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) and biomass burning (BB) emissions are the major sources of ambient air pollution. India has experienced a dramatic deterioration in air quality over the past few decades, but no systematic assessment has been made to investigate the individual contributions of ANTHRO and BB emissions changes over the long term in India, extension to non-urban areas. In this study, we conducted a comprehensive analysis of the long-term trends of particulate matter with aerodynamic diameters < 2.5 µm ($PM_{2.5}$) 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 revealed a substantial nationwide increase in annual mean $PM_{2.5}$ (6.71 µg m^{-3} decade^{-1}) and O_3 (7.08 ppbv decade^{-1}), with the Indo-Gangetic Plain (IGP) and eastern central India as hotspots for $PM_{2.5}$ and O_3 trend changes individually. Noteworthy substantial O_3 decreases were observed in the northern IGP which were potentially linked to NO titration due to a surge in NOx emissions. Sensitivity analyses highlighted ANTHRO emissions as primary contributors to rising $PM_{2.5}$ 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 $PM_{2.5}$ and O_3 changes were comparable with or even exceeding ANTHRO emissions in specific areas. We further estimated that the elevated air pollutants were associated with increased premature mortality attributable to $PM_{2.5}$ and O_3, leading to 97.83 K and 73.91 K per decade. Despite a per capita decrease in the IGP region, the increased population offset its effectiveness.

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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 µm ($PM_{2.5}$) 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 modeling studies indicated that districts exceeding India's annual ambient standard of 40 µg 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 $PM_{2.5}$ 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 NOy, to higher altitudes where the ozone lifetime is prolonged, facilitating 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 (NOx), 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

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quality and human health because large amounts of compounds are emitted into the atmosphere, namely carbon dioxide (CO_2), NOx, 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 analyze 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 O_3, 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.

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2 Methods

2.1 CAM-chem model configuration

We simulated surface $PM_{2.5}$ and O_3 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 was run at 1.25° (longitude) × 0.9° (latitude) horizontal resolution with 32 vertical levels reaching ~45 km. We configured 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 adopted 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 was considered the surface level, and all the model outputs were then regridded to a finer resolution 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 were 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 includes 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, were then re-speciated to match the chemical species in the latest CESM2 model,

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following the steps introduced by Emmons et al. (2020). Interpolation of the emission inventory from its original resolution (0.5° × 0.5°) to the target model resolution (0.9° × 1.25°) before being input into the model. Global historical BB emissions were 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 were 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).

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2.2 Numerical experiments designs

The standard simulation (BASE) was driven by year-to-year variability ANTHRO and BB emissions from 1995 to 2014, as described above. To separate the contributions from these two emission sources, we then conducted two sensitivity simulations in which ANTHRO emissions (FixAN) and BB emissions (FixBB) were fixed at 1995 levels individually, while all other parameters were kept consistent with the BASE (Table 1). Subtracting the BASE from each sensitivity 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).

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Table 1. Model simulations performed in this study

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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" indicates that only global ANTHRO emissions were set to 1995 conditions in the simulation. "FixBB", indicates that only global BB emissions were set to 1995 level.

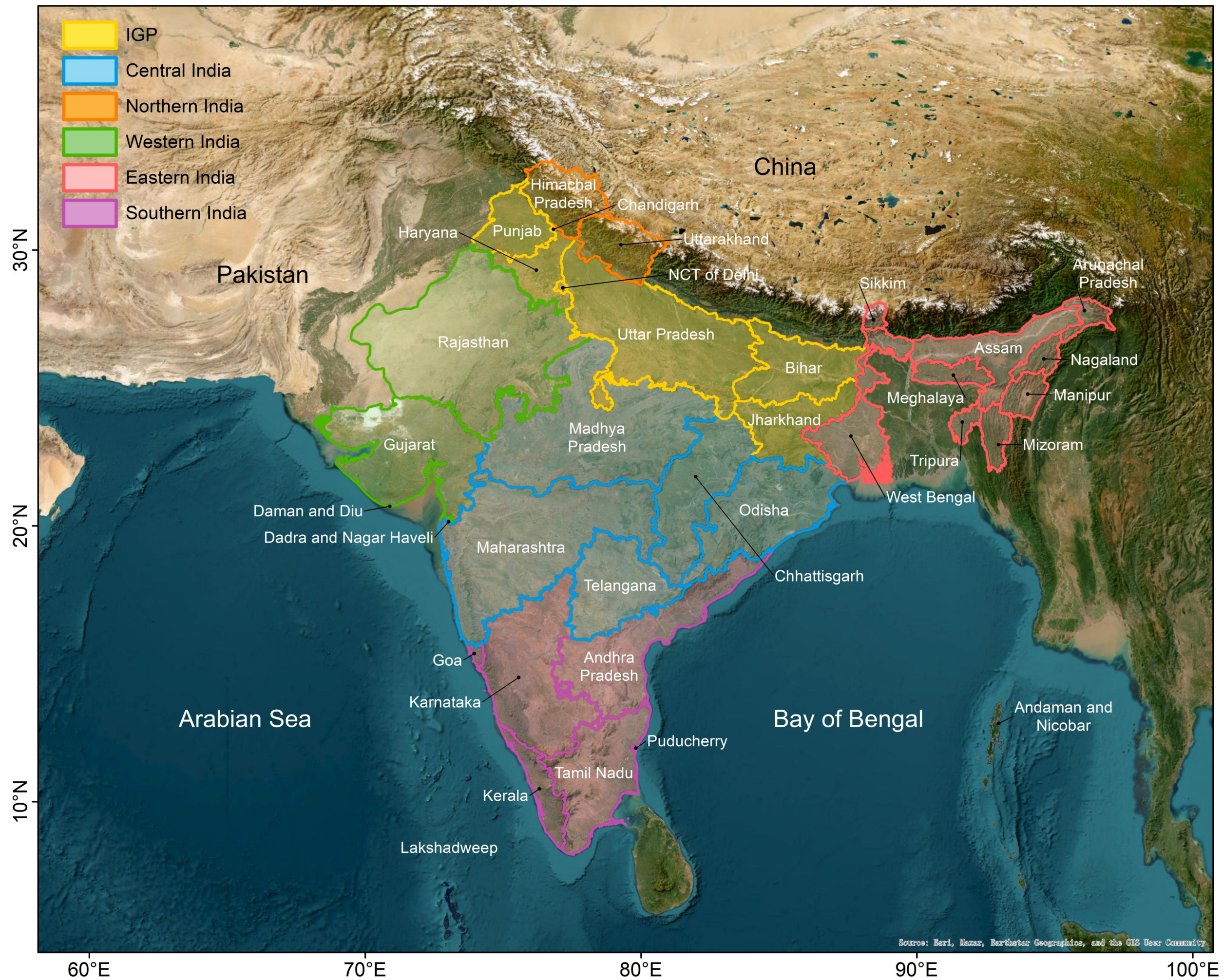

Figure 1. A map of India marked into six regions based on meteorological conditions and aerosol variability (adapted from David et al. 2018).

