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Anthropogenic and Natural Causes for the Interannual Variation of $PM_{2.5}$ in East Asia During Summer Monsoon Periods From 2008 to 2018

Danyang Ma[1], Min Xie[1,2,*] , Huan He[1,2], Tijian Wang[3,*] , Mengzhu Xi[1], Lingyun Feng[1], Shuxian Zhang[1], Shitong Chen[1]

[1] School of Environment, Nanjing Normal University, Nanjing 210023, China.

[2] Carbon monitoring and digital application technology center, Carbon peak and carbon neutralization strategy institute of Jiangsu Province, Nanjing 210023, China.

[3] School of Atmospheric Sciences, Nanjing University, Nanjing 210023, China.

Correspondence to: Min Xie(minxie@njnu.edu.cn); Tijian Wang (tjwang@nju.edu.cn)

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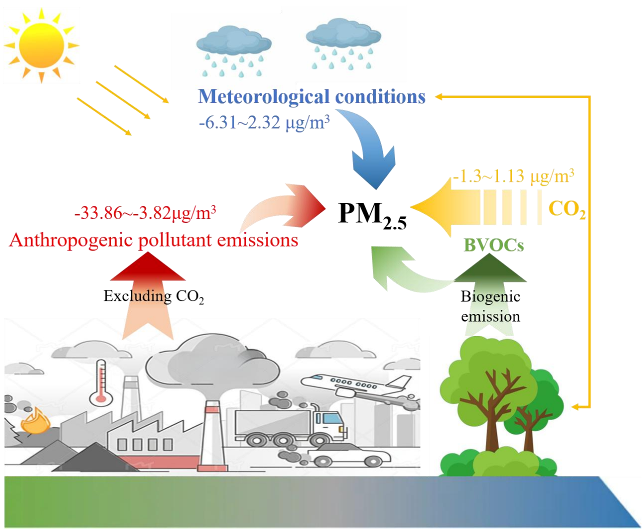
Abstract. There was a significant difference in near-surface $PM_{2.5}$ changes across China after the implementation of the Clean Air Action Plan in 2013. This study used the regional climate-chemistry-ecosystem coupled model, RegCM-Chem-YIBs, to investigate interannual variations in $PM_{2.5}$ across East Asia from 2008 to 2018. The drivers of $PM_{2.5}$ variability were examined from Anthropogenic and Natural perspectives. Compared to 2008, $PM_{2.5}$ showed little variation during 2009–2013 (the PreG phase). However, during 2014-2018 (the PostG phase), a substantial decline in $PM_{2.5}$ was simulated, particularly in the North China Plain (-36.76 μg/m³) and the Sichuan Basin (-33.96 μg/m³). Anthropogenic pollution emissions were the primary drivers of $PM_{2.5}$ reductions, contributing -10.39 to -3.82 μg/m³ in the PreG period and -33.86 to -8.45 μg/m³ in the PostG period. The influence of meteorological conditions on $PM_{2.5}$ during the PreG phase (-6.31 to 2.32 μg/m³) was comparable to that of anthropogenic pollutant emissions. Additionally, in the vegetation-rich region, the impact of CO_2 changes on $PM_{2.5}$ was comparable to that of anthropogenic pollutant emissions. Our study comprehensively examined the drivers of $PM_{2.5}$ concentration changes from 2008 to 2018. We highlight a significant intensification in the contribution of anthropogenic pollutant emissions and reveal that, in regions characterized by dense vegetation, changes in CO_2 concentrations exert a pronounced impact on $PM_{2.5}$ variations.

Graphical Abstract：



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1 Introduction

Fine particulate matter ($PM_{2.5}$) is one of the primary atmospheric pollutants in China(Fontes et al., 2017), posing significant risks to human respiratory health(Feng et al., 2016; Xing et al., 2016). Long-term exposure to $PM_{2.5}$ can lead to respiratory diseases such as chronic bronchitis, emphysema, and asthma(Kim et al., 2015; Pui et al., 2014; Xing et al., 2016). Additionally, $PM_{2.5}$ is critical as a short-lived species influencing atmospheric radiation processes(Hu et al., 2017). It affects the radiative energy balance of the Earth-atmosphere system by scattering or reflecting solar radiation (direct effect)(Wu et al., 2021) and altering cloud microphysical properties (indirect effect)(Wang et al., 2018a; Wu et al., 2021).

With China's rapid economic development, widespread $PM_{2.5}$ pollution became prevalent across the country in the early 21st century(Ma et al., 2016). In the most severely polluted urban areas, the annual average $PM_{2.5}$ concentration exceeded 100 μg/m^3 (Van Donkelaar et al., 2010). From 2000 to 2008, the national average $PM_{2.5}$ concentration in China was 49.4 ± 14.2 μg/m^3. In eastern China, the average concentration was 55.4 ± 16.1 μg/m^3, while the Beijing-Tianjin-Hebei region experienced levels as high as 62.1 ± 22.5 μg/m^3. The Yangtze River Delta saw an average concentration of 63.0 ± 11.1 μg/m^3, the Pearl River Delta recorded 52.4 ± 5.8 μg/m^3, and the Sichuan Basin averaged 61.6 ± 13.4 μg/m^3 (Wei et al., 2021). To mitigate the severe $PM_{2.5}$ pollution, China implemented the Clean Air Action Plan in 2013(Li et al., 2019). This policy led to a significant nationwide decrease in $PM_{2.5}$ concentrations(Zhang et al., 2019), marking a notable improvement in air quality ever since 2013 (Vu et al., 2019; Li et al., 2018).

The variation in $PM_{2.5}$ concentrations is influenced by three key factors: anthropogenic pollutant emissions, meteorological conditions(Xiao et al., 2021), and Carbon dioxide (CO_2) changes. Anthropogenic pollutant emissions encompass industrial production, transportation, and energy consumption(An et al., 2019), which release amounts of primary $PM_{2.5}$, as well as the precursors of secondary $PM_{2.5}$ such as volatile organic compounds (VOCs) (Kurokawa and Ohara, 2020) and nitrogen oxides (NO_x) (Wu et al., 2020; Zheng et al., 2021a; Kurokawa and Ohara, 2020). Consequently, reducing these emissions is essential for mitigating $PM_{2.5}$ concentrations, as they directly contribute to both the formation and persistence of particulate pollution(Zheng et al., 2018; Zhang et al., 2019).

