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Long-term changes in black carbon aerosols and their health effects in rural India during the past two decades (2000–2019)

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ABSTRACT

Black carbon (BC) is a short-lived atmospheric aerosol having light absorbing properties with climate-changing potential. In addition, BC aerosols are also responsible for several adverse health effects including cardiovascular and respiratory problems. Here, we examine the long-term changes in BC, using MERRA-2 (Modern-Era Retrospective analysis for Research and Applications) and Emissions Database for Global Atmospheric Research (EDGAR) data for the period 2000–2019, and the associated health burden in rural India. This study finds a decreasing trend in BC in the rural IGP (Indo-Gangetic Plain) and NWI (North West India) during 2007–2019, at about -0.004 and –0.005 μg/m^3/yr, respectively. A significant reduction in BC (from 0.03 to 0.01 μg/m^3/yr after 2006) is observed in the rural Peninsular India (PI), where the reduced wind speed limits the transport of BC aerosols from other regions and thus, limits the BC concentration there. Our assessment finds that government policies such as BS (Bharat Stage) emission norms, electrification of rail routes, use of electric and compressed natural gas-based vehicles, the transformation of brick kilns to zig-zag technology, mechanised farming for on-site handling of crop residues and recent changes in atmospheric drivers (e.g. winds in IGP) contributed to this reduction in BC. However, the health burden associated with BC causes the highest all-cause mortality to be around 5,17,651 and 34,082 inhabitants in winter (December-February) and post-monsoon (October-November) seasons, respectively, in the rural IGP in the latest year 2019. In brief, the reduction of BC in rural India indicates that it complements the government policies. However, an improvement in the policy implementation might prove to be conducive to reduce the BC-driven mortality and regional climate warming.

1. Introduction

Short-lived climate forcers (SLCFs) are gases and particulates that persist in the atmosphere for relatively brief durations, spanning from less than a day to potentially extending over a few years (Bowerman et al., 2013). These substances contribute positively to the overall radiative forcing (RF) of global climate. SLCFs such as black carbon (BC) primarily impact the climate in the near-term because they remain in the atmosphere for a shorter period (Myhre et al., 2014). These pollutants exacerbate global warming and pose risks to human health. Although SLCFs influence Earth's radiation equilibrium and have worldwide consequences, their most noticeable impact is regional. Specifically, BC is the second most influential atmospheric component in regional climate warming, following carbon dioxide (CO_2) (Bond et al., 2013). It contributes significantly to direct radiative forcing, surpassing all greenhouse gases except CO_2 and possibly methane (CH_4) (Myhre et al., 2014). Unlike CO_2, which requires international efforts to mitigate its escalating global impact, BC is a pollutant that can be significantly reduced through interventions at local and regional levels (Bowerman et al., 2013; Shindell et al., 2012). This offers a promising avenue as BC originates from inefficient combustion processes from sources such as motor vehicles, commercial activities (e.g. brick making), residential heating, cooking and biomass burning, all of which can be more effectively regulated and technologically improved. Similarly, measures targeting BC reduction may mitigate other co-pollutants (including carbon monoxide, volatile organic compounds and organic carbon).

Several epidemiological studies have highlighted the detrimental health effects of $PM_{2.5}$ (fine particulate matter), mainly its association with increased mortality rates worldwide (Ostro, 2004; Krewski et al., 2009; Laden et al., 2006). Although research suggests that BC, a marker of traffic-related fine particulate pollution, may pose a higher health risk than other $PM_{2.5}$ components, there is an ongoing debate about which specific constituent of $PM_{2.5}$ is the most harmful. Some studies suggest that as BC, which indicates air pollution from fine particulates related to

