation; residential, commercial, other; solvents production and application; waste and international shipping (Hoesly et al., 2018). The air pollutants from the CEDS inventory, especially the NMVOC, are then re-specified to match the chemical species in the latest CESM2 model, following the steps introduced by Emmons et al. (2020). Interpolation of the emission inventory from its original resolution

($0.5^{\circ} \times 0.5^{\circ}$) to the target model resolution ($0.9^{\circ} \times 1.25^{\circ}$) before being input into the model. Global historical BB emissions are sourced from van Marle et al. (2017) at monthly temporal resolution and 0.5° native resolution, with all emissions occurring at the surface. Additionally, the biogenic emissions are calculated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN v2.1). More emissions used are described in Emmons et al. (2020).

100 **2.2 Numerical experiments designs**

As described above, the standard (BASE) simulation is driven by year-to-year variability of ANTHRO and BB emissions from 1995 to 2014. To separate the contributions from these two emission sources, we then conduct two sensitivity simulations in which ANTHRO emissions (FixAN) and BB emissions (FixBB) are fixed at 1995 levels individually, while all other parameters are kept consistent with the BASE simulation (Table 1). Subtracting each sensitivity
105 simulation from the BASE simulation enables quantifying the influences of changes in ANTHRO and BB emissions on air quality and its associated health burden in India, respectively. We will discuss the air quality and mortality burden changes in six Indian regions based on meteorological conditions and aerosol variability (Fig. 1).

Table 1. Model simulations performed in this study

Simulation	Anthropogenic emissions	Biomass burning emissions
BASE	V	V
FixAN	1995	V
FixBB	V	1995

"V" indicates that particular input is subject to interannual variation in the simulation during the period 1995-2014, "FixAN"
110 indicates that only global ANTHRO emissions are fixed to 1995 levels in the simulation. "FixBB" indicates that only global BB emissions are fixed to 1995 levels.

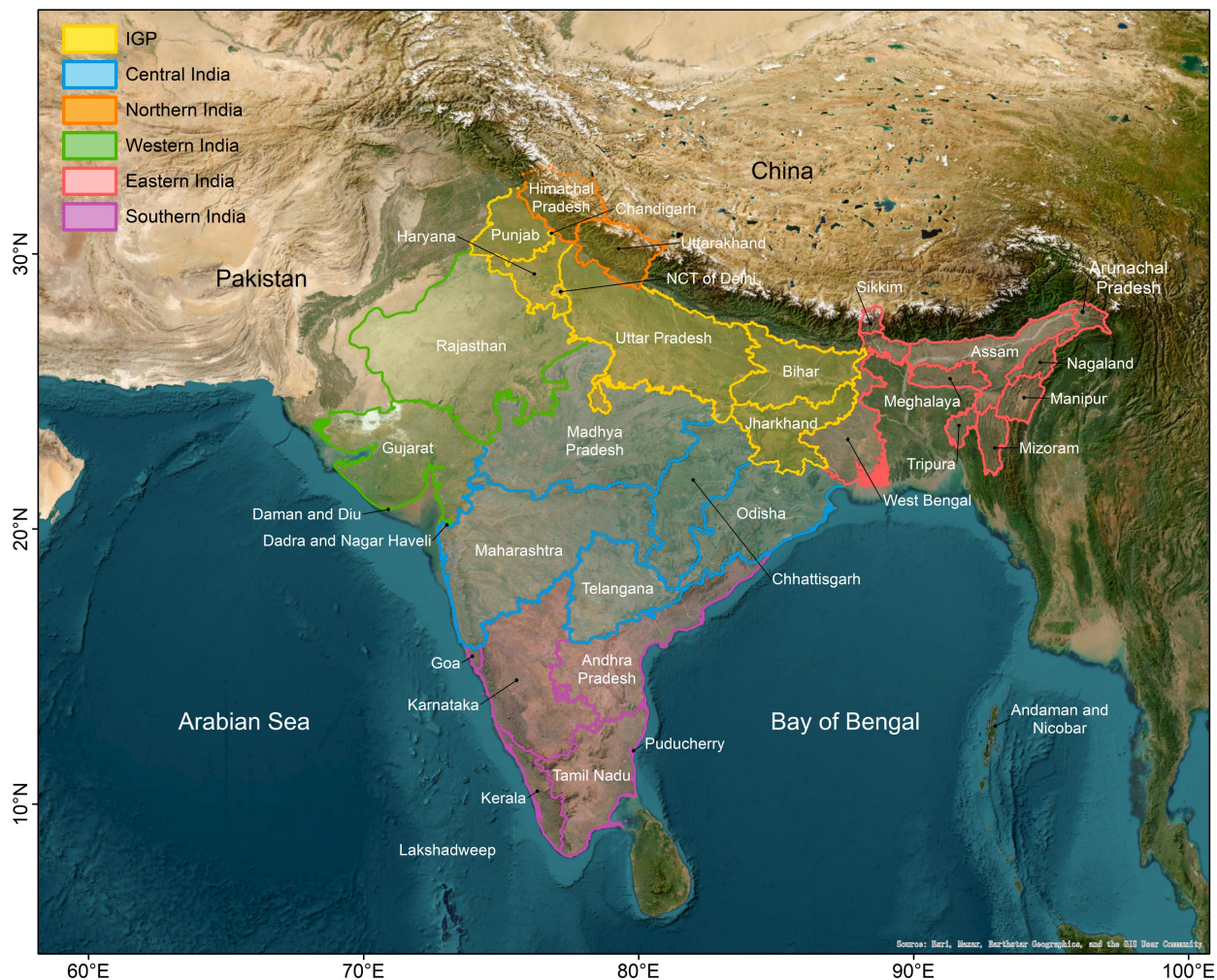


Figure 1. A map of India marked into six regions based on meteorological conditions and aerosol variability (adapted from David et al. 2018).

115 2.3 Trend estimation

In this study, we apply the Theil-Sen estimator (Theil, 1992; Sen, 1968) to calculate the magnitude of trends in surface PM_{2.5} and O₃ concentrations and the attributed mortality burden spanning from 1995 to 2014. The Theil-Sen estimator is a robust non-parametric method for trend analysis based on the median slope, which is insensitive to outliers and highly competent in identifying the slope of non-normally distributed data, as described in eq 1. This method has been widely used

120 to analyse temporal trends in air pollutants that are always non-normally distributed (e.g., Munir et al., 2013; Sarkar et al., 2019; Vanem and Walker, 2013; Wan et al., 2023).

$$Slope = Median \frac{(x_i - x_j)}{(t_i - t_j)} \quad (1)$$

Where x_i and x_j represent the concentrations of either PM_{2.5}, O₃ or attributed premature mortality at the time t_i and t_j ($i > j$), respectively, for the same parameter. $Slope > 0$ indicates an increasing trend; $Slope < 0$ indicates a decreasing trend.

125 In complement to the Theil-Sen estimator, we use the nonparametric Mann-Kendall test to assess the significance of temporal trends within the data series (Zhang et al., 2022a, b). Both the Mann-Kendall test and Theil-Sen estimator require independence and randomness in the data, making them suitable for identifying monotonic trends. According to previous studies, p-value less than 0.05 is most commonly treated as the absolute threshold of statistical significance (Christiansen et al., 2020; Wang et al., 2021; Zhou et al., 2017). The above methods are completed by implementing a Python program with the package “pymannkendall”, as detailed at <https://pypi.org/project/pymannkendall/>, last accessed on March 20, 2024.