Meteorological conditions play a significant role in influencing near-surface $PM_{2.5}$ concentrations(Chen et al., 2020b; Xiao et al., 2021). Elevated temperatures can accelerate atmospheric chemical reactions(Mousavinezhad et al., 2021), including oxidation and photochemical processes, thereby promoting the formation of $PM_{2.5}$ (Zhong et al., 2018). Precipitation aids in removing particulate matter from the atmosphere through wet deposition(Zhang et al., 2013), effectively reducing $PM_{2.5}$ pollution levels (Wu et al., 2018). Additionally, wind speed and direction are crucial factors in the transport and dispersion of particulate matter (Li et al., 2017). Higher wind speeds facilitate the dispersion of particulate matter over a wider area, reducing its local accumulation and mitigating air pollution in specific regions (Li et al., 2017; Zhang et al., 2018). The increase in planetary boundary layer height (PBLH) strengthens atmospheric upward motion(Ait-Chaalal et al., 2016), thereby reducing near-surface $PM_{2.5}$ concentrations (Pan et al., 2019).

Changes in CO_2 concentrations can influence $PM_{2.5}$ pollution levels through several mechanisms. Firstly, elevated CO_2 concentrations impact the atmospheric radiation balance, altering the distribution and intensity of precipitation(Cao et al., 2012), which directly affects

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$PM_{2.5}$ concentrations by influencing wet deposition rates(Zhang et al., 2022). Additionally, Changes in CO_2 concentrations can affect vegetation photosynthesis and growth, which alter the emissions of biogenic volatile organic compounds (BVOCs) that can participate in atmospheric chemical reactions to form secondary organic aerosols (SOA), and thereby impact atmospheric $PM_{2.5}$ concentrations(Sun et al., 2013; Sun et al., 2012). Isoprene is the most abundant species among BVOCs, so changes in CO_2 concentrations can indirectly affect near-surface $PM_{2.5}$ concentrations by influencing isoprene emissions from vegetation(Sun et al., 2013; Lin et al., 2013; Kramer et al., 2016).

Numerous studies have used statistical models and numerical simulations to investigate the impacts of meteorological conditions and anthropogenic emissions on $PM_{2.5}$ concentration changes. The results consistently indicate that changes in anthropogenic emissions are the primary driver of $PM_{2.5}$ variation. Zhang et al. (2019) using the WRF-CMAQ model at the national scale, found that meteorological conditions accounted for only 9 % of the total decline in $PM_{2.5}$ concentrations during 2013–2017 in China, suggesting that emission reductions were the dominant factor. Similarly, based on a multiple linear regression model, Chen et al. (2020a) reported that anthropogenic emission reductions contributed 73 %, 87 %, and 84 % to the $PM_{2.5}$ decline in the North China Plain, Yangtze River Delta, and Pearl River Delta, respectively, while the contribution of meteorological conditions ranged from 10 % to 26 %. Cheng et al. (2019) employing the WRF-CMAQ model, found that the decrease in $PM_{2.5}$ concentrations in Beijing over the same period was mainly attributable to local (65.4 %) and regional (22.5 %) emission reductions, with meteorological conditions accounting for only 12.1 %.

Current research primarily emphasizes the impact of anthropogenic pollutant emissions(Zheng et al., 2018) and meteorological changes on $PM_{2.5}$ concentrations(Zhang et al., 2019; Zhai et al., 2019), while the potential influence of CO_2 concentration changes on $PM_{2.5}$ pollution levels remains largely underexplored. Additionally, following the implementation of the Clean Air Action Plan in 2013, significant decreases in $PM_{2.5}$ concentrations were observed in China. Concurrently, CO_2 levels continued to rise(Xu et al., 2022), with the influence of CO_2 on $PM_{2.5}$ strengthening annually. Therefore, it is essential to analyze the evolution of $PM_{2.5}$ concentrations from 2008 to 2018 in detail, and attribute changes in $PM_{2.5}$ levels to every factor, such as anthropogenic emissions, meteorological conditions, and CO_2 variations.

2 Methods and data

2.1 Model description

In this study, we employed the coupled regional climate-chemistry-ecology model RegCM-Chem-YIBs (Xie et al., 2019; Xie et al., 2024). The RegCM-Chem component simulates key meteorological variables, including temperature, humidity, precipitation, and radiation, along with atmospheric pollutants including ozone and particulate matter (Shalaby et al., 2012). The YIBs (Yale Interactive Biosphere Simulator) model focuses on simulating vegetation physiological processes, such as ozone-induced damage, photosynthesis, and respiration(Lei et al., 2020). Additionally, it computes important land surface parameters, including CO_2 flux, biogenic volatile organic compound (BVOC) emissions, and stomatal conductance (Yue and Unger, 2015). The RegCM-Chem and YIBs models exchange variables every 6 minutes, facilitating dynamic coupling between regional climate, atmospheric chemistry, and ecosystem processes. The RegCM-Chem-YIBs model simulated both primary and secondary $PM_{2.5}$

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emissions, including dust, black carbon, organic carbon, sulfates, nitrates, and ammonium. The structure of the model is shown in Fig. 1.

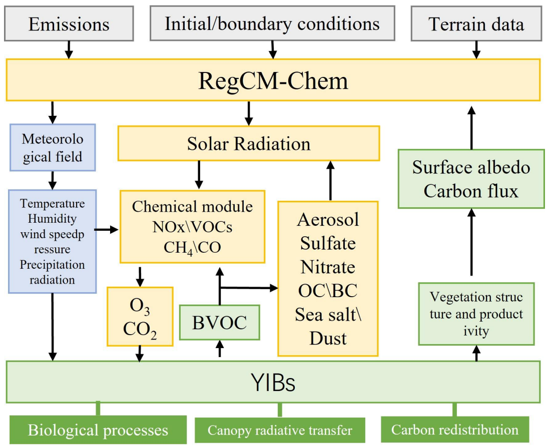

Figure 1. Framework of the RegCM-Chem-YIBs Model.

2.2 Model configurations

The study area covers the entire East Asian region, with a horizontal grid resolution of 60 km, centered at 36°N and 107°E. A terrain-following coordinate system was used vertically(Bleck and Benjamin, 1993), dividing the atmosphere into 18 layers from the surface to 50 hPa.