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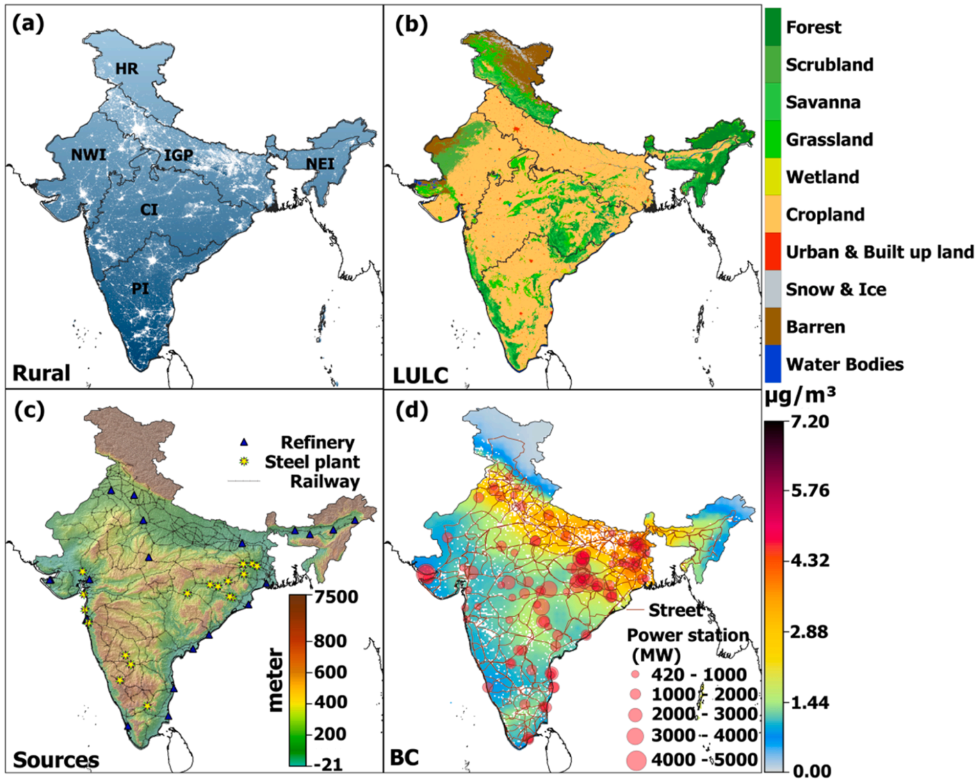

Fig. 1. (a). The rural India; (b) LULC classification; (c) Topography overlaid with major sources of BC including refineries, steel plants and railways and (d) Annual average BC concentration in rural India using the MERRA-2 data during 2000–2019 overlaid with street map and Thermal power stations. Here, LULC is Land use land cover and MW is Megawatts. The regions marked are IGP as Indo-Gangetic Plain, CI as Central India, NWI as North West India, PI as Peninsular India, HR as Hilly Region and NEI as North East India.

traffic may present a higher health risk to humans than $PM_{2.5}$ originating from other sources (Laden et al., 2006; Smith et al., 2009; Ostro et al., 2010).

Recent assessments, such as those conducted by the World Health Organisation, underscore the heightened health risks associated with BC when compared to PM_{10} and $PM_{2.5}$ measured in μg/m^3, suggesting that BC may serve as a more reliable indicator of harmful particulate matter from combustion sources than overall $PM_{2.5}$ mass measurements (Janssen et al., 2012). Research conducted in China suggests a stronger link between particles derived from combustion, notably BC, and cardiovascular mortality (CVM) (Wang et al., 2013). Studies in both the United States and China indicate that cardiovascular disease (CVD) morbidity and CVM per unit of aerosol mass are significantly greater for BC compared to unspecified PM, about 6–26 times higher (Li et al., 2016; Ostro, 2004; Laden et al., 2006). In contrast to regions like the Indo-Gangetic Plain (IGP), where the BC burden is significant, areas like the United States exhibit a considerably lower BC burden, estimated to be approximately 18–20 times lower than $PM_{2.5}$ (Janssen et al., 2011; Ostro et al., 2007).

Limited studies on long-term variability in BC (Ahmed et al., 2014; McDonald et al., 2015; Sharma et al., 2004, 2006; Chen et al., 2016), including those from the Asian region, suggest a declining trend in BC concentrations. A study conducted by Manoj et al. (2019) across 13 different stations in India using Aerosol Radiative Forcing over India network (ARFINET) observations from 2007 to 2016 indicates a similar decrease in BC levels, but the reasons behind such reductions need further analysis.

Recent research has increasingly utilised causal discovery as a prominent method to elucidate the interconnections among different climate drivers. It is a fundamental scientific approach essential for accurate forecasting, thorough explanation, and informed decision-making. For instance, causal discovery techniques have recently been applied to investigate the relationships among surface temperature and geophysical drivers (Kumar et al., 2023), and identify potential drivers of ozone (Kumar et al., 2022a). Though previous studies have focused on examining the impacts of PM and BC across various measurement stations in cities (Safai et al., 2012; Rajeevan et al., 2019), only a few have addressed rural pollution in India, notably studies by Ravishankara et al. (2020) and Pathak and Kuttippurath (2022). The latter reported high mortality due to cardiopulmonary disease and lung cancer associated with $PM_{2.5}$ in rural areas of districts in central IGP. Compared to $PM_{2.5}$, exposure to BC generally leads to a higher burden of CVM, ranging from 2 to 4 times higher and 10 times in some regions of Punjab and Haryana (Verma et al., 2022). This disparity arises because BC primarily originates from combustion processes, whereas other components of $PM_{2.5}$ in the atmosphere stem from a mixture of natural sources (such as crustal or marine sources) and human activities, including secondary chemical reactions (Ostro et al., 2007).