2.3 Trend estimation

In this study, we applied the Theil-Sen estimator (Theil, 1992; Sen, 1968) to calculate the magnitude of trends in surface $PM_{2.5}$ and O_3 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 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 $PM_{2.5}$, O_3, or attributed premature mortality at 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.

In complement to the Theil-Sen estimator, we used the nonparametric Mann-Kendall test to assess the significance of temporal trends within the data series (Zhang et al., 2022a, b). 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 were completed by implementing a Python program with the package "pymannkendall", as detailed at https://pypi.org/project/pymannkendall/, last accessed on March 20, 2024.

2.4 Mortality burdens of surface $PM_{2.5}$ and O_3 in India

Based on an integrated exposure-response function utilized in the most recent GBD studies, we estimated the mortality burden associated with long-term exposure to ambient annual $PM_{2.5}$ and 6-month running average of daily maximum 8-hr average (6mMDA8) O_3 in India spanning from 1995 to 2014, as described in eq 2.

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

Where $\Delta Mort$ refers to the annual mortality burden attributed to long-term $PM_{2.5}$ or O_3 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 O_3 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.

Following our previous work (Zhang et al., 2021b), we obtained 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 disease was estimated using an integrated exposure-response model (IER) constructed by Burnett et al. (2014) and updated in GBD2017. The RR for long-term O_3 exposure was 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 estimated the O_3-related mortality burden for the COPD in India during the same period. Population distribution with age stratification data (pop) was 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 was calculated by weighting the population of all

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grid cells inside each administrative region or country. Additionally, we calculated mortality rates per capita (avoid deaths per 100,000 people) in each administrative region to exclude the influence of varying populations.

3 Results and discussion

3.1 CAM-chem evaluation

We performed 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 compared our simulated $PM_{2.5}$ and O_3 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 selected 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 indicated 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 showed 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 O_3, our model showed 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 O_3, which was 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 was partly caused by the coarse resolution, which leads to diluted emissions of O_3 precursors and then simulated high O_3 production. Figure 2 compared our study with several previous studies and other publicly available $PM_{2.5}$ and O_3 datasets, as detailed in Tables S2 and S3. The comparisons indicated our simulated results using the CAM-chem agree very well with previous studies for both $PM_{2.5}$ and O_3, based on either the various metrics, such as annual averages, pop-weighted averages or annual maximum of 6mMDA8 O_3, 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 and Wustl Extracts dataset. A consistent increasing trend was 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 being 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 O_3 is higher (Fig. S4). The underestimation of the

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surface $PM_{2.5}$ was partly caused by the missing model representation of nitrate and ammonium (Ren et al., 2023) and the secondary organic aerosol (Liu et al., 2021).

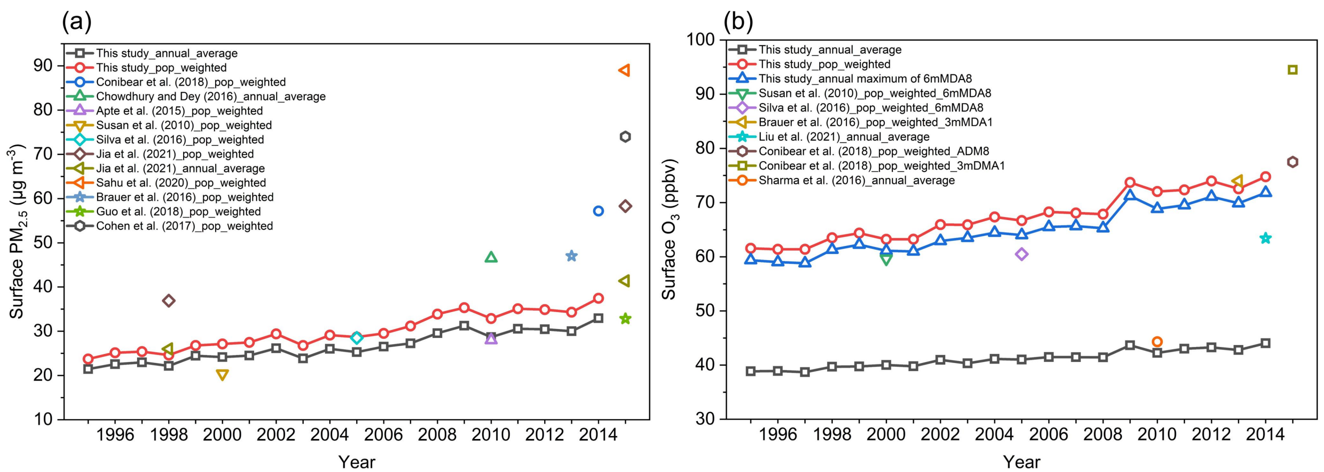


Figure 2. Comparison of annual $PM_{2.5}$ and O_3 concentrations in India with previous studies. Note that the metrics vary depending on the study.

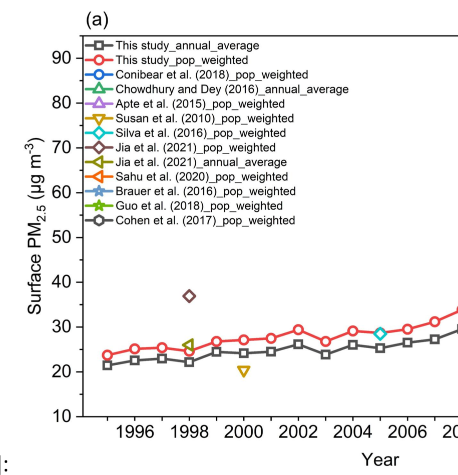
3.2 Spatial and temporal distribution of air pollution changes in India from 1995 to 2014