Anthropogenic pollutant emissions data were obtained from the Multi-resolution Emission Inventory for China (MEIC v1.4) developed by Tsinghua University(Geng et al., 2024). Surface CO_2 flux data were sourced from the National Oceanic and Atmospheric Administration (NOAA) CarbonTracker CT2019 dataset, which includes contributions from fossil fuel combustion, biomass burning, and ocean-atmosphere CO_2 exchange(Peters et al., 2007). Meteorological fields were derived from ERA-Interim reanalysis(Balsamo et al., 2015), while sea surface temperature data were taken from NOAA's weekly mean dataset(Huang et al., 2021). The model employed the Grell cumulus parameterization scheme, CCM3 radiation scheme, Holtslag PBL scheme for boundary layers, CBM-Z mechanism for meteorology and chemistry, and TUV photochemistry scheme.

2.3 Experiment settings

The numerical experiments are presented in Table 1. The SIM_{Base} experiment accounted for interannual variations in meteorological fields, CO_2 emissions, and anthropogenic pollutant emissions (excluding CO_2) from 2008 to 2018. Additionally, the $SIM_{MET=2008}$ and $SIM_{CO2=2008}$ experiments were designed, where meteorological fields and CO_2 emissions were fixed at their 2008 levels, respectively, while simulations were conducted for 2009–2018. The simulation period spans from April to August each year. Among them, the results from May to August, corresponding to the East Asian Summer Monsoon (EASM) period, were selected for analysis.

Changes in $PM_{2.5}$ concentrations were attributed to three main factors: anthropogenic pollution emissions, meteorological conditions, and CO_2 variations. By comparing the simulation results from different years in the SIM_{Base} experiment to those from 2008, we quantified changes

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in $PM_{2.5}$ concentrations relative to 2008 for the period 2009–2018. To evaluate the impact of meteorological conditions on $PM_{2.5}$ concentrations, we compared the results of the SIM_{Base} experiment with those of the $SIM_{MET=2008}$ experiment for the same year (SIM_{Base} - $SIM_{MET=2008}$). Similarly, the contribution of CO_2 changes to $PM_{2.5}$ variations was assessed by comparing the SIM_{Base} experiment with the $SIM_{CO2=2008}$ experiment (SIM_{Base} - $SIM_{CO2=2008}$). The contribution of anthropogenic pollutant emissions was then determined by subtracting the effects of meteorological and CO_2 changes from the total $PM_{2.5}$ variation.

The numerical experiments in this study follow the same design(Ma et al., 2023a) as in previous research. Therefore, the detailed parameterization schemes, settings, and model validation are provided in earlier publications and will not be repeated here. Overall, the R model effectively simulates meteorological factors, $PM_{2.5}$, and CO_2 concentrations in East Asia(Ma et al., 2023a; Ma et al., 2023b).

Table 1. The Numerical experimental in this study

Experiment	Time	Meteorological fields	CO_2 emissions	Anthropogenic pollutant emissions
SIM_{Base}	2008-2018	Varying	Varying	Varying
$SIM_{MET=2008}$	2009-2018	2008	Varying	Varying
$SIM_{CO2=2008}$	2009-2018	Varying	2008	Varying

3 Results and discussion

3.1 $PM_{2.5}$ variation

Figure 2 illustrates the changes in near-surface $PM_{2.5}$ concentrations across East Asia from 2009 to 2018. $PM_{2.5}$ concentrations are notably higher in the North China Plain, northeastern China, and eastern China (Shanghai, Jiangsu, Zhejiang), largely driven by industrial emissions, vehicle exhaust, coal combustion, and dust from human activities(Wang et al., 2017). In contrast, regions in western China (Yunnan, Gansu, Xinjiang) exhibit lower $PM_{2.5}$ levels due to limited industrial activity, lower population density, and more favorable meteorological conditions(Wei et al., 2021; Xue et al., 2020). Developed cities and industrial centers like the Pearl River Delta and Fuzhou (Fujian Province) continue encountering challenges related to $PM_{2.5}$ pollution. Moreover, the Sichuan region, characterized by its enclosed basin geography and high population density, also experiences high $PM_{2.5}$ pollution levels(Wang et al., 2018b). From 2009 to 2013, $PM_{2.5}$ concentrations in China remained relatively stable, with levels averaging around 90 μg/m³ in the North China Plain and Sichuan Basin. However, following the implementation of the Clean Air Action Plan in 2013, $PM_{2.5}$ levels significantly declined nationwide. By 2018, concentrations had dropped to below 50 μg/m³ across much of the country.

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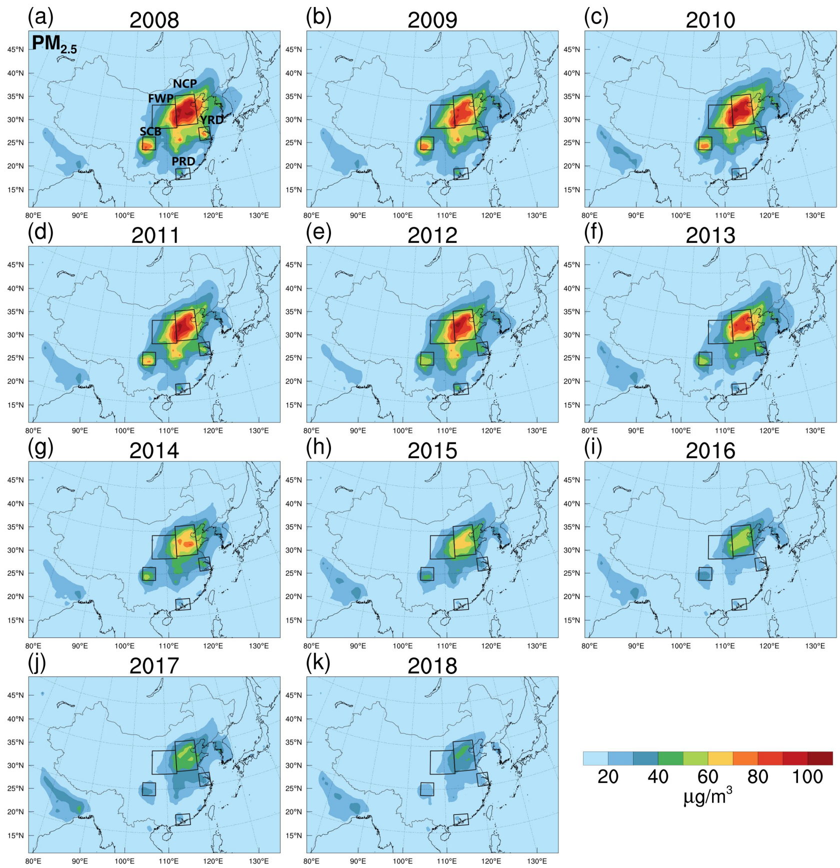

Figure 2. Near-surface $PM_{2.5}$ concentrations (μg/m³) over East Asia during May to August from 2009 (a) to 2018 (k). Key regions are highlighted by black boxes, including the North China Plain (NCP), Fenwei Plain (FWP), Yangtze River Delta (YRD), Pearl River Delta (PRD), and Sichuan Basin (SCB).