130 2.4 Mortality burdens of surface PM_{2.5} and O₃ in India

Based on an integrated exposure-response function utilized in the most recent GBD studies, we estimate the mortality burden associated with long-term exposure to ambient annual PM_{2.5} and the O₃ season daily maximum 8-h mixing ratio (OSDMA8) in India spanning from 1995 to 2014, as described in eq 2.

$$\Delta Mort = y_0 \times AF \times pop = y_0 \times \left(\frac{RR - 1}{RR} \right) \times pop \quad (2)$$

135 Where $\Delta Mort$ refers to the annual mortality burden attributed to long-term PM_{2.5} or O₃ exposure, and y_0 is the baseline mortality rate for a specific cause of disease. AF is the attributable fraction measuring the PM_{2.5} or O₃ exposure attributable disease burden, which is represented by $\frac{RR-1}{RR}$ (RR refers to relative risk). pop represents the exposed population above the age of 25 for each grid cell in the domain.

140 Following our previous work (Zhang et al., 2021b), we obtain the baseline mortality rate (y_0) for each country and 5-year age group from 1995-2014 from the GBD2017 project (GBD 2017 Risk Factor Collaborators., 2018). The RR of long-term PM_{2.5} exposure associated with mortality burden due to specific diseases was estimated using an integrated exposure-response model (IER) constructed by Burnett et al. (2014) and updated in GBD2017. The RR for long-term O₃ exposure is obtained from Turner et al. (2016) which indicated an RR of 1.12 (95% confidence interval (CI): 1.08, 1.16) for respiratory disease. The recent GBD2019 reported a relatively lower RR for the chronic obstructive pulmonary disease (COPD), a subcategory of respiratory disease (1.06, with 95% CI: 1.03, 1.10). To be comparable with the GBD2019 results, we also 145 estimate the O₃-related mortality burden for the COPD in India during the same period. Population distribution with age

stratification data (*pop*) is retrieved from the GBD2017 with a horizontal resolution of 0.1°. The population-weighted (pop-weighted) average of specific air pollutants discussed in the results is calculated by weighting the population of all grid cells inside each administrative region or country. Additionally, we calculate mortality rates per capita (avoid deaths per 100,000 people) in each administrative region to exclude the influence of varying populations.

150 **3 Results and discussion**

3.1 CAM-chem evaluation

We perform a comprehensive model evaluation by comparing our simulated monthly concentrations from the BASE with multiple datasets, including ground-based observations in India, historical multi-model simulation from the CMIP6 project, and different versions of multi-year reanalysis data from the Atmospheric Composition Analysis Group (ACAG) at Washington University in St. Louis, hereinafter referred as 'Wustl Extracts' (van Donkelaar et al., 2021). We also compare our simulated PM_{2.5} and O₃ with previously published studies in India using either global or regional chemical transport models (CTMs), as well as the concentration reported from the GBD2019. We select available ground-level PM_{2.5} observations over India from previous studies (Latha and Badarinath, 2005; Panwar et al., 2013; Reddy et al., 2012; Saradhi et al., 2008; Tiwari et al., 2009, 2013), which were also collected by the ACAG. The locations of these sites are listed in Table S1. Figure S1 indicates that the model exhibits good performance in capturing seasonal variations of surface PM_{2.5} observations, especially during the peak months, with correlation coefficients (R) ranging from 0.59 to 0.91. Two exceptions are Mumbai (with R of -0.16), where the model shows a contrasting trend for the seasonal PM_{2.5} characteristics (Fig. S1b), and Mukteshwar (with R of 0.45). One possible explanation is the potential underestimation of emission inventories, especially during early periods for developing regions, such as India (McDuffie et al., 2020; Wang et al., 2022; Agarwal et al., 2024). For O₃, our model shows an even higher R when compared with the available surface observation sites in India from 1997 to 2011 (Fig. S2). Unlike the underestimations of surface PM_{2.5} in India, the CAM-chem model tends to overestimate surface O₃, which is not very uncommon for global CTM and also frequently discussed in previous studies (Hou et al., 2023; Tilmes et al., 2015; Young et al., 2018; Zhang et al., 2021b). The overestimation is partly caused by the coarse resolution, which leads to diluted emissions of O₃ precursors and then simulated high O₃ production. Figure 2 compares our study with previous studies and other publicly available PM_{2.5} and O₃ datasets, as detailed in Tables S2 and S3. The comparisons indicate our simulated results using the CAM-chem agree very well with previous studies for both PM_{2.5} and O₃, based on either the various metrics, such as average O₃, pop-weighted average O₃ or OSDMA8, consistent with the findings within the multiple CMIP6 models (Turnock et al., 2020). Figure S3 further compares the long-term trend of annual surface PM_{2.5} concentrations from 1998 to 2014 in the BASE simulation and Wustl Extracts dataset. A consistent increasing trend is found in both datasets, with temporal R of 0.86 and lower estimations in our model. The model performs better in eastern India than in western India, with R usually larger than 0.9 and NMB lower than -25%. Similarly, compared to the simulated trend in our study with different versions of Wustl Extracts and the GBD2019, our simulated PM_{2.5} concentration

is lower, and the simulated O₃ is higher (Fig. S4). The underestimation of the surface PM_{2.5} is partly caused by the missing model representation of nitrate and ammonium (Ren et al., 2023) and the secondary organic aerosol (Liu et al., 2021).

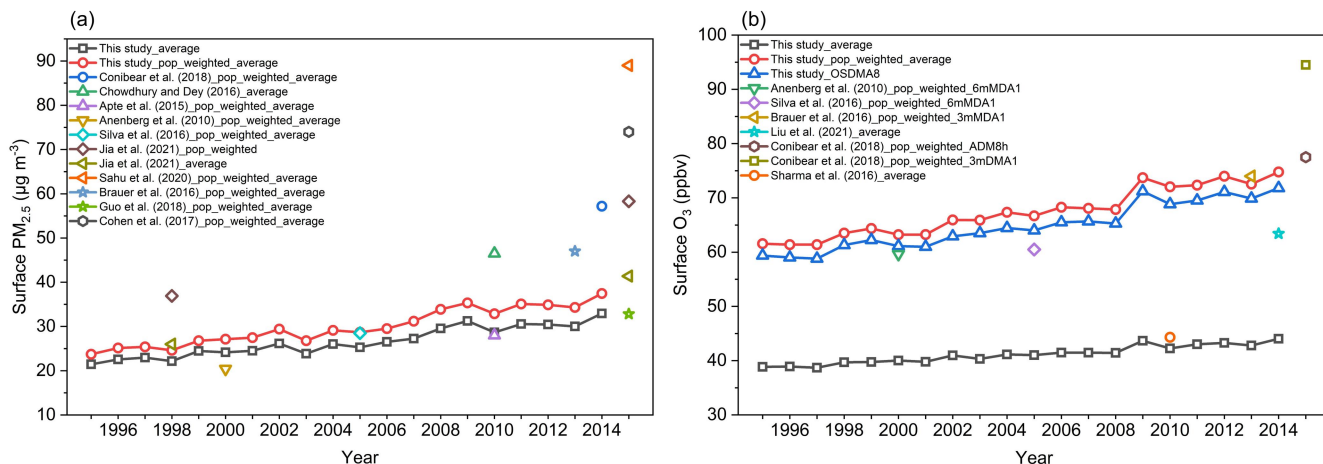


Figure 2. Comparison of annual PM_{2.5} and O₃ concentrations in India with previous studies. Note that the metrics vary depending on the study.