3.2.1 Historical emissions in India from 1995 to 2014

We first assessed the interannual variation of ANTHRO and BB emissions of CO, NOx, NMVOC, sulfur dioxide (SO_2), ammonia (NH_3), black carbon (BC), and organic carbon (OC) in India between 1995 and 2014 from the CEDS. Figure S5 indicated an overall increase in ANTHRO emissions before slowly falling after 2011. Significant inter-annual variations for 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 showed 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 O_3 in India from 1995 to 2014

From the BASE simulation, we estimated that the annual mean pop-weighted $PM_{2.5}$ and O_3 for India in 1995 and 2014 were 29.88 μg m^{-3} and 67.41 ppbv, respectively. Figure 3a, b showed 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 O_3 concentrations gradually increase 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 O_3 distribution in India were also



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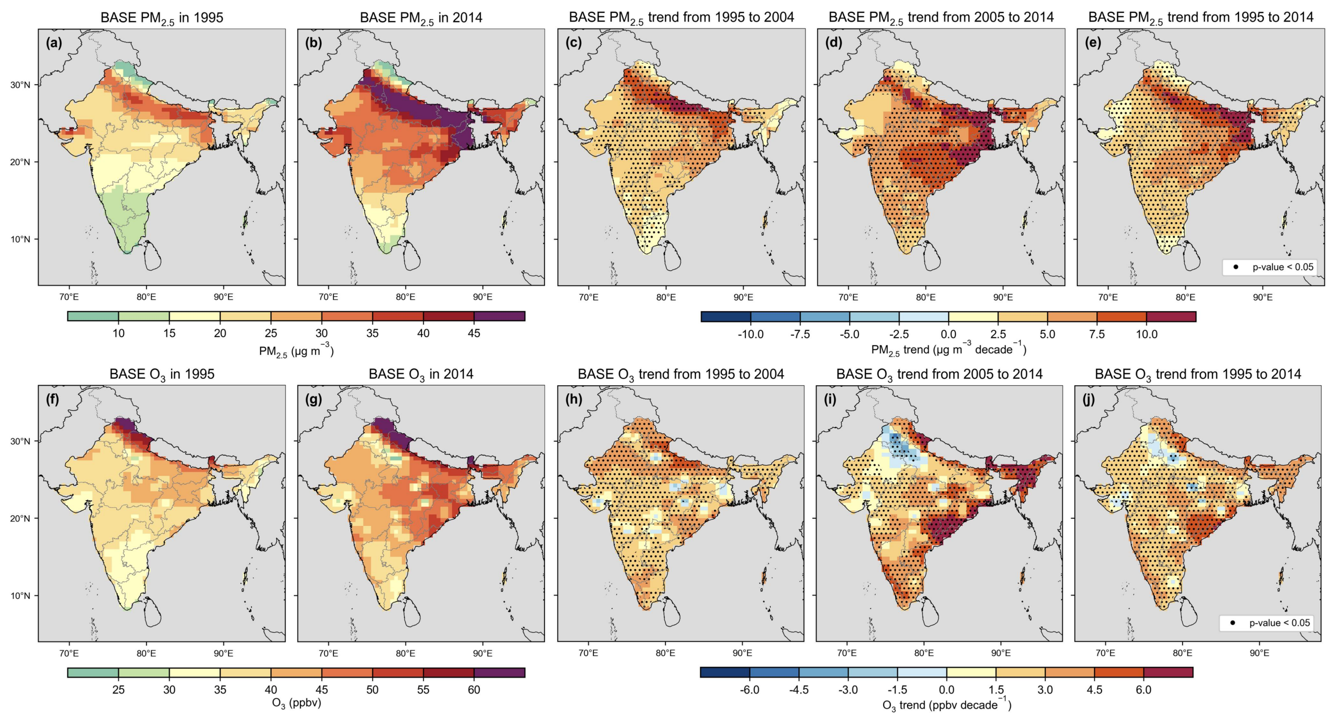
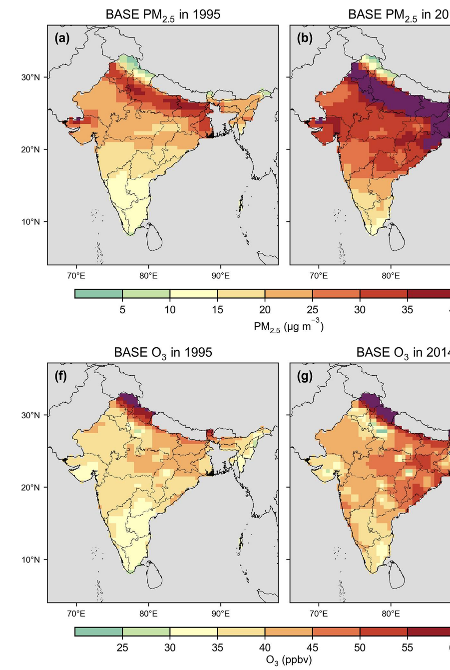


Figure 3. Spatial distributions of $PM_{2.5}$ (top panel) and O_3 (bottom panel) for annual average in 1995 (a, f) and 2014 (b, g), with the trends from 1995 to 2004 (c, h), 2005 to 2014 (d, i), and 1995 to 2014 (e, j). The black dot denotes the areas where the trend is statistically significant ($p < 0.05$). The units are μg m^{-3} for $PM_{2.5}$ (a,b) and ppbv for O_3 in (f, g), and μg m^{-3} per decade (μg m^{-3} $decade^{-1}$) for $PM_{2.5}$ trends (c,d,e), and ppbv per decade (ppbv $decade^{-1}$) for O_3 trends (h,i,j).

From Figure 3, we also found that both $PM_{2.5}$ and O_3 showed a statistically significant increasing trend all over the country from 1995 to 2014, with a nation-wide increasing rate of 6.71 μg m^{-3} decade $^{-1}$ ($p < 0.01$) for pop weighted $PM_{2.5}$ and 7.08 ppbv $decade^{-1}$ ($p < 0.01$) for pop-weighted O_3, 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 μg m^{-3} $decade^{-1}$. This notable rise can be attributed to the increased regional ANTHRO emissions (Fig. S6). For O_3, eastern central India experienced the highest O_3 increases, with an obvious increase in the eastern and the lowest increases in western India. One thing needs to be pointed out that in northern IGP, including New Delhi, significant O_3 decreases were also observed, which could be caused by the inhibited O_3 production due to NO titration as a result of dramatic increase in NOx emissions, as discussed in Karambelas et al. (2018). Splitting the trend into two periods (from 1995 to 2004 and from 2005 to 2014), we found a larger increasing trend in the latter period than that in the previous one for both $PM_{2.5}$ and O_3,



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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

Figure 4 showed the contributions of ANTHRO and BB emissions changes on area-weighted $PM_{2.5}$ and O_3 trends from 1995 to 2014. Not surprisingly that ANTHRO emissions changes dominated the $PM_{2.5}$ and O_3 deterioration in India. 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 O_3 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 O_3 trends were mostly dominated by the ANTHRO emissions changes all over India (Fig. S9a, c). Changes in BB emissions led to a slight increasing trend of $PM_{2.5}$ in most of India and a decreasing trend in eastern India, though neither of these trends was statistically significant. BB emissions seemed to increase O_3 in IGP and central India and decrease O_3 in western India, but the trends were insignificant either (Fig. S9b, d).