As India is an agrarian country with 67% of its population in rural regions, and there are only a few studies on rural pollution and the available are for specific regions or measurement stations, it is imperative to examine atmospheric pollution in rural India. Our previous studies have analysed the pollution in rural India in terms of $PM_{2.5}$ and NO_2 (Pathak and Kuttippurath, 2022; 2024). Therefore, here, we examine air pollution with respect to BC in rural India, and assess the associated health impacts and mortality for the past two decades (2000–2019). Since there was an anomalous reduction in BC during the COVID-19 lockdown period (Pathak et al., 2023), we have restricted our analysis up to 2019 to avoid its impact on the long-term analysis and trend estimates.

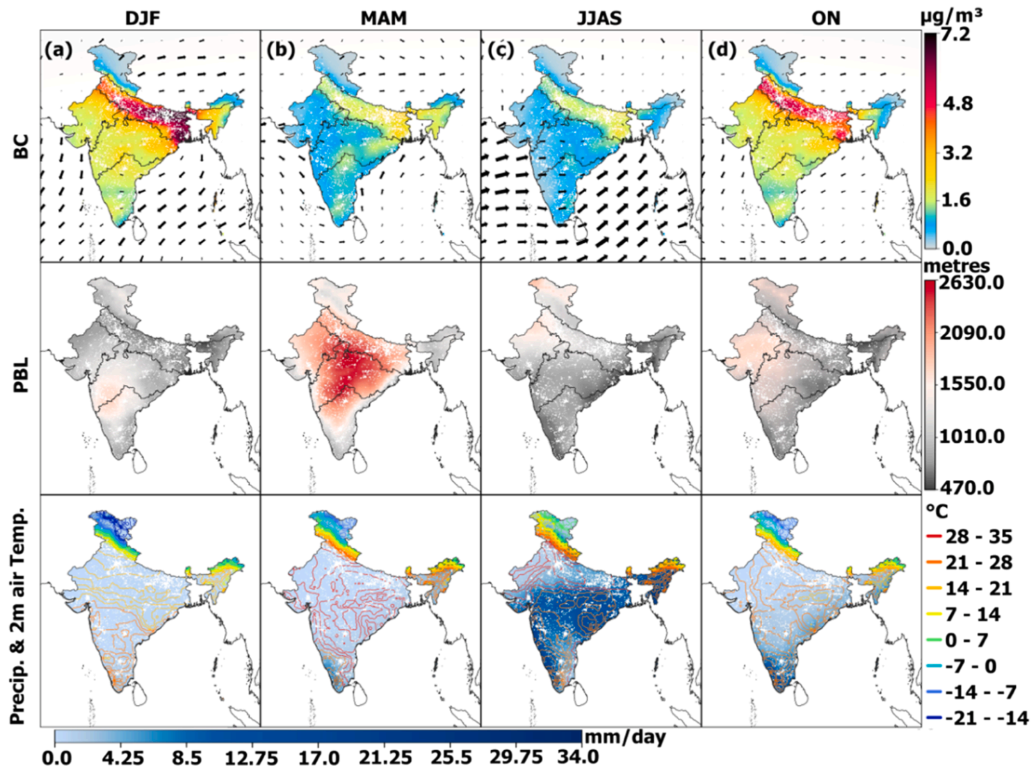

Fig. 2. Top: Seasonal BC concentration derived fromthe MERRA-2 data for the period 2000–2019. Second row: Seasonal changes in Planetary Boundary Layer (PBL) height from MERRA-2 during the same period. Third row from top: Seasonal changes in precipitation overlaid with 2 m air temperature from MERRA-2 during the same period. The seasons are DJF: Winter (December, January and February), MAM: Pre-monsoon (March, April and May), JJAS: Monsoon (June, July, August and September) and ON: Post-monsoon (October and November).