3.2 Spatial and temporal distribution of air pollution changes in India from 1995 to 2014

3.2.1 Historical emissions in India from 1995 to 2014

We first assess the interannual variation of ANTHRO and BB emissions of CO, NO_x, NMVOC, sulfur dioxide (SO₂), ammonia (NH₃), black carbon (BC), and organic carbon (OC) in India between 1995 and 2014 from the CEDS. Figure S5 indicates an overall increase in ANTHRO emissions before slowly falling after 2011. Significant inter-annual variations in BB emissions, such as in 1999, 2006, and 2009, were mainly caused by climate change-induced hot and arid conditions (Sahu et al., 2015). Figure S6 shows that ANTHRO emissions occurred predominately in IGP and central India, significantly increasing across all regions. Unlike other administrative regions, northern and eastern India, such as Punjab and Manipur, features a higher ratio of BB emissions to ANTHRO emissions.

3.2.2 The long-term trends of PM_{2.5} and O₃ in India from 1995 to 2014

From the BASE simulation, we estimate that the annual mean pop-weighted PM_{2.5} and O₃ for India in 1995 and 2014 were 29.88 $\mu\text{g m}^{-3}$ and 67.41 ppbv, respectively. Figure 3a, b show that annual average PM_{2.5} concentrations gradually rose from the south to the north, with high levels predominantly found in the IGP, mainly caused by high ANTHRO emissions (Fig. S6) and reduced ventilation due to obstruction by the Tibetan Plateau (Gao et al., 2018). Surface annual average O₃ concentrations gradually increased from west to east and south to north, with the highest levels concentrated in northern India and the eastern part of central India. The spatial patterns of the PM_{2.5} and O₃ distribution in India are also seen in several previous studies, though they only discussed one or several specific years (Jia et al., 2021; Pandey et al., 2021).

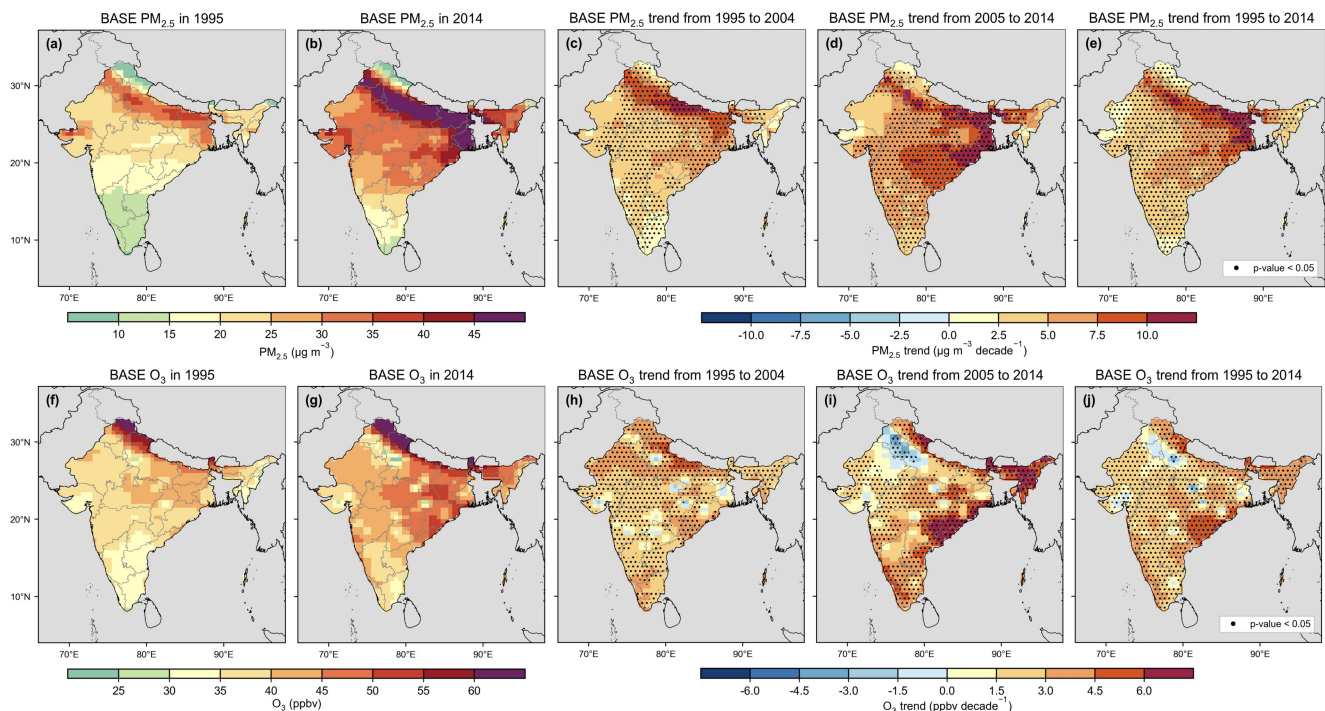


Figure 3. Spatial distributions of PM_{2.5} (top panel) and O₃ (bottom panel) for annual average in 1995 (a, f) and 2014 (b, g), with the trends from 1995 to 2004 (c, h), 2005 to 2014 (d, i), and 1995 to 2014 (e, j). The black dot denotes the areas where the trend is statistically significant ($p < 0.05$). The units are $\mu\text{g m}^{-3}$ for PM_{2.5} (a,b) and ppbv for O₃ in (f, g), and $\mu\text{g m}^{-3} \text{ decade}^{-1}$ for PM_{2.5} trends (c,d,e), and ppbv per decade (ppbv decade^{-1}) for O₃ trends (h,i,j).

From Figure 3, we also find that both PM_{2.5} and O₃ showed a statistically significant increasing trend all over the country from 1995 to 2014, with a nationwide increasing rate of $6.71 \mu\text{g m}^{-3} \text{ decade}^{-1}$ ($p < 0.01$) for pop-weighted PM_{2.5} and $7.08 \text{ ppbv decade}^{-1}$ ($p < 0.01$) for pop-weighted O₃, respectively (Fig. S7), which was mainly driven by rapid industrialization and substantial economy development (Pandey et al., 2014; Sadavarte and Venkataraman, 2014). However, distinct spatial heterogeneity for the increasing trend was observed for the two air pollutants. PM_{2.5} exhibited varying degrees of increase across India, with the most distinctive increase occurring in the IGP, where the maximum trend reached $12.60 \mu\text{g m}^{-3} \text{ decade}^{-1}$. This notable rise can be attributed to the increased regional ANTHRO emissions (Fig. S6). For O₃, eastern central India experienced the highest O₃ increases, with an obvious increase in the eastern and the lowest in western India. One thing needs to be pointed out that in northern IGP, including New Delhi, significant O₃ decreases were also observed, which could be caused by the inhibited O₃ production due to NO titration as a result of the dramatic increase in NO_x emissions, as discussed in Karambelas et al. (2018). Splitting the trend into two periods (from 1995 to 2004 and from 2005 to 2014), we find a larger increasing trend in the latter period than that in the previous one for both PM_{2.5} and O₃, which may be due to the rapid urbanization and growing transportation activities over populous regions (Fig. S8) in recent years in India (Gao et al., 2018).