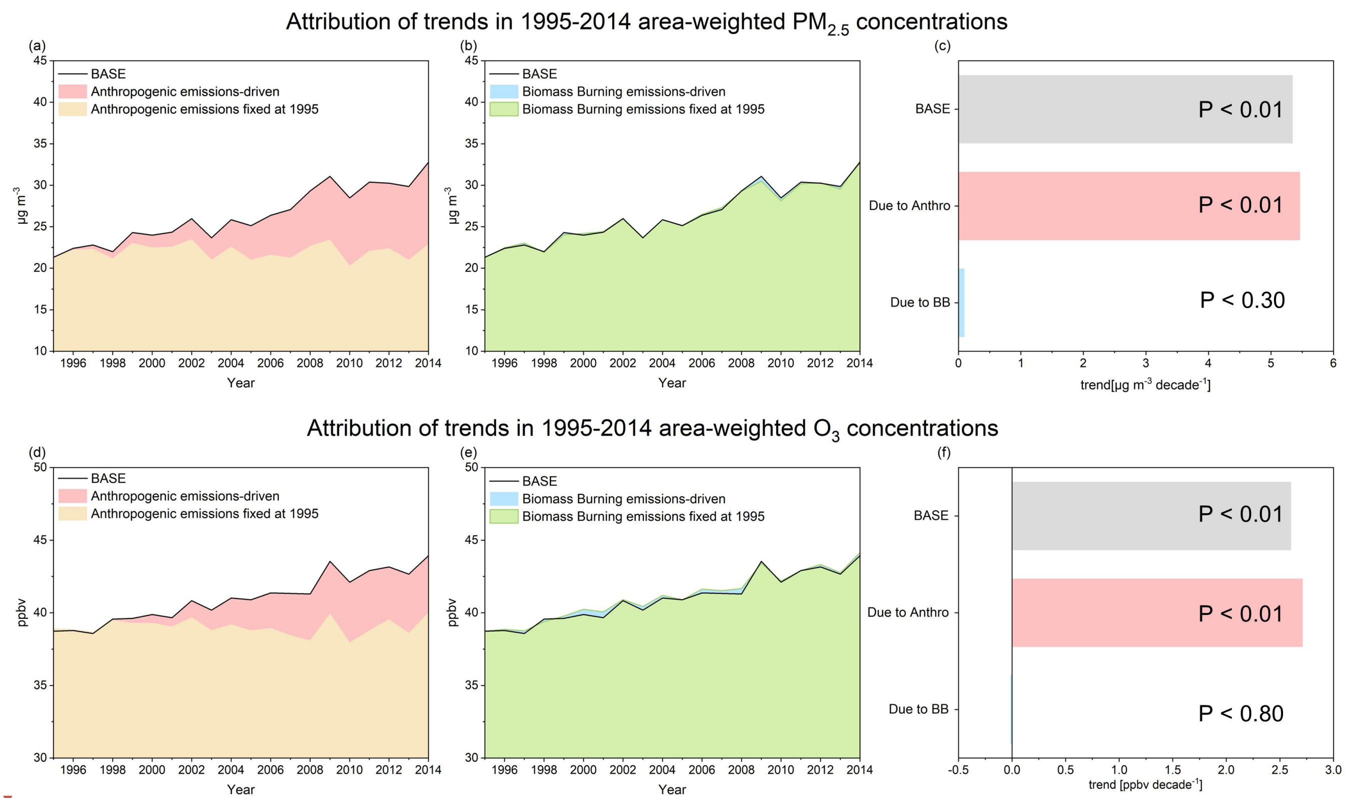

Figure 4. Drivers for trends of area-weighted (a-c) $PM_{2.5}$ and (d-f) O_3 in India in 1995-2014. The yellow shadings in (a, d) showed the evolution of model-simulated $PM_{2.5}$ and O_3 concentrations in the FixAN simulation, with the red shadings illustrating the estimation of the $PM_{2.5}$ and O_3 concentrations resulting from changes in ANTHRO emissions compared to the 1995 level. (b, e) as

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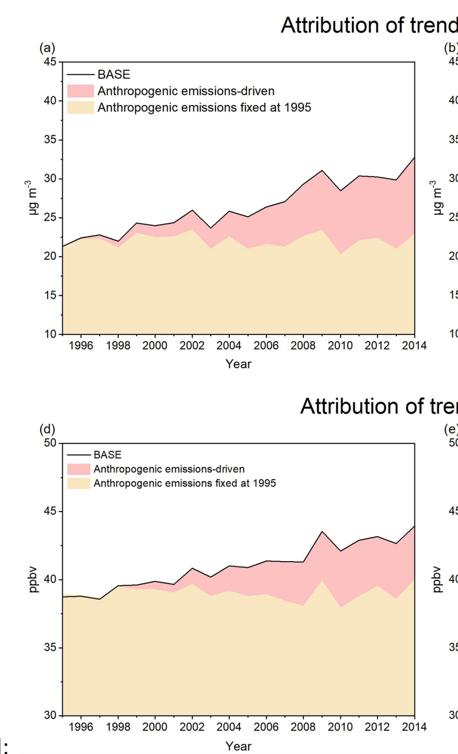

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for (a, d), but for impacts of changes in BB emissions. (c, f) denotes the estimated $PM_{2.5}$ and O_3 trends in India derived from the BASE simulation and impacts of ANTHRO and BB emissions, respectively.