Figure 3 and Table S1 present the changes in $PM_{2.5}$ concentrations relative to 2008 across East Asia from 2009 to 2018. Since 2008, most regions in China have seen varying degrees of $PM_{2.5}$ reduction. During the pre-governance period (PreG, 2009~2013), the largest decrease occurred in the Yangtze River Delta, with a reduction of 14.77 μg/m³, followed by the Sichuan Basin and Pearl River Delta, where concentrations dropped by 10.59 μg/m³ and 8.69 μg/m³, respectively. In contrast, the Fenwei Plain and Pearl River Delta experienced smaller changes, with reductions of less than 3 μg/m³. $PM_{2.5}$ concentrations across China significantly decreased after the implementation of the Clean Air Action Plan in 2013. The most notable reductions were simulated in the North China Plain and Sichuan Basin, where $PM_{2.5}$ concentrations dropped by 36.76 μg/m³ and 33.96 μg/m³, respectively. In the Fenwei Plain and Yangtze River Delta, $PM_{2.5}$ concentrations decreased by 22.16 to 27.89 μg/m³. In contrast, the Pearl River Delta saw a smaller reduction, with levels decreasing by just 8.03 μg/m³. This may be attributed to the region's significant influence from the summer monsoon and relatively lower impact from anthropogenic pollution emissions. Further analysis of these factors will be conducted in subsequent sections.

Overall, before 2013, near-surface $PM_{2.5}$ concentrations across China showed little variation.

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However, after 2013, a significant reduction in $PM_{2.5}$ pollution levels was simulated nationwide. Changes in $PM_{2.5}$ concentrations were attributed to three main factors: anthropogenic pollution emissions, meteorological conditions, and CO_2 variations. The following sections analyze each factor's contribution to the changes in $PM_{2.5}$ concentrations from 2008 to 2018.

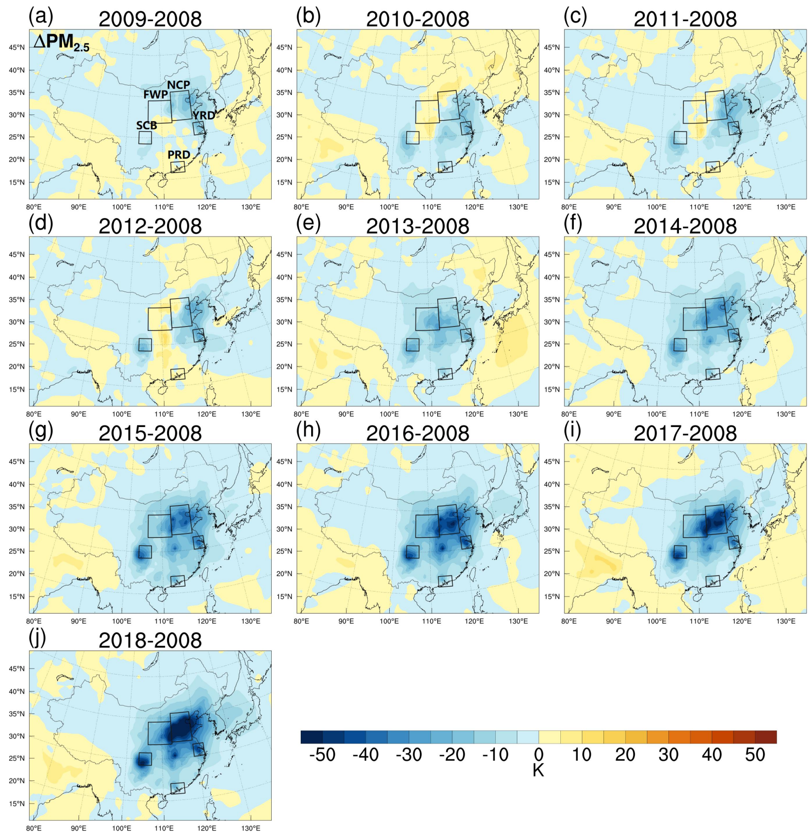

Figure 3. Changes in near-surface $PM_{2.5}$ concentrations (μg/m³) during May to August from 2009 (a) to 2018 (j) relative to 2008 in East Asia.

3.2 Contribution of meteorological conditions

As shown in Fig. 4, during the PreG period, the precipitation increased by 2–4 mm/day in China's eastern coastal and western inland regions, while it decreased by approximately 2 mm/day in central China. This increase in precipitation facilitates the reduction of near-surface $PM_{2.5}$ concentrations through wet deposition. Consequently, trends in $PM_{2.5}$ concentrations are inversely related to precipitation: concentrations decreased by 2–16 μg/m³ in the eastern coastal and western inland regions, while increased by 4–8 μg/m³ around 110°E in central China. Additionally, in northeastern and southwestern China, wind speeds increased by 1 to 2 m/s, contributing to the reduction of $PM_{2.5}$ concentrations. In contrast, decreased wind speeds in southeastern and central China facilitated the accumulation of $PM_{2.5}$. During the PostG period, the significant increase in temperature (Fig. 4l) promoted the formation of $PM_{2.5}$, leading to an

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expansion of the areas where $PM_{2.5}$ concentrations increased. Overall, $PM_{2.5}$ concentrations have decreased in the eastern coastal and western inland regions but increased in the central area of China.

Table S2 indicates that in the NCP region, precipitation increased by 0.58 to 0.6 mm/day, and wind speed rose by 0.17 to 0.26 m/s during the PreG and PostG periods, resulting in a decrease in near-surface $PM_{2.5}$ concentrations of 1.6 to 4.01 μg/m³. In the FWP region, $PM_{2.5}$ concentrations increased by 1 to 2.31 μg/m³, which was associated with a rise in temperature of 0.1 to 0.46 K and a significant decrease in PBLH of 108.5 to 15.3 m. In the YRD region, the increase in wind speed of 0.48 to 1.02 m/s facilitated a reduction in $PM_{2.5}$ concentrations by 0.43 to 0.61 μg/m³. Conversely, in the PRD region, reduced precipitation combined with increased temperature contributed to an increase in $PM_{2.5}$ concentrations, ranging from 0.11 to 1.49 μg/m³. In the SCB region, $PM_{2.5}$ concentrations rose by 0.29 μg/m³ during the PreG period, linked to a significant decrease in PBL height of 136.5 m. In the PostG period, $PM_{2.5}$ concentrations decreased by 1.14 μg/m³, attributed to an increase in precipitation (0.37 mm/day) and a decrease in temperature (0.14 K).