2. Study region, data and methods

2.1. Study area

The Indian subcontinent has a vast population that is majorly distributed in rural areas, and these regions have several sources of BC emissions such as biomass burning, industries and manufacturing units. Fig. 1 shows the study area and the critical sources marked to understand the relationships among BC and its sources in different regions. The rural regions considered in this study are IGP (Indo-Gangetic Plain), PI (Peninsular India), CI (Central India), HR (Hilly Region), NWI (North West India) and NEI (North East India), as shown in Fig. 1 a. We utilise nightlight collocated with Land Use Land Cover (LULC) and population density data for the year 2020 to identify areas characterised by urban settlement, transportation infrastructure and industrial activities. These data are then analysed together to distinguish rural regions from urban areas. For instance, high nightlight and population areas are primarily urban, but high vegetation areas with tiny population and low nightlight are often rural. However, the results reveal that numerous non-urban regions have a high population density ($>$ 1500 persons/km^2) comparable to urban regions. For instance, both rural and urban areas of IGP have a population density of $>$ 1500 persons/km^2 (Pathak and Kuttippurath, 2022). A detailed description of the methodology can be found in Pathak and Kuttippurath (2022) and Supplementary Section S1.1. The LULC of India as shown in Fig. 1 b is majorly covered with croplands, followed by forests, barren land and urban areas. These croplands are one of the major sources of BC emissions during the seasons of agriculture waste burning (AWB), which vary regionally in rural India (Jain et al., 2014). The topography of India as depicted in Fig. 1 c shows the low lying region of IGP, which is also the hotspot of BC (e.g. concentration $>$ 3.5 µg/m^3) as illustrated in Fig. 1 d. The other sources of BC include refineries, steel plants, railways, streets and power plants, as overlaid on Fig. 1 c and d.

To investigate the temporal evolution of BC emissions from different sources, we perform the regional source-wise inventory separately. The seasons are defined as December, January and February (DJF: winter); March, April and May (MAM: pre-monsoon); June, July, August and September (JJAS: monsoon), and October and November (ON: post-monsoon). The long-term trends are computed using simple linear regression with statistical significance determined based on the 95% confidence interval.

2.2. Data

We use the Modern-Era Retrospective analysis for Research and Applications Version 2 (MERRA-2) at a resolution of 0.5° $\times$ 0.625°, for monthly data of BC, 2 m air temperature (2mAT), wind speed (WS), specific humidity (SH), planetary boundary layer height (PBLH) and precipitation (P) (Gelaro et al., 2017). The Emissions Database for Global Atmospheric Research (EDGAR v8.1) is used for the power sector (PS), AWB, road transport (RTR), combustion from manufacturing (CFM), oil and refinery (OAR), and railways, pipeline and off-road transport (RPOR) inventory data (Crippa et al., 2018). The Terra Advanced Space-borne Thermal Emission and Reflection Radiometer (ASTER) Global Digital Elevation Model V002 is used for the assessment of surface elevation (Hulley et al., 2012). These datasets are presented in detail in Supplementary Section S1.2.

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model used in this study is explained in Supplementary Section S1.3 (Stein et al., 2015). Here, we have computed 72-hour backward trajectories for the representative year 2016, at 500 m above the ground to reduce the effect of friction and to represent the winds in the lower troposphere, at five distinct receptor sites in rural India (L_1: 29.84 °N, 77.28 °E; L_2: 26.4 °N, 90.75 °E; L_3: 26.16 °N, 79.85 °E; L_4: 20.92 °N,