It is well recognized that BB emissions usually featured a distinct seasonal trend, especially in India, where they were influenced by the monsoon. Hence, here we quantified the seasonal trend of $PM_{2.5}$ and O_3 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. From Fig. 5a-h, we found 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 μg m^{-3} $decade^{-1}$ 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 O_3 trend in India had a distinct spatial pattern across seasons (Fig. 5i-p). The ANTHRO emissions had a much stronger positive influence on the O_3 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 O_3 in the presence of NOx (Doherty et al., 2013; Jacob and Winner, 2009; Pusede et al., 2015). The decreased O_3 in the IGP was most pronounced in the DJF season (Fig. 5i), which was mainly attributed to lower solar radiation and titration of O_3 by higher NOx levels (Kumar et al., 2012). Additionally, the occurrence of winter monsoon led to extensive air subsidence in northern India, resulting in low net O_3 production and strong horizontal export, which ultimately leads to relatively low O_3 levels (Lu et al., 2018).

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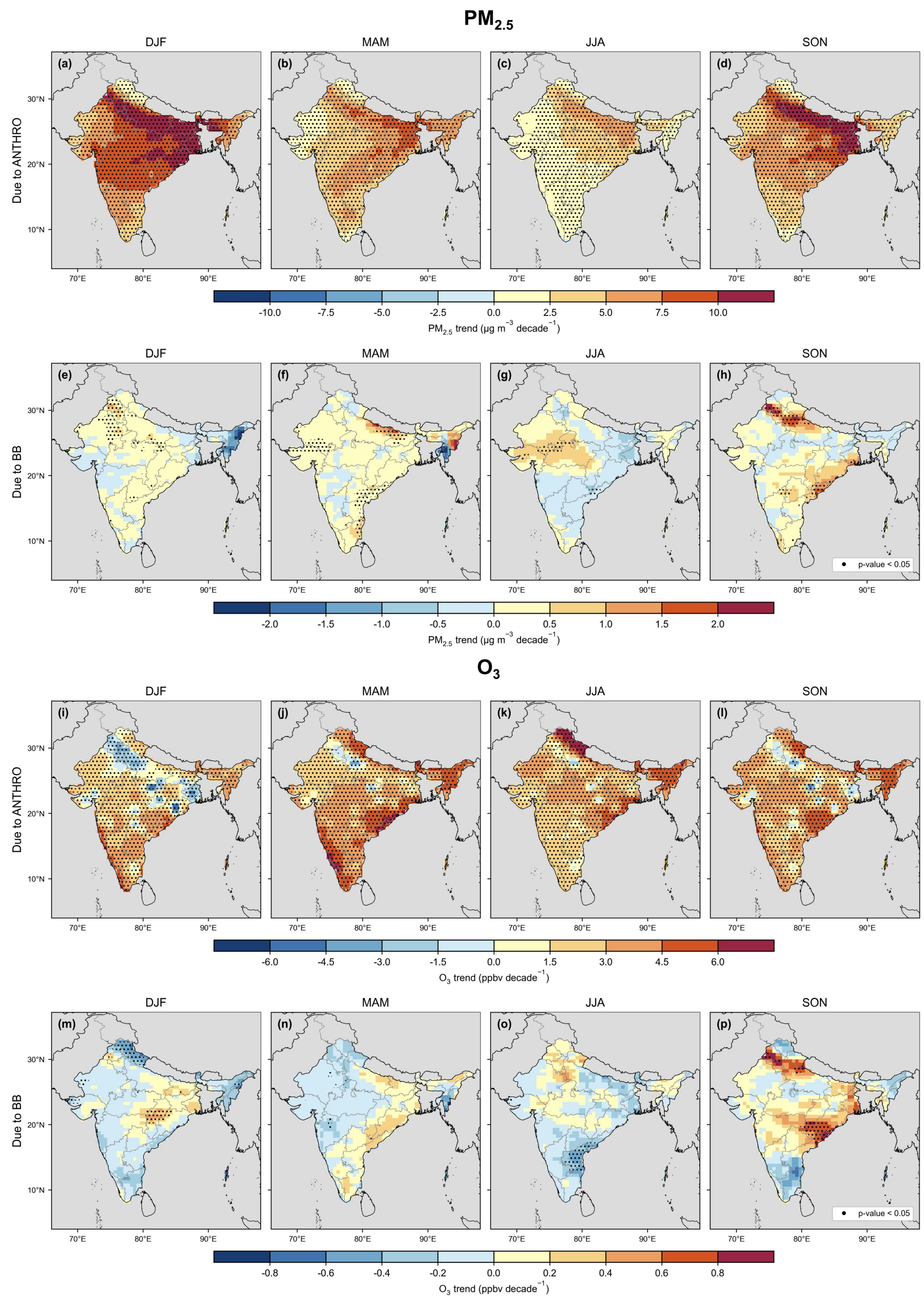