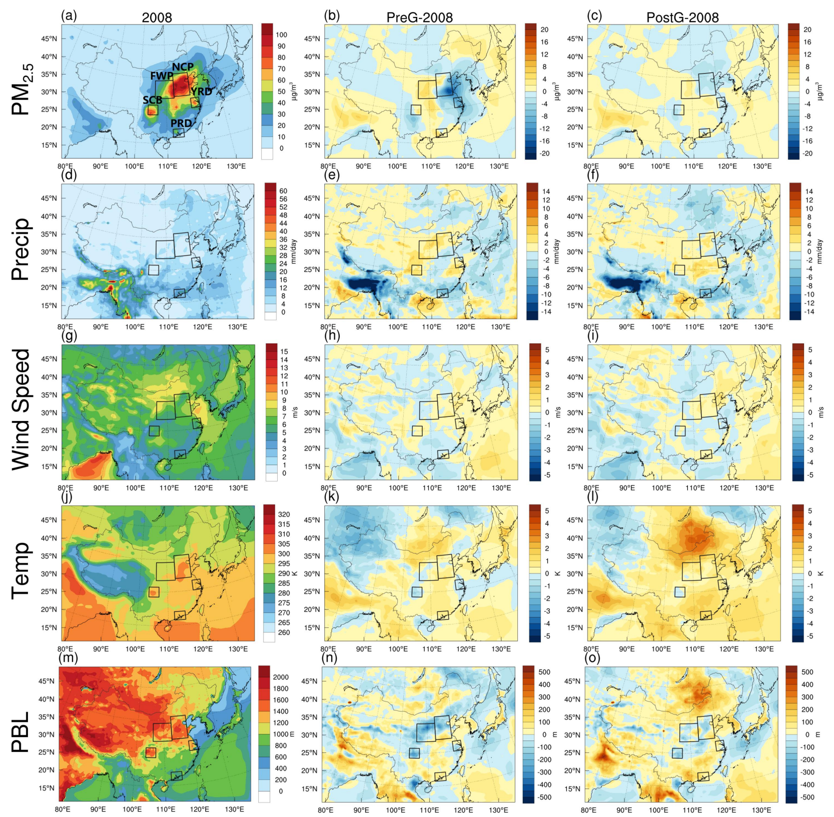

Figure 4. Impact of changes in meteorological conditions on MDA8 O_3 in East Asia during May to August from 2009 (a) to 2018 (j).

3.3 Contribution of CO_2

As shown in Fig. 5, Following the ongoing urbanization and industrialization, CO_2

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concentrations across East Asia rose by 2–10 ppm during both the PreG and PostG periods, with a sharper increase in the PostG period. CO_2 influences atmospheric $PM_{2.5}$ concentrations both through its radiative effects on precipitation and by altering BVOCs emissions from vegetation. Overall, CO_2 changes contributed to $PM_{2.5}$ variations across East Asia from 2008 to 2018, ranging from -4 to 6 μg/m³. $PM_{2.5}$ pollution levels generally increased in the PreG period, while reductions were more common in the PostG period.

Table S3 presents a detailed analysis of the five target regions. In northern China, particularly the NCP and FWP regions, limited vegetation coverage means CO_2 impacts surface $PM_{2.5}$ concentrations mainly through precipitation changes. In the PostG period, precipitation increased by 0.06–0.13 mm/day, lowering $PM_{2.5}$ concentrations by 0.98–1.3 μg/m³. Similarly, in the Sichuan Basin, precipitation rose by 0.21–0.64 mm/day, reducing $PM_{2.5}$ concentrations by 0.49–0.73 μg/m³ in the PreG and PostG period. However, in the YRD and PRD regions, where vegetation coverage is higher, CO_2 primarily impacts $PM_{2.5}$ concentrations by modulating BVOCs emissions. The impact can be either positive or negative(Possell et al., 2005), depending primarily on the balance between the inhibitory effects on synthase activity and the stimulatory effects of enhanced photosynthesis(Wilkinson et al., 2009). In the YRD region, isoprene fell by 0.32–0.58 μg/m³ during both periods, while precipitation rose by 0.09–0.13 mm/day, collectively reducing $PM_{2.5}$ by 0.02–0.05 μg/m³. In the PRD region, isoprene concentrations increased significantly by 0.31–0.92 μg/m³, while precipitation decreased by 0.33–1.02 mm/day. Consequently, $PM_{2.5}$ concentrations rose by 0.31–1.13 μg/m³ during both the PreG and PostG periods.

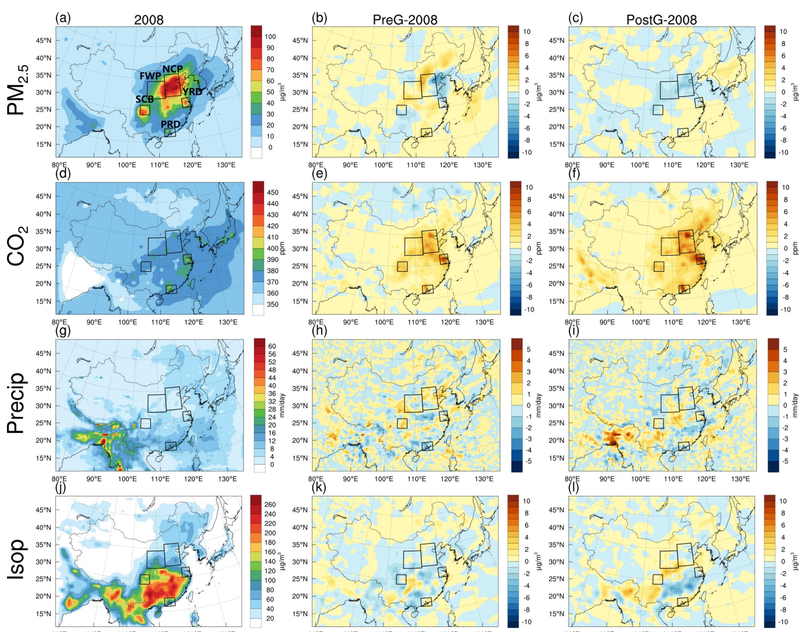

Figure 5. The impact of CO_2 concentration changes on $PM_{2.5}$ (μg/m³), CO_2 (ppm), precipitation (mm/day), and isoprene (μg/m³).