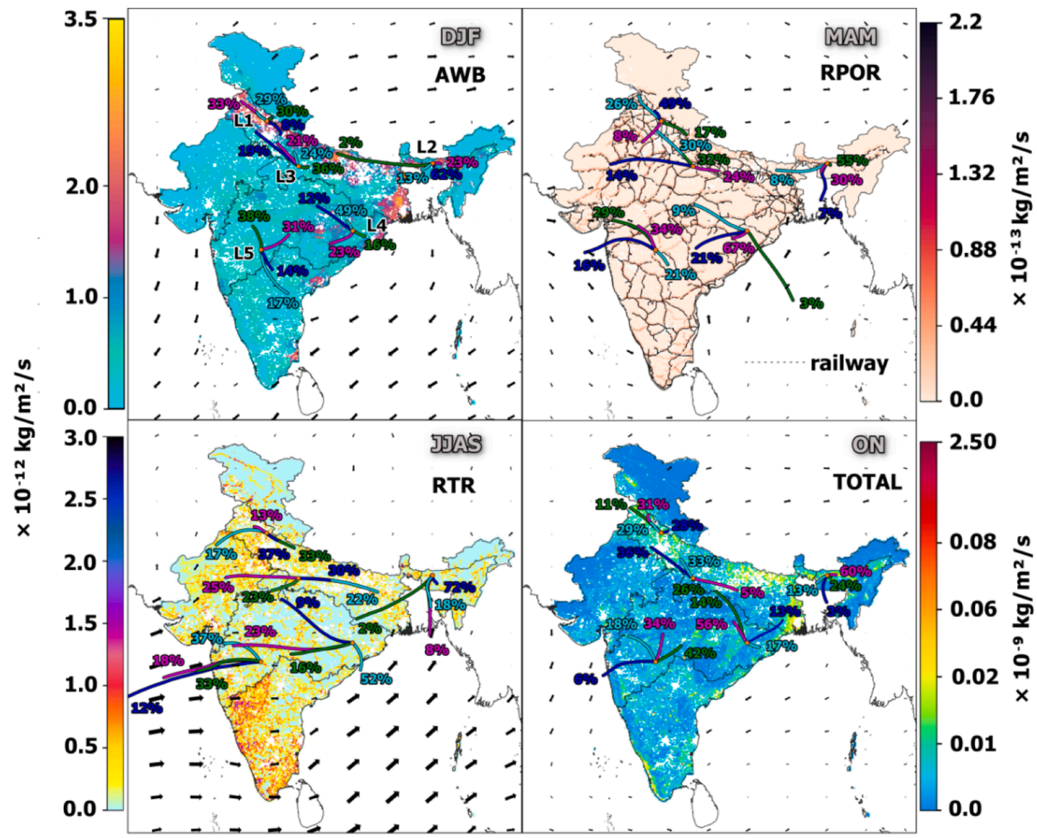

Fig. 3. The AWB, RPOR, RTR and total BC emissions during 2000–2019 overlaid with seasonal trajectories of air mass transport in rural regions for the year 2016. The sectors are AWB is Agriculture Waste Burning, RPOR (Railways, Pipeline and Off-Road transport), RTR (Road Transport no Resuspension) and TOTAL (all sectors included). The seasons are DJF: winter (December, January and February), MAM: pre-monsoon (March, April and May), JJAS: monsoon (June, July, August and September) and ON: post-monsoon (October and November).

84.25 °E and L_5: 19.38 °N, 76.81 °E). We have taken receptor sites at locations in rural regions of NEI, CI and IGP, as rural India is the focus of this study. For instance, the choice of locations was made in rural IGP due to the fact that the central IGP has high BC values, which might be due to emissions from local sources or long-range air mass transport, as also mentioned by Rana et al. (2019).

We utilise the Peter Clark momentary conditional independence (PCMCI$^+$) algorithm to explore the causal relationship between geophysical factors and BC concentration across the rural regions during the periods 2000–2006 and 2007–2019 (Runge et al., 2019). These periods are chosen in response to the reversal of BC trends in north India, particularly in IGP. A detailed description of the causal discovery is provided in Supplementary Section S1.4. We consider data including WS, SH, P, 2mAT and PBLH from MERRA-2 to elucidate the influence of various geophysical drivers on BC distribution.

2.3. Methods

The mortality associated relative risk (RR) as computed by Crippa et al. (2016) for BC using a prediction model is as follows:

$$RR = exp(\beta[C_n - C_0]) \tag{1}$$

where C_n and C_0 are the average and baseline BC concentration (μg/m^3) (assumed to be 5 μg/m^3), respectively, and β is the BC exposure associated risk coefficient for mortality. A detailed description of β for all-cause (ACM), cardiovascular (CVM) and respiratory (RM) mortality is provided in Supplementary Section S1.5.

The population attributable fraction (PAF) as computed using the methodology by Faridi et al. (2022):

$$PAF = (RR - 1)/RR \tag{2}$$

The health burden attributed to BC is estimated using the following equation by Verma et al. (2022):

$$NM = y_0 \times PAF \times PD \times a \tag{3}$$

where y_0 is the mortality rate including all age groups and sex (deaths per 100,000 people in a year) due to ACM, CVM and RM, PD is the gridded population density in the unit of 100,000 persons/km^2, and *a* is the area of the grid in km^2.

3. Results and discussion

3.1. Spatial distribution of BC aerosols in rural India

Fig. 2 illustrates the seasonal distribution of BC in rural India along with meteorological parameters, including 2 m wind speed, PBLH, precipitation and air temperature for the period 2000–2019. The highest BC concentration (> 7.2 μg/m^3) is detected in winter over the rural IGP, which is associated with enhanced wintertime anthropogenic activities (such as biomass burning for industrial and domestic purposes) with lower PBLH (< 1000 m), light precipitation rate (< 2 mm/day), low temperature (around 14–21 °C) and lower wind speed (< 4.5 m/s), compounded by the obstruction of atmospheric circulation by the Himalayan mountains (Guttikunda et al., 2014). An increasing gradient in BC is observed from the upper to the lower IGP. This is due to the north-westerly winds, which dominate IGP during winter months, and transport BC to the eastern India. These changes in BC aerosol-meteorology interactions are seasonal and are expected to influence the dispersion of BC over IGP and the nearby downwind regions.