3.3 Driving factor analysis for the air pollution changes in India

3.3.1 Contributions to the annual and seasonal trends

To disentangle the contributions of ANTHRO and BB emissions to long-term trends in PM_{2.5} and O₃ concentrations in India from 1995 to 2014, we first analyse their contributions to annual and seasonal trends (Figure 4). Not surprisingly, changes in ANTHRO emissions dominated the deterioration of PM_{2.5} and O₃ in India, consistent with studies based on observational and satellite data (Gurjar et al., 2016; Vohra et al., 2022). Changes in ANTHRO emissions alone increased area-weighted PM_{2.5} by 5.46 µg m⁻³ decade⁻¹ (p < 0.01) and area-weighted O₃ by 2.71 ppbv decade⁻¹ (p < 0.01), accounting for 102.21% and 104.11% of the total changes, respectively. The contributions of changes in BB emissions were relatively minor, with distinct interannual variations and seasonal variations. Spatially, we found that both the long-term PM_{2.5} and O₃ trends were mostly dominated by the ANTHRO emissions changes all over India (Fig. S9a, c). Changes in BB emissions led to a slightly increasing trend of PM_{2.5} in most of India and a decreasing trend in eastern India, though neither was statistically significant. BB emissions seemed to increase O₃ in IGP and central India and decrease O₃ in western India, but the trends were insignificant (Fig. S9b, d).

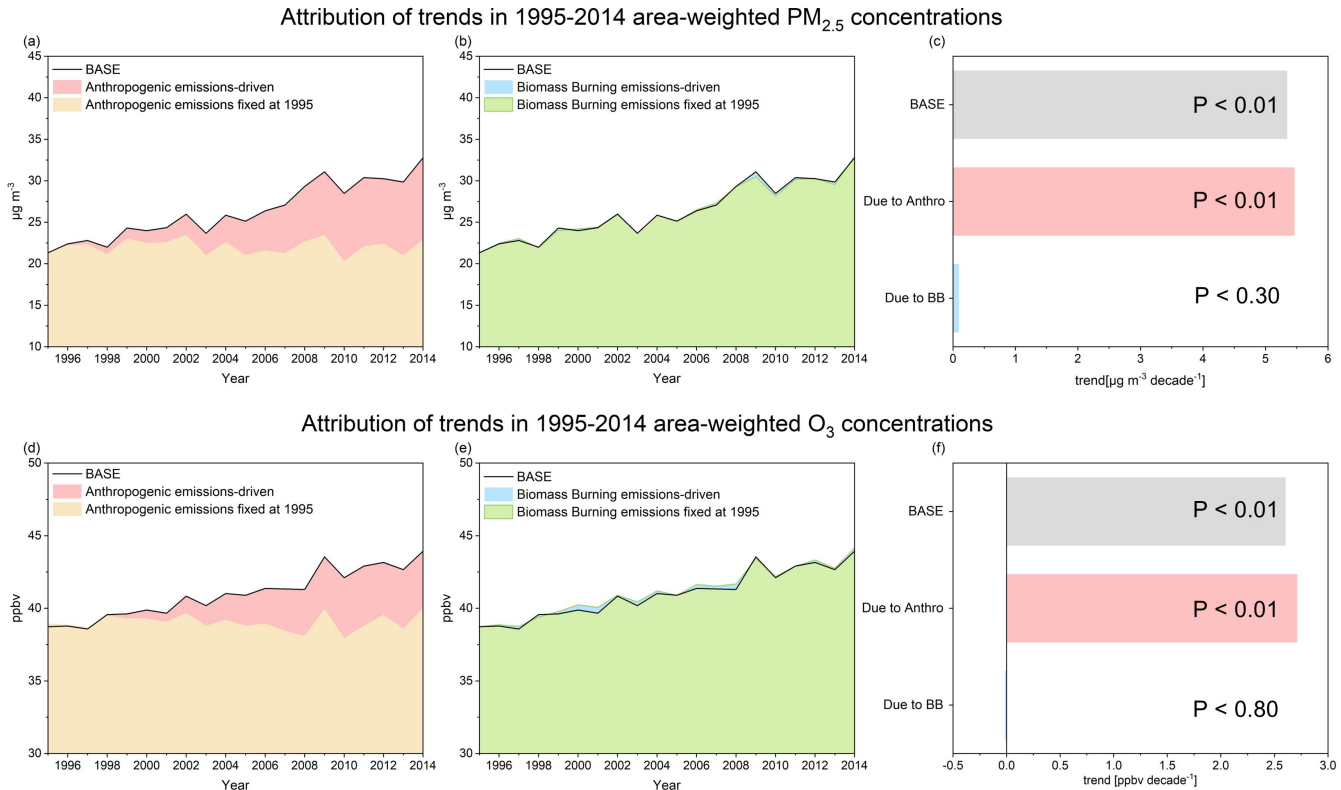
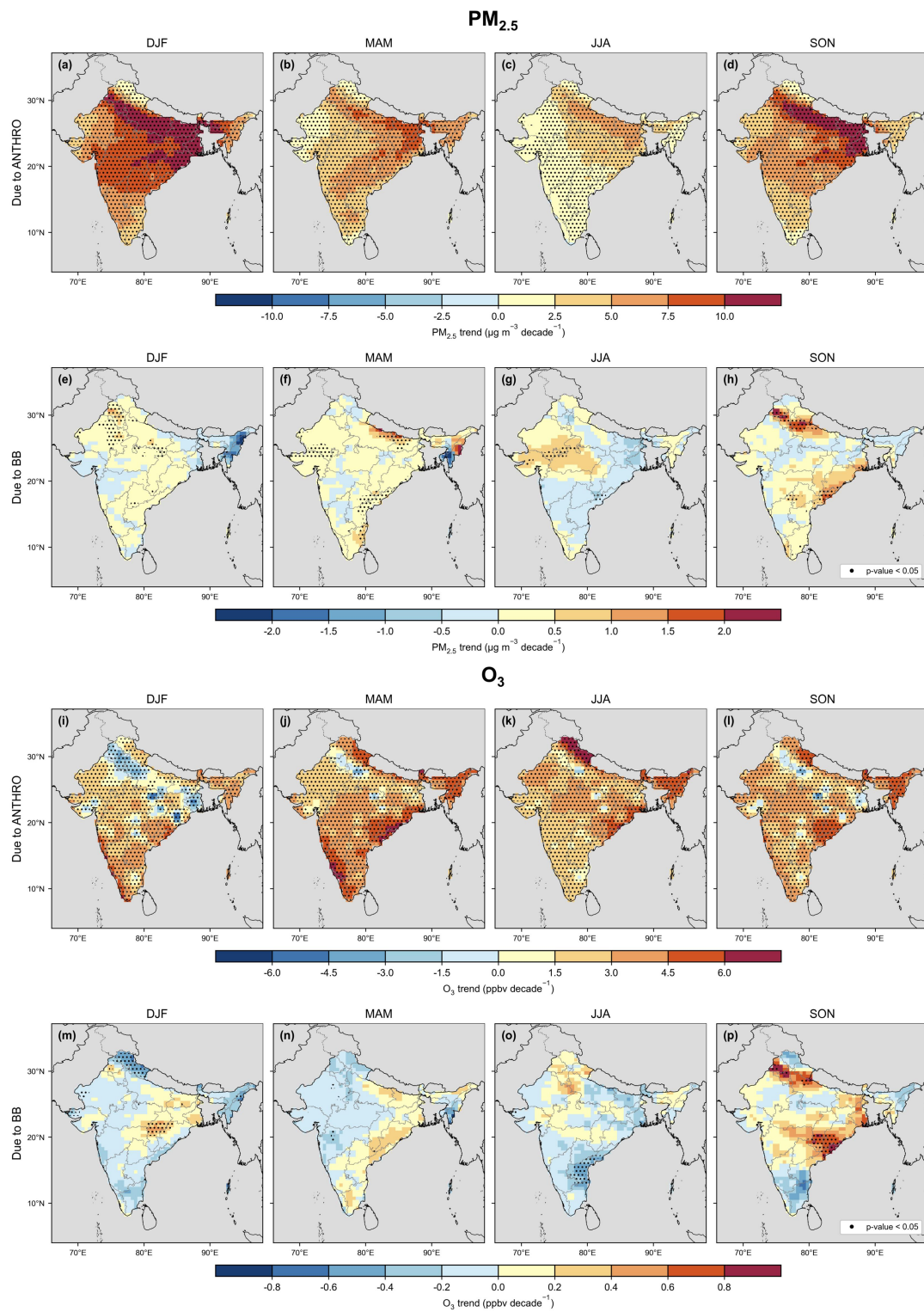