3.4 Contribution of anthropogenic pollutant emissions

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Figure 6 illustrates a significant downward trend in $PM_{2.5}$ concentrations across East Asia since 2008. During the PreG period, $PM_{2.5}$ levels decreased by 5 to 10 μg/m³. Following the implementation of the Clean Air Action Plan in 2013, a marked reduction in $PM_{2.5}$ concentrations was simulated. The most substantial decreases occurred in the NCP and SCB region, with approximately 60 μg/m³. Anthropogenic pollutant emissions emerged as the primary drivers of this decline, with their spatial distribution and magnitude of impact closely corresponding to the overall changes in $PM_{2.5}$ concentrations. In contrast, the effects of changing meteorological conditions and CO_2 concentrations on $PM_{2.5}$ levels in East Asia were relatively minor, ranging between -5 to 5 μg/m³. Meteorological conditions have reduced $PM_{2.5}$ concentrations in the eastern coastal and western regions of China, while increasing them in the central region. In the PostG period, the extent of $PM_{2.5}$ concentration increases has expanded. The impact of CO_2 concentration changes on $PM_{2.5}$ levels shows different trends in the PreG and PostG periods. In the PreG period, changes in CO_2 concentrations primarily led to an increase in $PM_{2.5}$ concentrations. However, in the PostG period, the rise in CO_2 concentrations began to have a negative impact, leading to a reduction in $PM_{2.5}$ concentrations.

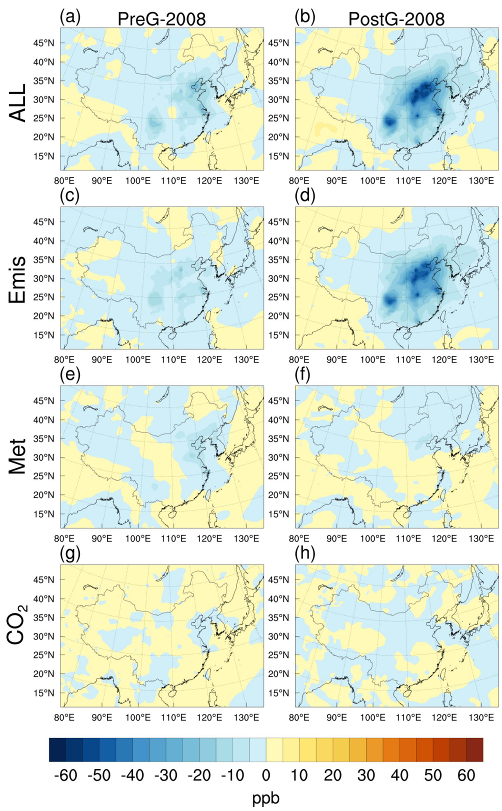

Figure 6. The changes in $PM_{2.5}$ in the East Asia region relative to 2008 (All), and the changes in $PM_{2.5}$ attributed to variations of anthropogenic pollutant emissions (Emis), meteorological conditions (Met), and CO_2 concentrations (CO_2).

Based on Fig. 7 and Table S4, $PM_{2.5}$ concentrations in the NCP region decreased by 5.28 μg/m³ during the PreG period and by 33.86 μg/m³ in the PostG period. Anthropogenic pollution

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emissions were the primary driver of these changes. During the PreG period, the influence of meteorological conditions on $PM_{2.5}$ was comparable to that of anthropogenic pollution emissions, with changes in meteorology contributing -4.01 μg/m³ and emissions contributing -5.28 μg/m³. However, in the PostG period, the impact of meteorological factors diminished to -1.6 μg/m³, indicating that anthropogenic pollution emissions became the predominant factor in the reduction of $PM_{2.5}$ concentrations. In contrast, the effect of changes in CO_2 concentrations on $PM_{2.5}$ levels was relatively minor, ranging from -1.3 to 0.6 μg/m³.

The situation in the FWP region is similar to that of the NCP region, with anthropogenic emissions as the primary driver of reduced $PM_{2.5}$ concentrations. During the PreG and PostG periods, the contributions of anthropogenic emissions to $PM_{2.5}$ levels were -5.75 μg/m³ and -22.18 μg/m³, respectively. In contrast, meteorological conditions contributed to an increase in $PM_{2.5}$ concentrations, with a contribution of 2.32 μg/m³ in the PreG period, comparable to the impact of anthropogenic emissions. Meanwhile, the influence of CO_2 concentrations on $PM_{2.5}$ levels was relatively minor.

In the YRD region, anthropogenic emissions are the primary driver of reduced $PM_{2.5}$ concentrations. Due to its location in eastern China, the YRD region is more affected by the EASM, resulting in more pronounced effects of changing meteorological conditions on $PM_{2.5}$ levels compared to the NCP and FWP regions. During the PreG period, the impact of meteorological conditions on $PM_{2.5}$ concentrations reached as high as -6.31 μg/m³.

In the PRD region, changes in anthropogenic pollution emissions have contributed to a reduction in $PM_{2.5}$ concentrations, ranging from -8.45 to -3.82 μg/m³. However, changes in meteorological conditions and CO_2 concentrations have led to increases in $PM_{2.5}$ levels, ranging from 0.11 to 1.49 μg/m³. Similar to the YRD region, the effects of changing meteorological conditions on $PM_{2.5}$ concentrations are significant, peaking at 1.49 μg/m³ during the PreG period. Located in southeastern coastal China, the Pearl River Delta's rich vegetation cover enhances the impact of CO_2 changes on $PM_{2.5}$ concentrations. During the PreG period, the influence of CO_2 concentration changes on $PM_{2.5}$ levels reached 1.13 μg/m³, comparable to the effect of anthropogenic emissions (-3.82 μg/m³). In the PostG period, the impact of CO_2 changes (0.31 μg/m³) surpassed that of meteorological conditions (0.11 μg/m³).