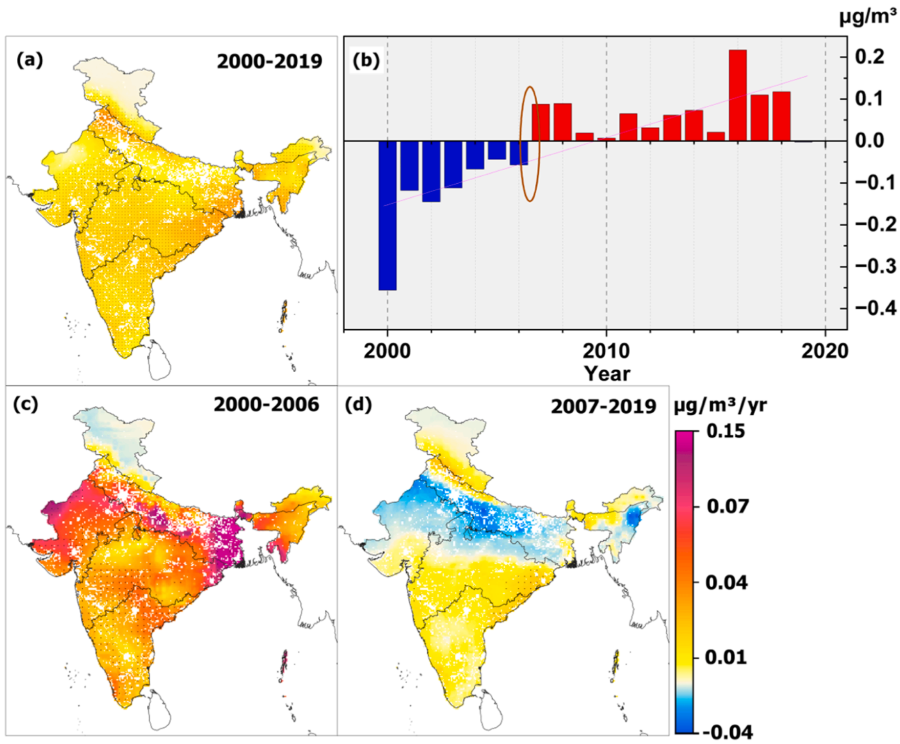

Fig. 4. a. Trends of BC concentrationin in rural India during the period 2000–2019; b: Anomaly of BC in rural India during the same period; c: Trend in BC concentration for the period 2000–2006; d: Trend in BC concentration during 2007–2019 in rural India. The stippled regions represent statistical significance at the 95% confidence interval.

However, the rural HR is the cleanest of all regions with BC concentration < 1 μg/m^3 throughout the year, which might be attributed to the absence of emission sources, as shown in Figs. 1 and 2.

The pre-monsoon season has a relatively lower BC concentration throughout rural India, particularly over the rural IGP, with significant contribution from AWB, typically undertaken after the wheat harvest before the monsoon (Lan et al., 2022). This reduction might be attributed to the enhanced PBLH (> 2000 m) and dry deposition in the absence of precipitation in all rural regions. The south PI receives early rainfall showers from the southwest monsoon in this season, causing wet deposition of BC and reduction even with lower PBLH (< 1000 m). The cleanest among all is the monsoon season with high precipitation throughout rural India, causing escalated wet deposition of BC in the rural regions of western PI (> 20 mm/day), IGP (around 5 mm/day), CI and NEI. The post-monsoon shows high concentrations of BC due to AWB following the rice harvest (Lan et al., 2022), where the retreating monsoon in IGP along with reduced wind speed, PBLH and air temperature, plays a key role in enhancing BC concentrations. The rural IGP shows the highest BC concentrations throughout the year as compared to other parts of India. This might be attributed to a large number of sources including brick kilns, many of which utilise outdated and inefficient combustion methods, relying on a combination of biomass and coal for fuel (Guttikunda et al., 2014). Furthermore, states such as Bihar, Chhattisgarh, West Bengal, Orissa and Jharkhand in IGP are home to the largest coal mines in India, which are often accompanied by clusters of power plants in close proximity (Guttikunda and Jawahar, 2014). Additionally, several large-scale power plants are located in Haryana, Punjab, Uttar Pradesh and Delhi, and are making IGP the most polluted region in the country. These sources contribute to unceasing background concentrations of BC throughout the year together with the seasonal AWB in the pre (wheat harvest: April - May) and post-monsoon (rice harvest: October - November) seasons.