Figure 4. Drivers for trends of area-weighted (a-c) PM_{2.5} and (d-f) O₃ in India in 1995-2014. The yellow shadings in (a, d) show the evolution of model-simulated PM_{2.5} and O₃ concentrations in the FixAN simulation, with the red shadings illustrating the estimation of the PM_{2.5} and O₃ concentrations resulting from changes in ANTHRO emissions compared to the 1995 level. (b, e) as

for (a, d), but for impacts of changes in BB emissions. (c, f) denotes the estimated PM_{2.5} and O₃ trends in India derived from the BASE simulation and impacts of ANTHRO and BB emissions, respectively.

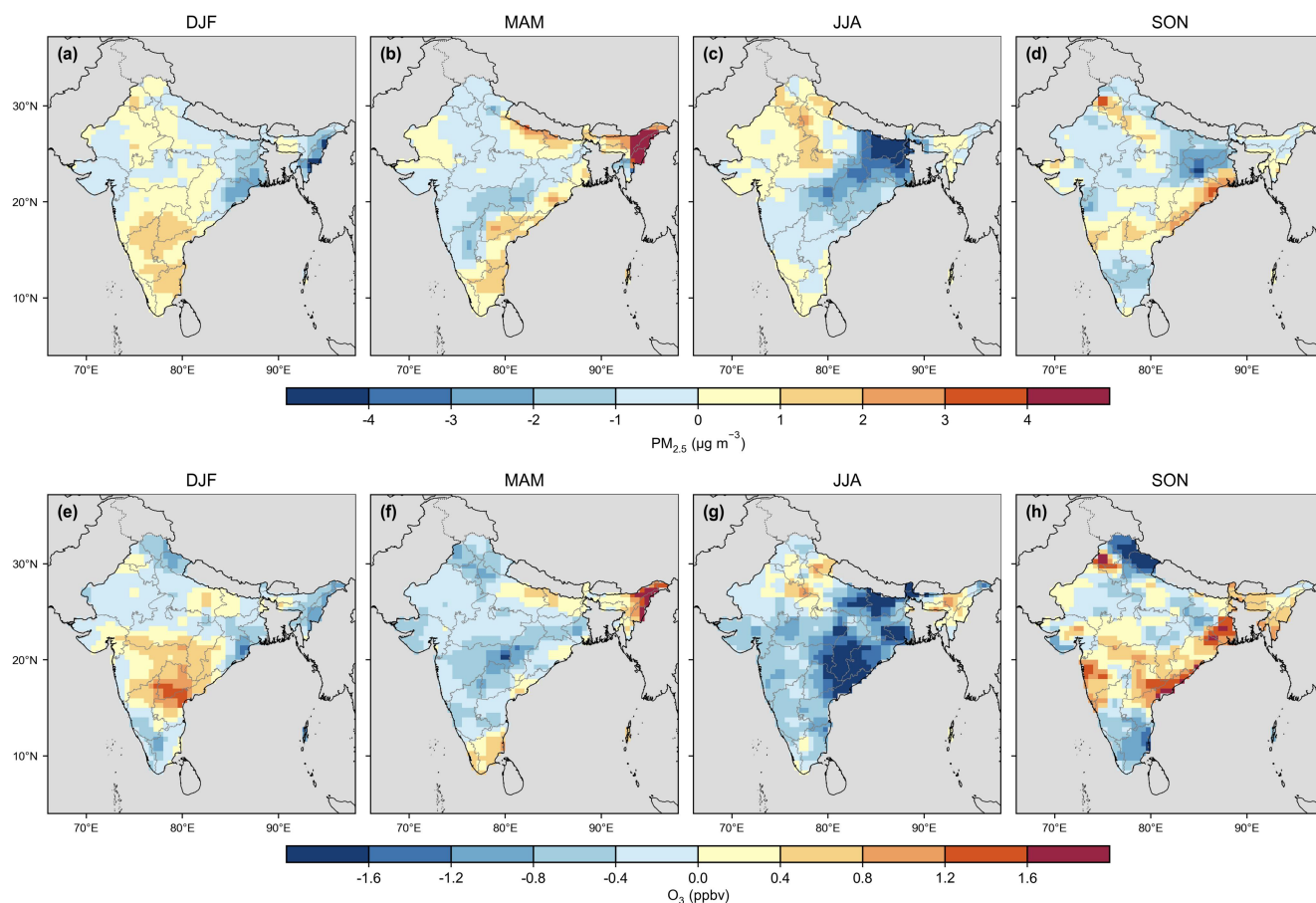
It is well recognized that BB emissions usually feature a distinct seasonal trend, especially in India, where monsoons influence them. Hence, here we quantify the seasonal trends of PM_{2.5} and O₃ from ANTHRO and BB emissions for DJF (December-January-February), MAM (March-April-May), JJA (June-July-August, monsoon season), and SON (September-October-November, post-monsoon season) from 1995 to 2014 by subtracting the FixAN or FixBB simulation from the BASE simulation. The annual trends for PM_{2.5} and O₃ for each season were subsequently estimated using the Theil-Sen estimator and the Mann-Kendall test. From Fig. 5a-h, we find that the contributions of ANTHRO emissions had consistent spatial patterns for the seasonal PM_{2.5} trend, with larger influences in the post-monsoon seasons (DJF and SON), which was estimated to be responsible for PM_{2.5} enhancement by as high as 17.08 $\mu\text{g m}^{-3} \text{ decade}^{-1}$ because of decreased vertical dispersion and diffusion of aerosol caused by lower solar radiation during winter and surface wind speeds (Bran and Srivastava, 2017). The contributions of ANTHRO emissions during the MAM and JJA were modulated as a result of increased precipitation, strong air convergence, and uplift strong air convergence during the presence of the summer monsoon, which impeded the accumulation of PM_{2.5} concentrations at ground level (Bran and Srivastava, 2017; Gao et al., 2020; Lu et al., 2018). Unlike PM_{2.5}, the contributions of ANTHRO emissions changes on surface O₃ trend in India had a distinct spatial pattern across seasons (Fig. 5i-p). The ANTHRO emissions had a much stronger positive influence on the O₃ increases in northern, eastern central, and eastern India during JJA and SON, while it had the largest increases in southern India in the pre-monsoon season (MAM, Fig. 5j). It was reported that the stronger solar radiation and higher temperature in MAM were attributed to an increase in the photochemical efficiency of O₃ in the presence of NO_x (Doherty et al., 2013; Jacob and Winner, 2009; Pusede et al., 2015). The decreased O₃ in the IGP was most pronounced in the DJF season (Fig. 5i), mainly attributed to lower solar radiation and titration of O₃ by higher NO_x levels (Kumar et al., 2012). Additionally, the occurrence of winter monsoon led to extensive air subsidence in northern India, resulting in low net O₃ production and strong horizontal export, ultimately leading to relatively low O₃ levels (Lu et al., 2018).