In the SCB region, the basin topography results in relatively minor effects of meteorological conditions and CO_2 concentration changes on $PM_{2.5}$ levels, with contributions ranging from -1.14 to 0.29 μg/m³ during both the PreG and PostG periods. In contrast, anthropogenic emissions are the primary drivers of reduced $PM_{2.5}$ concentrations, exerting an impact of -32.09 μg/m³ during the PostG period.

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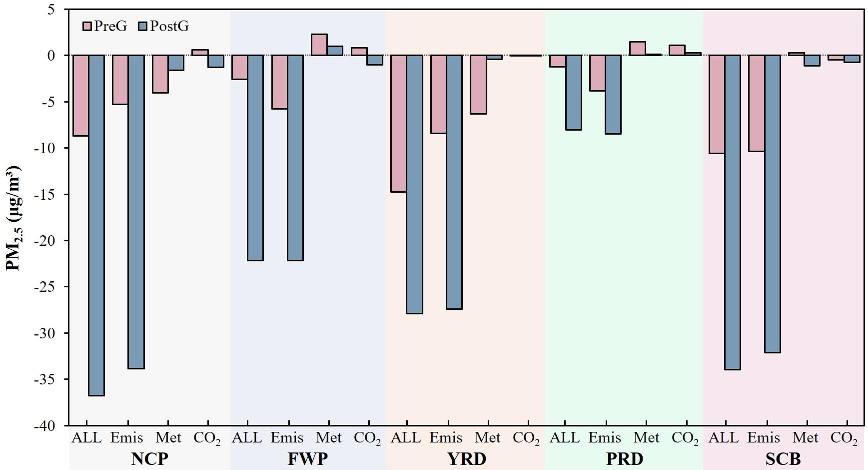

Figure 7. The total changes in $PM_{2.5}$ concentrations (All) for the North China Plain (NCP), Fenwei Plain (FWP), Yangtze River Delta (YRD), Pearl River Delta (PRD), and Sichuan Basin (SCB) relative to 2008, along with the variations in $PM_{2.5}$ due to anthropogenic pollutant emissions (Emis), meteorological conditions (Met), and CO_2 concentration (CO_2) changes.

3.5 Attribution of Changes in $PM_{2.5}$

Figure 8 illustrates that $PM_{2.5}$ concentrations remained relatively stable across the five regions during the PreG period. However, in the PostG period, following the implementation of the Clean Air Action Plan, significant reductions in $PM_{2.5}$ concentrations were simulated in the NCP, FWP, YRD, and SCB regions, while the PRD region showed the smallest decrease.

Anthropogenic pollutant emissions are the primary factor driving $PM_{2.5}$ concentration reductions across the five regions, with their impact increasing linearly over time. In the PreG period, meteorological conditions had a relatively stronger influence on $PM_{2.5}$ levels, occasionally surpassing the effects of anthropogenic emissions. For example, in 2013, the meteorological and emission impacts on $PM_{2.5}$ in the NCP region were -17.35 μg/m^3 and 4.49 μg/m^3, respectively. Similarly, in the FWP region from 2013 to 2015, meteorological impacts ranged from -16.9 to -15.36 μg/m^3, while emissions affected $PM_{2.5}$ concentrations between -15.8 and -2.27 μg/m^3. The influence of meteorology also exceeded that of emissions in the YRD region during 2011–2012 and in the PRD region in 2010. Even in the SCB region, where meteorological impacts on $PM_{2.5}$ were relatively minor, meteorological effects in 2010 (8.59 μg/m^3) were comparable to emissions (-14.67 μg/m^3).

The influence of CO_2 concentration changes on $PM_{2.5}$ levels was generally minor but, in the densely vegetated PRD region, could be comparable to the effects of emissions and meteorology. The influences of CO_2 concentration, emissions, and meteorology on $PM_{2.5}$ are -0.25 to 3.11 μg/m^3, -6.19 to -1.47 μg/m^3, and -0.5 to 3.11 μg/m^3, respectively from 2009 to 2013.

Our attribution analysis of $PM_{2.5}$ concentration changes is mainly consistent with previous studies, which have indicated that variations in anthropogenic emissions were the primary driver of $PM_{2.5}$ changes in China during 2013-2017, with meteorological conditions contributing approximately 9 %-26 % (Zhang et al., 2019; Chen et al., 2020a; Cheng et al., 2019). In our study, relative to 2008, the average contribution of anthropogenic emissions during the PreG period was 89.08 %, while meteorological conditions contributed 16.45 %. In the PostG period, following the implementation of the Clean Air Action Plan, the influence of anthropogenic emissions further increased to 96.26 %, whereas the contribution of meteorological conditions declined to 1.60 %. This finding underscores that the impact of changes in anthropogenic emissions on $PM_{2.5}$ concentrations was markedly enhanced after 2013. Notably, changes in CO_2 concentrations had a significant impact on $PM_{2.5}$ levels, contributing -5.46 % during the PreG period and 2.14 % during the PostG period, with the latter effect surpassing that of meteorological conditions.