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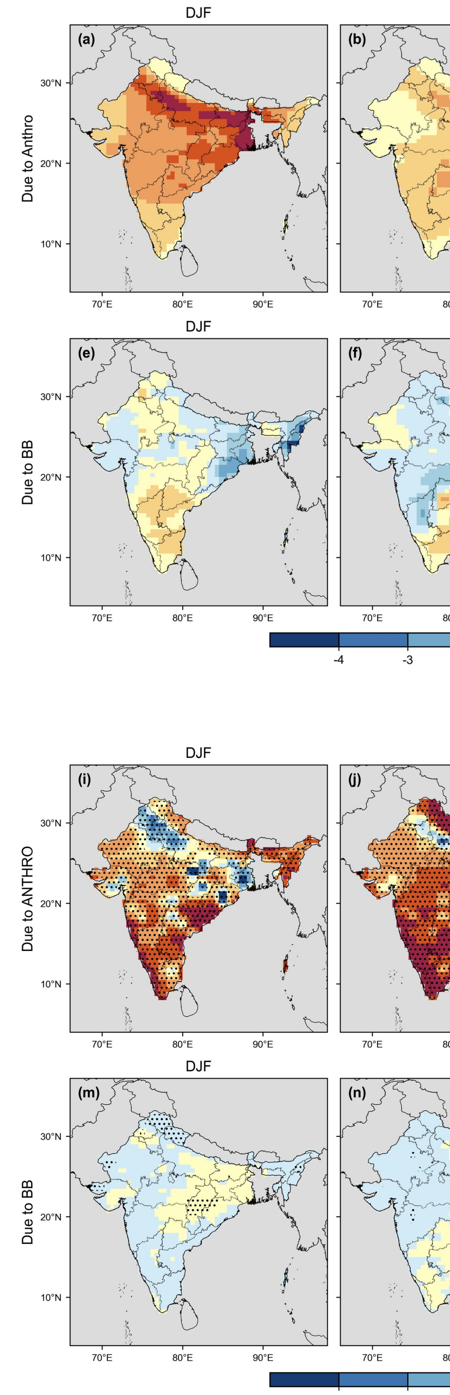
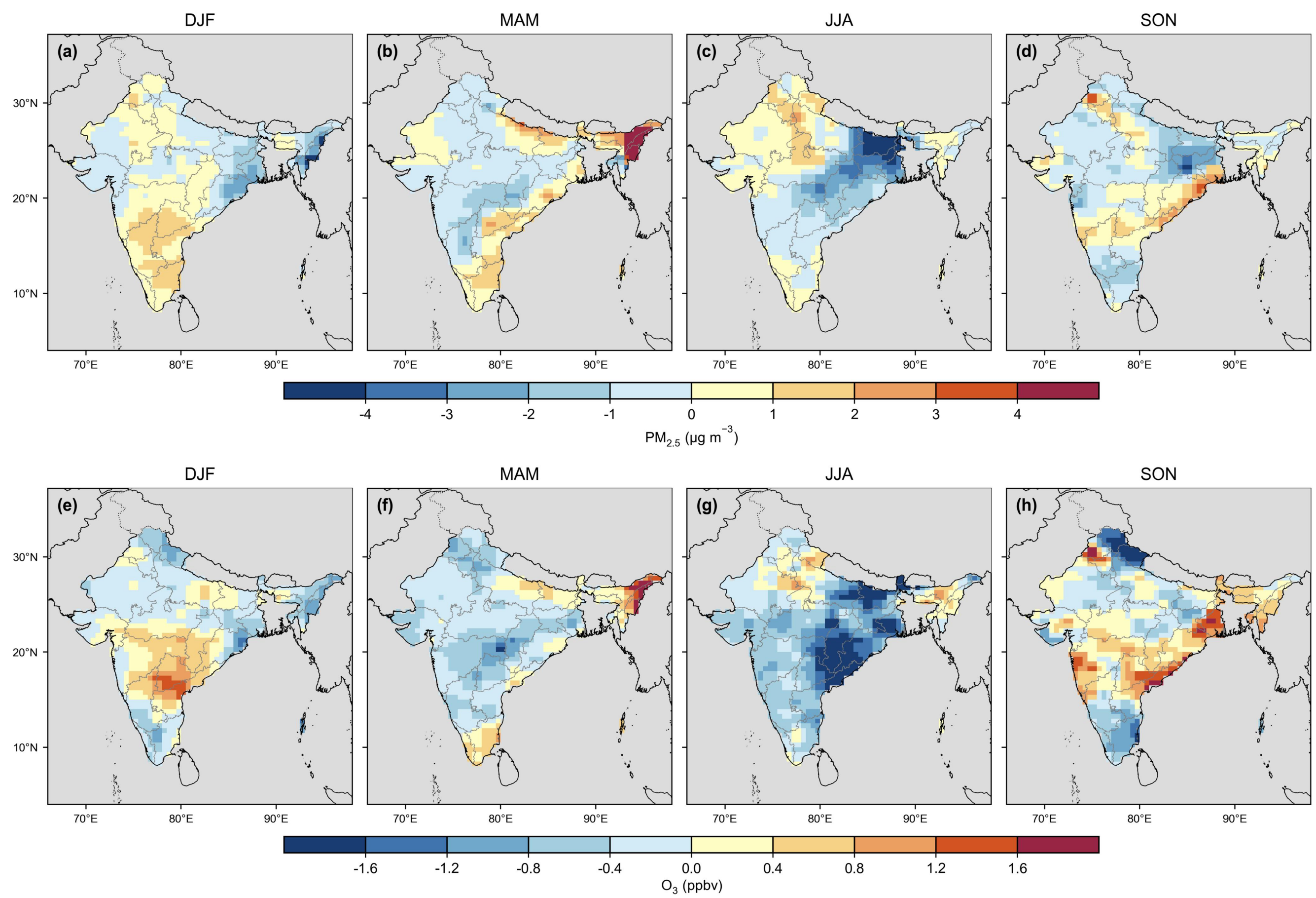


Figure 5. Seasonal patterns of (a-d) ANTHRO and (e-h) BB emissions contributions for the trends of $PM_{2.5}$ in India from 1995 to 2014 and (i-p) for O_3. The units are μg m^{-3} per decade for $PM_{2.5}$ and ppbv per decade for O_3. The dots in the plots indicate statistically significant trends, with p-values less than 0.05.

3.3.2 Contributions to the seasonal air quality changes

Figure 6 showed the spatial distributions of BB contributions for seasonal $PM_{2.5}$ and O_3 changes between 1995 and 2014, respectively, as detailed in Table S4. The changes in BB emissions from 1995 to 2014 contributed significantly to the $PM_{2.5}$ increases in eastern India (over 20 μg m^{-3}) with a high incidence of forest fires (Jena et al., 2015). It also resulted in an increase of O_3 by more than 4 ppbv in eastern India in MAM. Contributions to seasonal $PM_{2.5}$ and O_3 changes from BB were comparable or even exceeding the 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 μg m^{-3} and 6.46 ppbv for $PM_{2.5}$ and O_3, respectively (Fig. S11). Therefore, we concluded that the BB emissions in India posed a great threat to the air quality and thus could not be overlooked.



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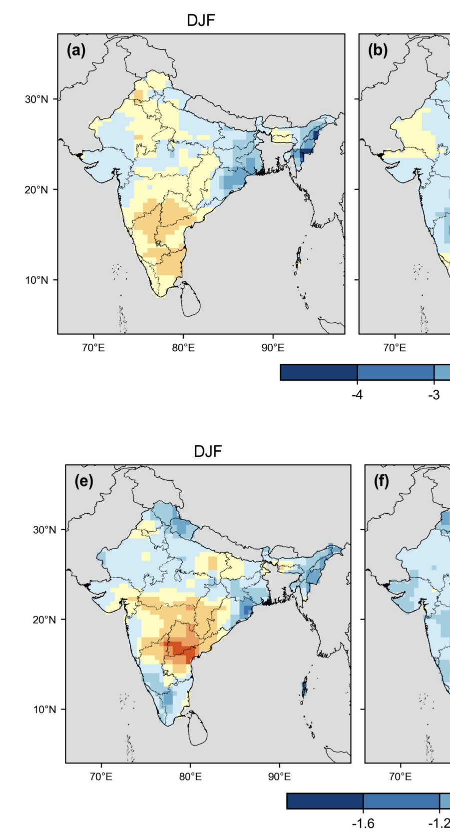