260 **Figure 5. Seasonal patterns of (a-d) ANTHRO and (e-h) BB emissions contributions for the trends of PM_{2.5} in India from 1995 to 2014 and (i-p) for O₃. The units are $\mu\text{g m}^{-3}$ per decade for PM_{2.5} and ppbv per decade for O₃. The dots in the plots indicate statistically significant trends, with p-values less than 0.05.**

3.3.2 Contributions to the seasonal air quality changes

BB emissions exhibit a high degree of interannual variability, leading to less clear trends in the annual data. Thus, 265 Figure 6 focuses on the spatial distributions of BB contributions for seasonal PM_{2.5} and O₃ changes between 1995 and 2014 rather than trends, as detailed in Table S4. These contributions are quantified by subtracting the FixBB simulation in 2014 from the BASE simulation in 2014. The changes in BB emissions from 1995 to 2014 contributed significantly to the PM_{2.5} increases in eastern India (over $20 \mu\text{g m}^{-3}$) with a high incidence of forest fires (Jena et al., 2015). It also led to an increase of O₃ by more than 4 ppbv in eastern India in MAM. Contributions to seasonal PM_{2.5} and O₃ changes from BB were 270 comparable to or even exceeded those from ANTHRO in some regions, such as Manipur and Nagaland (Fig. S10). With a higher BB fraction in other years, such as 1999, these contributions could even be even higher, reaching up to $46.03 \mu\text{g m}^{-3}$ and 6.46 ppbv for PM_{2.5} and O₃, respectively (Fig. S11). Therefore, despite their variability, the BB emissions in India posed a great threat to the air quality and thus could not be overlooked.



275 **Figure 6. Spatial distributions of the BB contribution for seasonal (a-d) $\text{PM}_{2.5}$ and (e-h) O_3 changes from 1995 to 2014 for DJF, MAM, JJA, and SON. The contributions from BB were calculated as the differences between the BASE and FixBB simulation in 2014. The units are $\mu\text{g m}^{-3}$ and ppbv.**

3.4 Long-term trends of premature mortality due to $\text{PM}_{2.5}$ and O_3 in India

We estimate that the national mortality burden attributable to ambient $\text{PM}_{2.5}$ exposure rose significantly from 698.29
 280 thousand in 1995 to 893.33 thousand in 2014, at a rate of 97.83 thousand per decade ($p < 0.01$, Figure 7a). Similarly, the
 mortality burden attributable to O_3 exposure also notably rose from 414.50 thousand in 1995 to 580.03 thousand in 2014,
 73.91 thousand per decade ($p < 0.01$). The hotspots of premature mortality attributable to $\text{PM}_{2.5}$ and O_3 exposure occurred in
 New Delhi and IGP regions in 1995 and 2014 (Fig. 7b-e), coincidentally with the dense population (Fig. S8). We find that
 Uttar Pradesh, Bihar, West Bengal, and Haryana, four states within the IGP region, accounted for 41.00% and 39.77% of the
 285 national premature mortality due to $\text{PM}_{2.5}$ and O_3 in 2014, respectively. Considering this heterogeneous spatial distribution, it
 is imperative for the IGP region to implement stronger air pollution control policies to safeguard human health, as discussed
 by Jia et al. (2021). Our estimations for the O_3 -related mortality burden are higher than those reported from the GBD2019
 (Fig. S12) since we applied a higher RR and used larger baseline mortality rates (see Methods section 2.4). After

recalculating the O₃-related mortality burden using the GBD2019 metrics, we report an increasing trend of 29.74 thousand deaths per decade⁻¹ for O₃-related mortality, comparable to the GBD2019 estimation of 33.24 thousand deaths per decade⁻¹. However, our estimated mortality burdens are still slightly higher than the GBD2019 due to the O₃ overestimation in our model (Fig. 2 and Fig. S4).