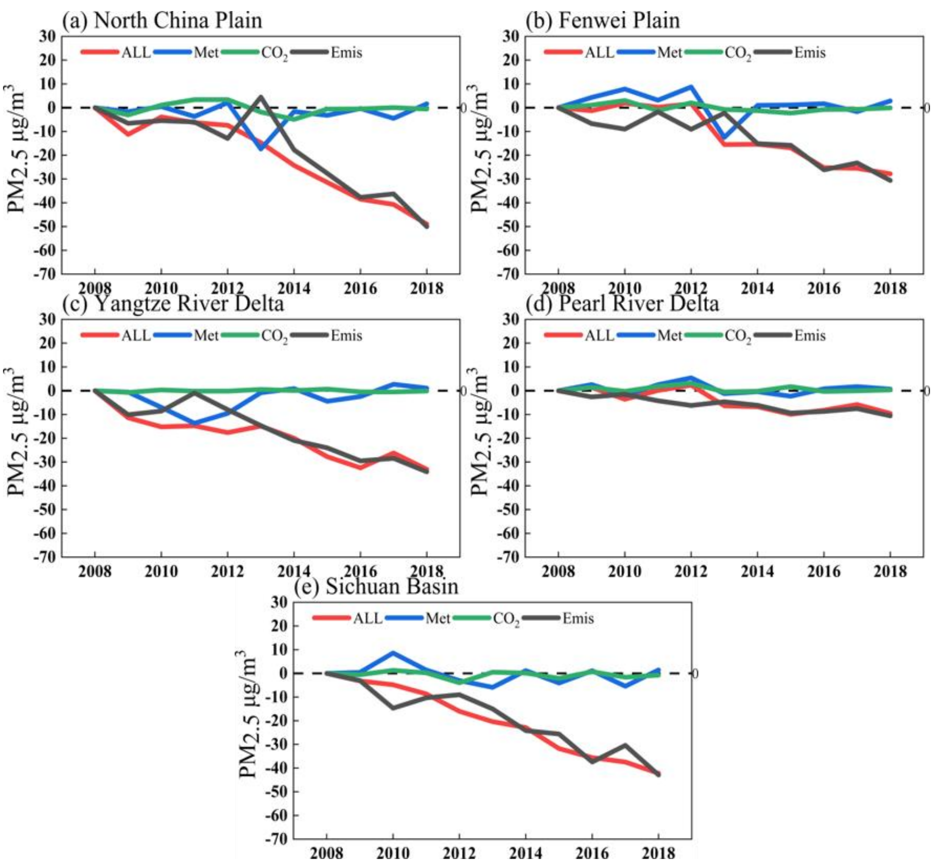

Figure 8. Changes in $PM_{2.5}$ concentrations in East Asia from 2008 to 2018 (ALL) and the contributions of anthropogenic pollutant emissions (Emis), meteorological conditions (Met), and CO_2 concentration changes (CO_2) to $PM_{2.5}$ concentrations by Region (Units: μg/m^3).

3.6 Uncertainties

The uncertainties in the MEIC emission inventory primarily arise from activity data, emission factors, spatial and temporal allocation methods, and the implementation status of pollution control measures (Hong et al., 2017; Zheng et al., 2021b), all of which may affect the accuracy of simulation results. Future improvements can be achieved by employing more refined and

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accurate emission inventories.

In addition, the use of a 60 km low-resolution grid limits the ability to represent local topography and physical processes, thereby introducing simulation errors (Harris et al., 2016; Ringler et al., 2013). Given that this study employs a fully coupled regional climate-chemistry-ecology model with extended simulation periods (three sets of 10-year simulations) and a broad regional scope (covering the entire East Asia region), computational resource constraints necessitated the use of 60 km grids. Numerous studies have employed the RegCM-Chem-YIBs model at a 60 km grid resolution to systematically analyze $PM_{2.5}$, O_3, CO_2, and the regional climate over East Asia (Ma et al., 2023a; Ma et al., 2023b; Xu et al., 2023; Gao et al., 2021). These demonstrate its robustness and reliability in simulating East Asian atmospheric and climatic conditions. Future studies could enhance simulation accuracy by increasing computational resources and employing higher-resolution grids.

4 Conclusions

This study employed numerical experiments with the RegCM-Chem-YIBs model to analyze the interannual variability of near-surface $PM_{2.5}$ concentrations in East Asia from 2008 to 2018. The analysis examines the drivers of annual $PM_{2.5}$ changes in detail, focusing on three key factors: anthropogenic pollutant emissions, meteorological conditions, and CO_2 concentration changes.

Compared to 2008, $PM_{2.5}$ concentrations in East Asia exhibited minimal change during the PreG stage, with most areas showing variations between -10 and 5 μg/m^3. In contrast, following the implementation of the Clean Air Action Plan, $PM_{2.5}$ concentrations decreased significantly during the PostG stage. This reduction was especially notable in the NCP and the SCB region, with declines of 36.76 μg/m^3 and 33.96 μg/m^3, respectively.

Anthropogenic pollutant emissions are the primary driver of the decline in $PM_{2.5}$ concentrations in East Asia, with their impact on $PM_{2.5}$ levels increasing linearly over time. During the PreG and PostG stages, the contributions of anthropogenic pollution emissions to $PM_{2.5}$ concentrations in the NCP, FWP, YRD, PRD, and SCB regions ranged from -10.39 to -3.82 μg/m^3 and -33.86 to -8.45 μg/m^3, respectively.

Changes in meteorological conditions have led to decreased $PM_{2.5}$ concentrations along China's eastern coastal and western inland regions, while increasing $PM_{2.5}$ levels in central areas. During the PreG stage, the influence of these meteorological changes on $PM_{2.5}$ concentrations was comparable to that of anthropogenic emissions, ranging from -6.31 to 2.32 μg/m^3.

CO_2 indirectly influences $PM_{2.5}$ concentrations by affecting precipitation and isoprene emissions from vegetation. In the sparsely vegetated NCP and FWP regions, CO_2 impacts near-surface $PM_{2.5}$ primarily through changes in precipitation. Conversely, in the vegetation-rich PRD region, CO_2 affects $PM_{2.5}$ concentrations mainly by altering isoprene emissions, with an impact comparable to that of anthropogenic emissions. From 2009 to 2013, the effects of anthropogenic pollution emissions and CO_2 changes on $PM_{2.5}$ ranged are -0.25 to 3.11 μg/m^3 and -6.19 to -1.47 μg/m^3, respectively.

In summary, $PM_{2.5}$ concentrations in East Asia have significantly declined since 2013, primarily driven by changes in anthropogenic pollutant emissions. During several years of the PreG period, variations in meteorological conditions affected $PM_{2.5}$ levels to a degree comparable to that of anthropogenic pollutant emissions. However, following the implementation of the Clean Air Action Plan in 2013, the influence of anthropogenic pollution emissions increased significantly, while the impact of meteorological factors diminished considerably. This simulation underscores the critical importance of stringent air pollution control measures in mitigating $PM_{2.5}$ concentrations. Moreover, we highlight that in regions with dense vegetation cover, changes in CO_2 concentrations play a noteworthy role in regulating $PM_{2.5}$ levels, with the average effect during the PostG phase even surpassing that of meteorological conditions. Given the sustained rise in CO_2 levels in recent years, it is imperative to integrate the modulatory effects of CO_2 into $PM_{2.5}$ simulating models and control strategies.

Data availability

ERA-Interim data are available at https://apps.ecmwf.int/datasets/data/interim-full-daily/. MEIC data are available at http://meicmodel.org/?page_id=560. CarbonTracker data are available at https://gml.noaa.gov/aftp/products/carbontracker/co2/CT2019/.

Author contributions

MX, TW, and DM: designed the research, and **DM:** carried it out. **MX, and LF:** provided technical support on the RegCM-Chem-YIBs mode. **SZ, and SC:** improved the paper. **HH:** improved and edited the paper.

Competing interests:

The contact author has declared that none of the authors has any competing interests.

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