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Figure 6. Spatial distributions of the BB contribution for seasonal (a-d) $PM_{2.5}$ and (e-h) O_3 changes from 1995 to 2014 for DJF, MAM, JJA, and SON. The contributions from BB were calculated as the differences between BASE and FixBB in 2014. The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

3.4 Long-term trends of premature mortality due to $PM_{2.5}$ and O_3 in India

We estimated that the national mortality burden attributable to ambient $PM_{2.5}$ exposure rose significantly from 698.29 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, being 73.91 thousand per decade ($p < 0.01$). We observed that the hotspots of premature mortality attributable to $PM_{2.5}$ and O_3 exposure occurred in New Delhi and IGP regions in 1995 and 2014 (Fig. 7b-e), coincidently with the dense population (Fig. S8). We found that Uttar Pradesh, Bihar, West Bengal, and Haryana, four states within the IGP region, accounted for 41.00% and 39.77% of the national premature mortality due to $PM_{2.5}$ 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 in Jia et al. (2021). Our estimations for the O_3-related mortality burden were 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 O_3-related mortality burden using the GBD2019 metrics, we reported an increasing trend of 29.74 thousand deaths per decade^{-1} for O_3-related mortality, comparable to the GBD2019 estimation of 33.24 thousand deaths per decade^{-1}. However, our estimated mortality burdens were still slightly higher than the GBD2019 due to the O_3 overestimation in our model (Fig. 2 and Fig. S4).

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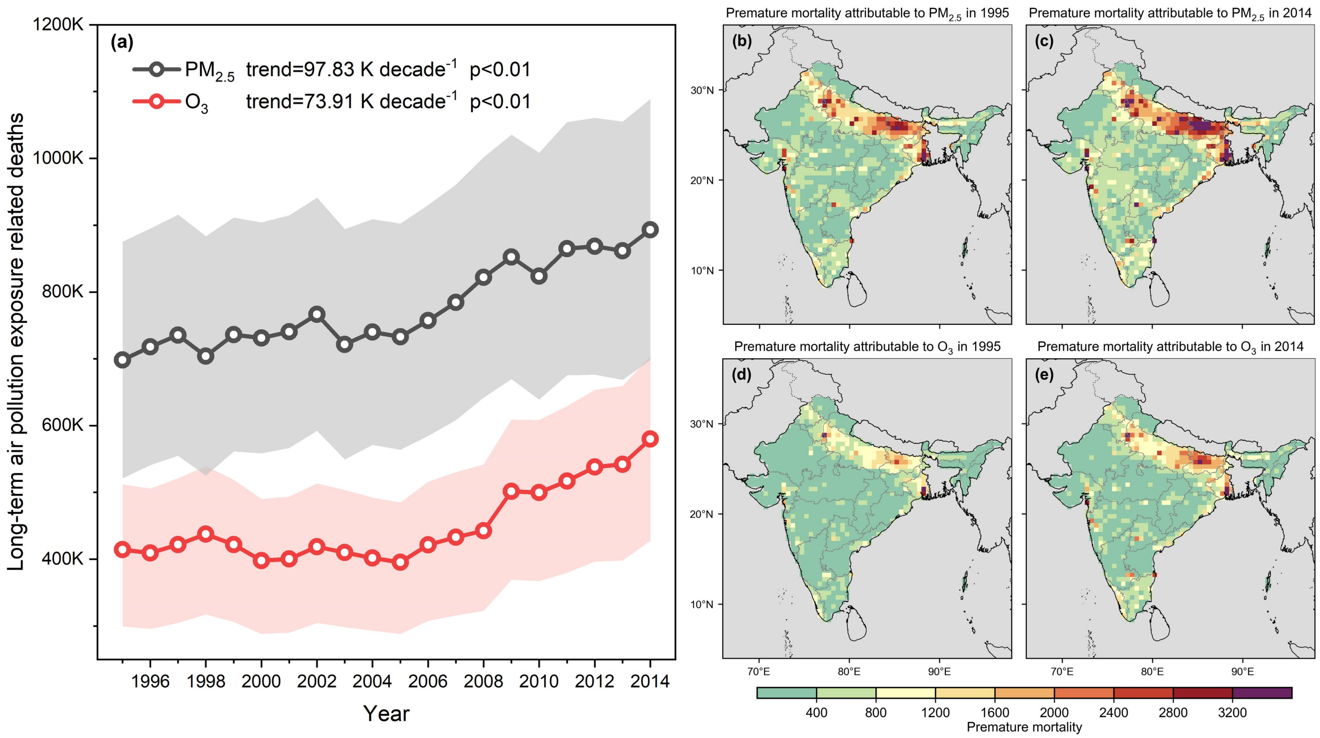
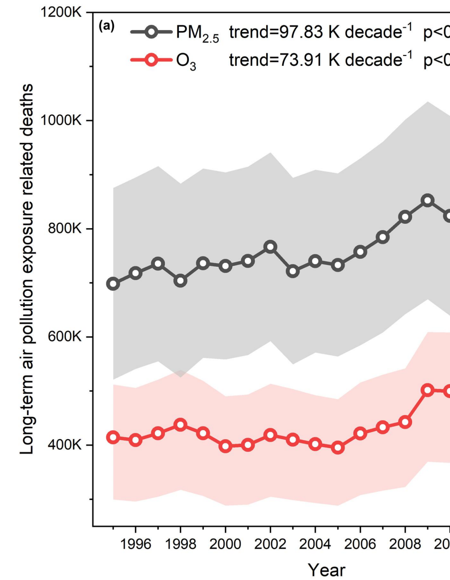


Figure 7. Spatial-temporal change of mortality burden attributable to $PM_{2.5}$ and O_3. (a) interannual variation from 1995 to 2014. The shaded area indicates the range of 95% confidence interval (gray indicates half of the range). (b-e) spatial distributions of the average annual premature mortality attributable to (b-c) $PM_{2.5}$ and (d-e) O_3 in 1995 and 2014.

To isolate the effects of population heterogeneous among regions, we also quantified 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 O_3-attributable premature mortality per capita resembled that of $PM_{2.5}$, but 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 O_3- 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 O_3 in the IGP region was mainly driven by the increased population (Fig. S8).