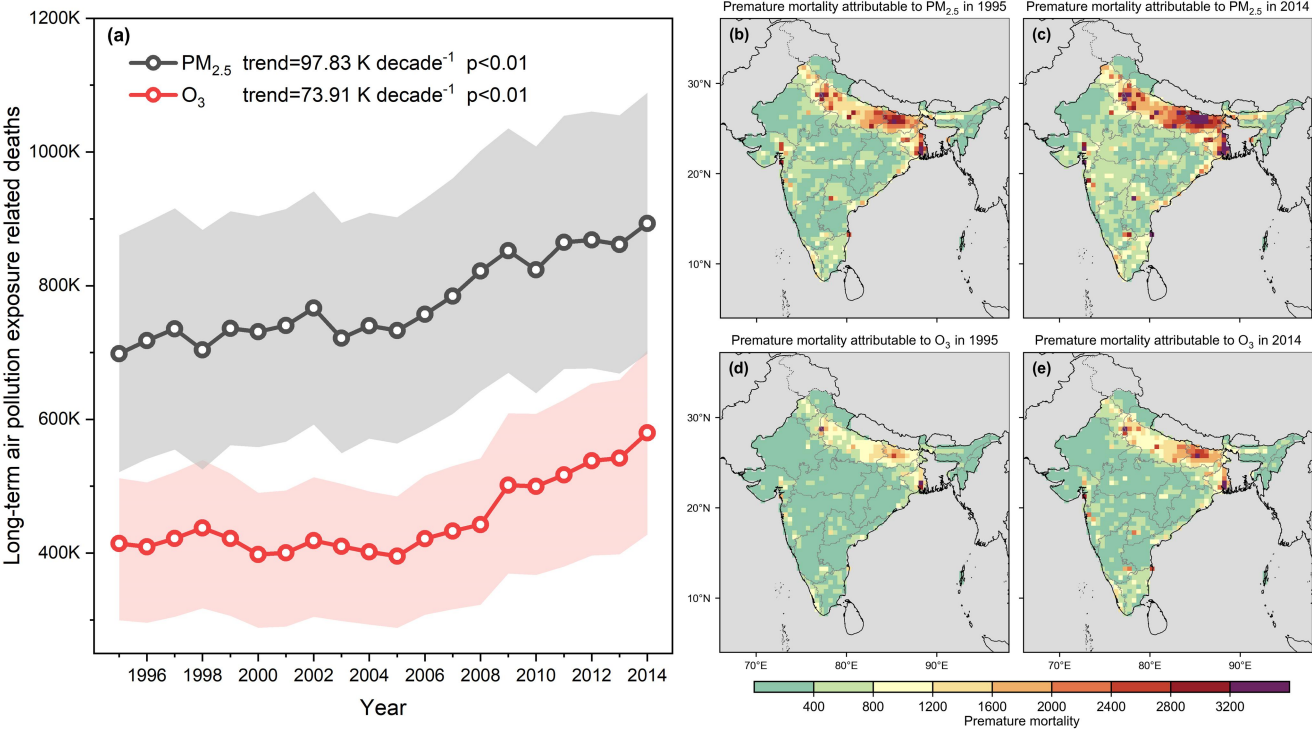


Figure 7. Spatial-temporal change of mortality burden attributable to PM_{2.5} and O₃. (a) interannual variation from 1995 to 2014. The shaded area indicates the range of 95% confidence interval account for RR estimates of long-term exposure to PM_{2.5} and O₃ (gray indicates half of the range). (b-e) spatial distributions of the average annual premature mortality attributable to (b-c) PM_{2.5} and (d-e) O₃ in 1995 and 2014.

To isolate the effects of population heterogeneous among regions, we also quantify the mortality burden changes per capita (avoided deaths per 100,000 people) from 1995 to 2014 (Fig. 8). PM_{2.5}-attributable premature mortality per capita was higher in the IGP and eastern India, with the highest in Chandigarh (427.2), followed by Sikkim (153.6), Meghalaya (140.3), and NCT of Delhi (126.1) in 1995 (Fig. S13). The spatial distribution of O₃-attributable premature mortality per capita resembled that of PM_{2.5}. However, the values were relatively smaller, with the maximum value also appearing in Chandigarh (288.0), followed by Sikkim (120.2), Meghalaya (68.6), and NCT of Delhi (68.0) in 1995 (Fig. S13). Over the period from 1995 to 2014, PM_{2.5}- and O₃- attributable premature mortality per capita decreased in the north and increased in the south (Fig. 8), indicating that the increasing trend of premature mortality attributable to PM_{2.5} and O₃ in the IGP region was mainly driven by the increased population (Fig. S8).

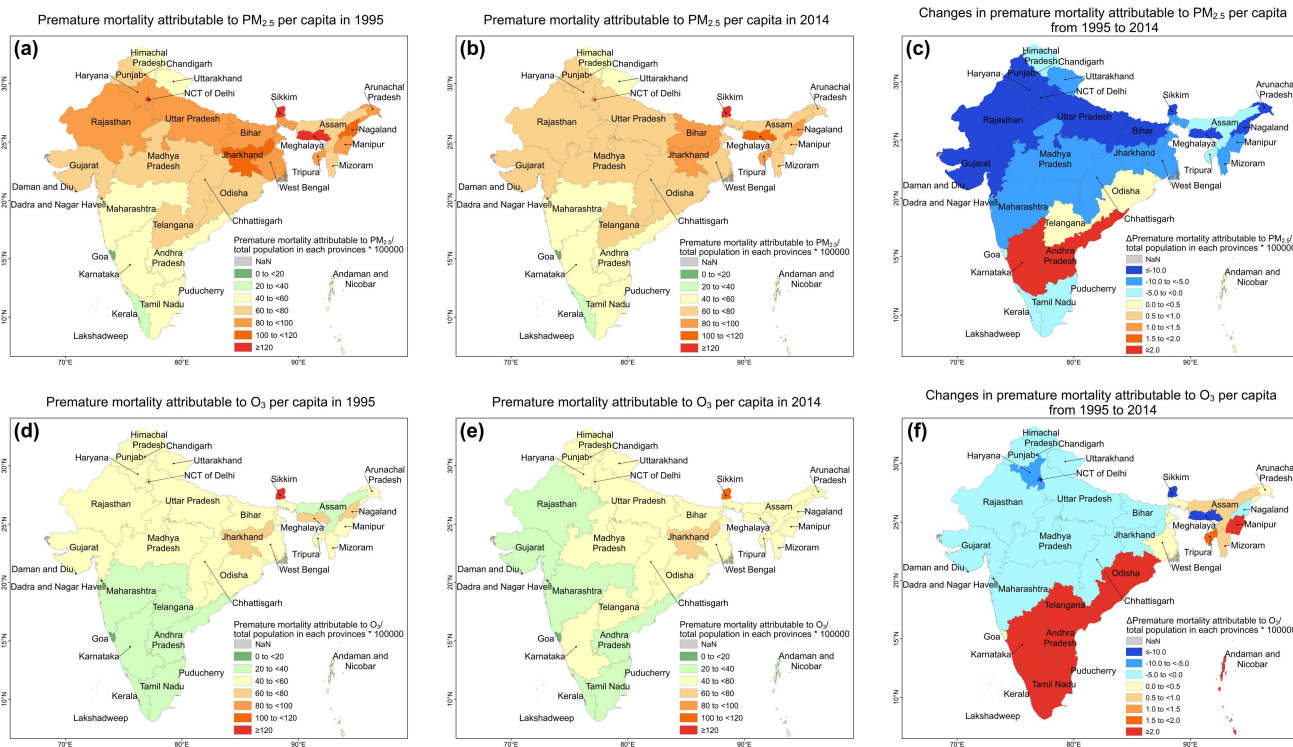


Figure 8. Spatial distributions of premature mortality attributable to PM_{2.5} or O₃ per capita (avoid deaths per 100,000 people) in (a, d) 1995, (b, e) 2014, and (c, f) changes from 1995 to 2014 in the state of India.

Figure 9 shows that changes in ANTHRO emissions from 1995 to 2014 increased premature mortality per capita attributable to PM_{2.5}, with the higher values located mainly in eastern IGP and central India. Changes in BB emissions increased premature mortality attributable to PM_{2.5} per capita in eastern, western, and southern India and decreased in IGP and central India. The state with the largest increase was Manipur (2.55), followed by Nagaland (2.06), associated with the high incidence of wildfires in these regions. The state that experienced the largest decrease was Jharkhand (-1.71), with Bihar (-1.02) followed behind. To explore contribution changes from ANTHRO and BB emissions, we estimate the premature mortality attributable to PM_{2.5} per capita in 2000, 2005, and 2010-2014 in Table S5, respectively, consistent with the demonstrations from GBD2017. There was a sharp rise in contributions to premature mortality attributable to PM_{2.5} from changes in ANTHRO emissions from 1995 to 2014. Not surprisingly, the premature mortality attributable to PM_{2.5} from changes in BB emissions fluctuated greatly from 1995 to 2014. In 2000, a year with high BB emissions (Fig. S5), the contributions of changes in BB emissions to the premature mortality attributable to PM_{2.5} in the states of Mizoram, Nagaland, Arunachal Pradesh, and Tripura reached 5.14, 4.90, 4.86, and 4.17, respectively, which exceeding the contributions of changes in ANTHRO emissions in that year (Table S5).