3.2. Emissions and transport of BC

The sector-wise distribution of regional BC in rural India is depicted in Fig. S1 using EDGAR v8.1 data for the period 2000–2019. The highest emissions are observed in IGP from AWB (2×10^{-12} kg/m^2/s), which shows a continuous increase in emissions from 2000 to 2019, followed by CFM (1×10^{-12} kg/m^2/s), RTR (0.3×10^{-12} kg/m^2/s) and PS. To identify the transport of air mass towards the selected sites in rural India, we run three-day backward trajectories at five receptor locations L_1, L_2, L_3, L_4 and L_5 for the year 2016 (Fig. 3). The AWB, RPOR, RTR and total BC emissions during 2000–2019 overlaid with trajectories in rural regions for the representative year 2016 are also shown in Fig. 3. The air mass transport to all the receptor sites is influenced by seasonal variation in meteorology, including humidity, temperature, PBLH, and wind speed and direction. Among the sources, biomass burning (including AWB) is one of the prime emitters of BC in rural India. The air mass along with emissions is transported to several cities, towns and nearby rural areas that are downwind of the source. During winters, about 62% and 64% of trajectories arriving at L_1 and L_3, respectively, are from the upper IGP dominated by OAR, transportation and manufacturing emissions throughout the year, and the seasonal AWB emissions. Shorter clusters carry emissions from the upwind direction, making enhanced BC around that region, particularly during winters, which is also influenced by lower PBL and reduced wind speed to accumulate pollution there.

At L_2, a longer air-flow trajectory with a 2% contribution carries total emissions of about 0.02×10^{-9} kg/m^2/s from the central IGP, whereas the cluster with 13% contribution is originated from Bangladesh and the rest 85% from local sources within the rural NEI. The rural CI with clusters of industries, power plants, steel plants and manufacturing units show high emissions of BC (Fig. S1) from the CFM sector (1.1×10^{-12} kg/m^2/s), followed by AWB, RTR and PS. The emissions from sectors get transported to locations L_4 and L_5 along with the air, as shown in Fig. 3. All rural regions show high emissions from the manufacturing sector,