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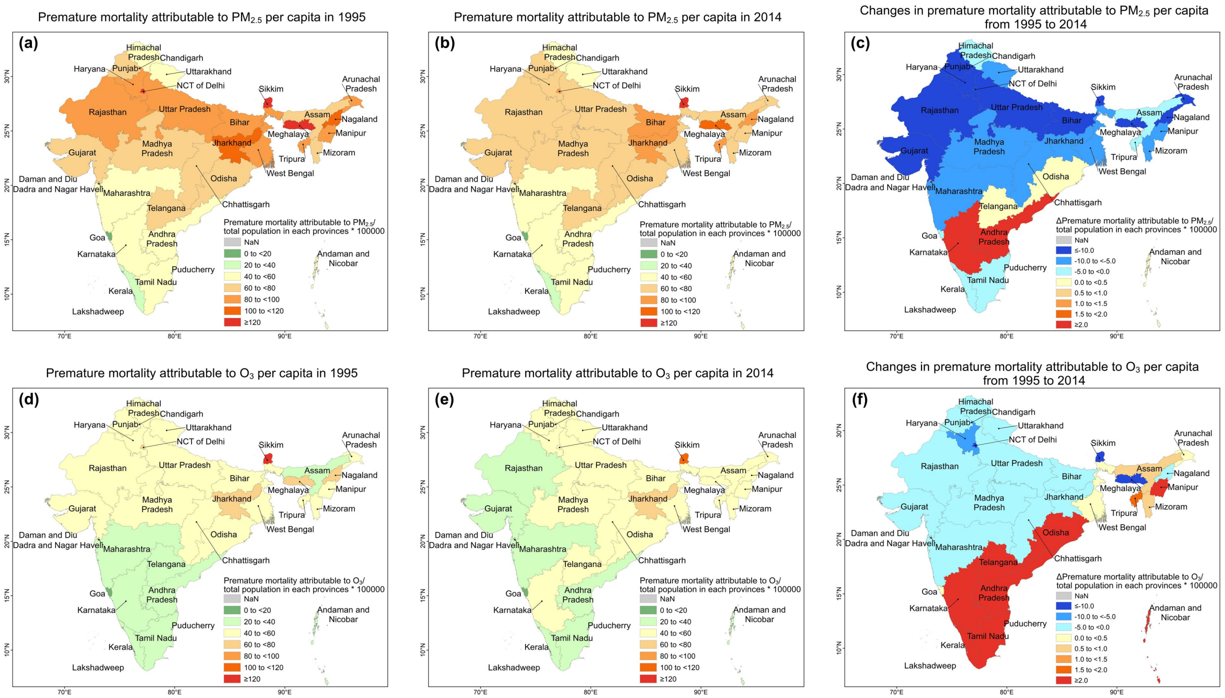

Figure 8. Spatial distributions of premature mortality attributable to $PM_{2.5}$ or O_3 per capita (avoid deaths per 100,000 people) in (a, d) 1995, (b, e) 2014, and (c, f) changes from 1995 to 2014 in the state of India.

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Figure 9 showed 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), which was 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 estimated 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).

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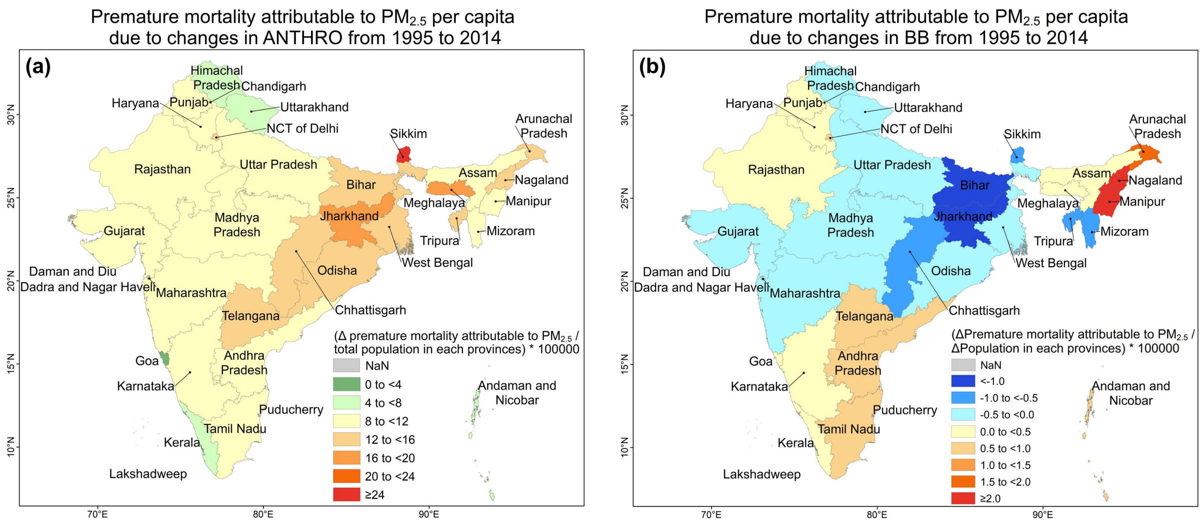

Figure 9. Spatial distributions of contributions to premature mortality attributable to $PM_{2.5}$ per capita (avoided deaths per 100,000 people) from changes in (a) ANTHRO and (b) BB emissions from 1995 to 2014.

4 Conclusions

In this study, we applied 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 O_3 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 μg m^{-3} decade^{-1} ($p < 0.01$) from 1995 to 2014, dominated by the ANTHRO emissions (5.46 μg 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 O_3, 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 found that O_3 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 μg m^{-3} decade^{-1}, $p < 0.30$) or O_3 (-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 μg m^{-3} and 6.46 ppbv for $PM_{2.5}$ and O_3, respectively.

Further estimation of mortality burden showed 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 O_3 (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 revealed a notably higher

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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 suggested that population growth is the primary factor driving the trend of premature mortality.

Our study was subject to several uncertainties and limitations. First of all, the coarser resolution in the global model (0.9° × 1.25°) was frequently found to be unable to realistically represent the complex physical and chemical processes of regional-scale air pollution, especially for O_3 (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 O_3 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 applied the RR derived from a global study instead of India which could potentially have a higher value. Thus, our estimations for the air pollution-related mortality burden could be conservative. More epidemiology studies should be carried out in India to retrieve their own 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.

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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. analyzed 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.

Acknowledgments

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