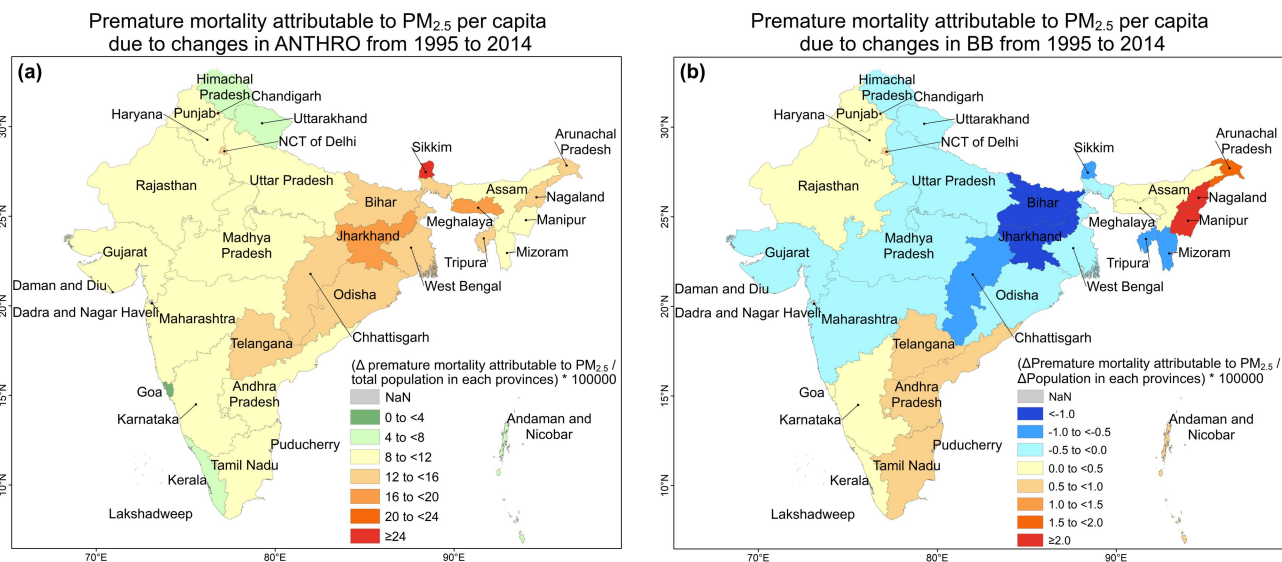


Figure 9. Spatial distributions of contributions to premature mortality attributable to PM_{2.5} per capita (avoided deaths per 100,000 people) from changes in (a) ANTHRO and (b) BB emissions from 1995 to 2014.

4 Conclusions

In this study, we apply a state-of-the-art global CTM (CAM-chem) to provide a detailed assessment of long-term trends of the ambient annual mean PM_{2.5} and O₃ in India and their health burden from 1995 to 2014, as well as the driving factor from ANTHRO and BB emissions changes. The annual mean area-weighted PM_{2.5} over India increased at 5.34 $\mu\text{g m}^{-3}$ decade⁻¹ ($p < 0.01$) from 1995 to 2014, dominated by the ANTHRO emissions (5.46 $\mu\text{g m}^{-3}$ decade⁻¹, $p < 0.01$). The highest and fastest PM_{2.5} growth was in the IGP regions due to the rapid industrialization, urbanization, and transportation growth. For annual mean area-weighted O₃, the increase was 2.60 ppbv decade⁻¹ ($p < 0.01$), dominated by the ANTHRO emissions as well (2.71 ppbv decade⁻¹, $p < 0.01$). We find that O₃ concentrations were highest in northern India, with the fastest growth occurring in northern, central, and eastern India. The contributions from BB emissions for the long-term trends were not significant for either PM_{2.5} (0.09 $\mu\text{g m}^{-3}$ decade⁻¹, $p < 0.30$) or O₃ (-0.01, $p < 0.80$) and showed significant seasonal variations due to large inter-annual variability features. However, when we examine the air quality changes in specific years, such as 1999 and 2014, when there were larger BB activities in India, we found that the contributions from BB could be comparable to or even exceed those from ANTHRO during DJF and MAM, reaching over 46.03 $\mu\text{g m}^{-3}$ and 6.46 ppbv for PM_{2.5} and O₃, respectively.

Further estimation of mortality burden shows a 27.93% (698.29 to 893.33 thousand) increase in premature mortality attributable to PM_{2.5} between 1995 and 2014 (22.94% for 2005-2014), and a 39.93% (414.50 to 580.03 thousand) increase for O₃ (44.54% increasing during 2005-2014). Changes in ANTHRO and BB emissions were responsible for an

enhancement of premature mortality attributable to PM_{2.5} by 88.78% (97.83 thousand per decade, $p < 0.01$) and 0.02% (2.38 thousand per decade, $p < 0.10$). After removing the effect of population growth, our analysis reveals a notably higher mortality burden per capita attributable to PM_{2.5} in the IGP regions. However, it was noteworthy that the mortality burden per capita in these regions exhibited a significant decline over the period of 1995-2014, despite the increasing trend of premature mortality. This suggests that population growth was the primary factor driving the trend of premature mortality.

Our study is subject to several uncertainties and limitations. First of all, the coarser resolution in the global model ($0.9^\circ \times 1.25^\circ$) frequently cannot realistically represent the complex physical and chemical processes of regional-scale air pollution, especially for O₃ (Yue et al., 2023). Moreover, missing chemical mechanisms in the model, such as the lack of representations of nitrate and ammonium (Ren et al., 2023) and the secondary organic aerosol (Liu et al., 2021), prevented the model from accurately simulating PM_{2.5} concentration, especially during heavily polluted regions, such as China and India (Turnock et al., 2020). Another major uncertainty originated from the inaccurate emission inventory, especially for developing regions in early periods, as reported by the global datasets (Paulot et al., 2018; Wang et al., 2022). Zhang et al. (2021b) revealed that model performance with global CEDS inventory tends to predict lower bias for surface PM_{2.5} and higher bias for surface O₃ compared with a regional emission inventory (MEIC) in China due to disparities in spatial allocation. Xie et al. (2024) also highlighted a significant underestimation of agricultural fires in the inventory. Moreover, the uncertainty from health functions ranging from the choice of the exposure-response functions (Ostro et al., 2018; Giani et al., 2020) and the uncertainties of the baseline mortality rates both had different impacts on human health (Lelieveld et al., 2015; Pozzer et al., 2023). Meanwhile, in estimating the mortality burden, we apply the RR derived from a global study rather than using values specific to India, which could potentially be lower (Brown et al., 2022). Thus, our estimations for the air pollution-related mortality burden could be overestimated. More epidemiology studies should be conducted in India to retrieve their RR. Finally, another limitation in our experimental design was that we set global fixed emissions for ANTHRO and BB instead of in India only, ignoring the impact of intercontinental transportation.

Code and data availability

The CESM model code is available at https://www.cesm.ucar.edu/models/cesm2/release_download.html (last access: 12 March 2024). Observation data is available at <https://wustl.box.com/s/79pfex658crbq4dykxh51vvfdpksfhj5> (last access: 29 March 2024), which is collected by the Atmospheric Composition Analysis Group (ACAG) at Washington University in St. Louis.

Author contributing

B.L. analysed the simulation results and wrote the manuscript. Y.Z. and T.T. conceived the idea, designed and conducted the experiment. Y.Z., T.T., H.Z., J.H., J.M., W.W., and L.X. revised the original paper. All authors contributed to the manuscript.

Competing interests

The authors declare that they have no conflict of interest.

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