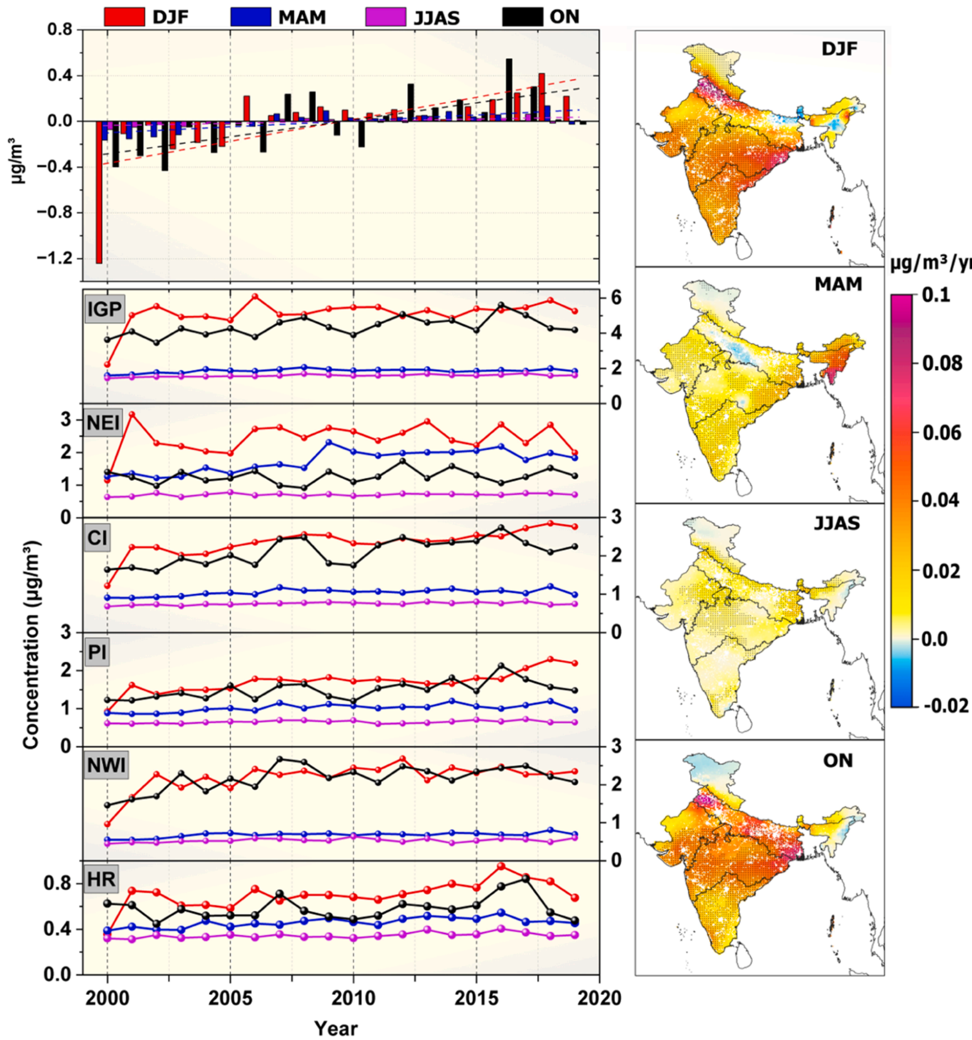

Fig. 5. Left side panels: The seasonal anomalies in BC concentration in rural India (Top panel) together with region-wise seasonal BC concentration (bottom panels) during 2000–2019. Right side panels: The seasonal trend in BC concentration in rural India during the same period. The seasons are DJF: winter (December, January and February), MAM: pre-monsoon (March, April and May), JJAS: monsoon (June, July, August and September) and ON: post-monsoon (October and November). The stippled regions represent statistical significance at the 95% confidence interval.

followed by AWB, except the rural IGP, where AWB emissions are the primary source of BC. Note that emissions from the OAR sector were reduced in rural PI, CI, NWI and HR after 2006.

During pre-monsoon, a cluster originating from the Thar desert in India (Fig. 3) contribute 14% at L_3, transporting natural soil dust and small amounts of BC and OC to the rural IGP and CI. However, small concentrations of BC are observed due to higher wind speeds and the presence of a well-defined seasonal PBLH extending beyond 2600 m height there (see Fig. 2). Around 30% air transport occurs from upper to central IGP, causing enhanced BC concentration in that region. At L_2, around 85% of the contribution is from local air transport including AWB emissions, prevalent in the rural NEI during this season.

In the monsoon season, BC particles undergo long-range transport, where the length and speed of airflow trajectories influence their deposition capacity. However, increased precipitation leads to wet deposition, diminishing the impact of these longer airflow trajectories. The most extended inland air flow identified at L_3 shows that 25% contribution is from the rural NWI and 30% from local sources east of this receptor site (Fig. 3). This season shows the lowest concentration of BC (Fig. 2), followed by pre-monsoon.

In addition, in the rural IGP, post-monsoon season is influenced by AWB, which is associated with harvesting Kharif crops, leading to elevated levels of BC. During this period, at L_1, approximately 89% of the air mass originates from the states of Haryana and Punjab, with the remaining 11% from Punjab in the western Pakistan and Lahore that carry BC air to rural IGP (e.g. Rana et al., 2019). Additionally, air from surrounding regions upwind of IGP also contributes to the high BC concentrations within the region. Given the consistently high aerosol levels observed in IGP throughout most seasons, the central region of IGP, which exhibits the highest concentration levels, can be described as an "aerosol super-hotspot" (Kuttippurath and Raj, 2021). The highest AWB emissions are observed ($> 3.0 \times 10^{-12}$ kg/m^2/s) in the rural IGP, NEI and CI, whereas RTR emissions are highest in the rural PI (around 1.5×10^{-12} kg/m^2/s) due to its dense road network, followed by NWI (around 1×10^{-12} kg/m^2/s) and IGP (1×10^{-12} kg/m^2/s). The emissions from RPOR sector are distributed along the railway network throughout the country, particularly in IGP (0.6×10^{-13} kg/m^2/s). The total emissions are high in the rural IGP (around 0.02×10^{-9} kg/m^2/s) compared to other regions due to the significant sources such as AWB, RTR, PS and CFM.

3.3. *Significant changes in BC in rural India*

The trend in BC concentration in rural India during 2000–2019 is depicted in Fig. 4 a. The rural areas of upper and lower IGP, NEI and eastern CI exhibit a significant positive trend of 0.02 μg/m^3/yr. The annual anomaly over rural India, as shown in Fig. 4 b, indicates its complete reversal from 2006 onwards, i.e. -0.35 μg/m^3 in 2000 to +0.1 μg/m^3 in 2019. Fig. 4 c and d show the trends in BC concentration in rural India during 2000–2006 and 2007–2019, respectively. During 2000–2006, positive trends in BC (> 0.06 μg/m^3/yr) were found in rural India, which might be due to continuous increase in CFM and AWB emissions (Fig. S1) in all regions, particularly in the rural IGP, CFM (1 to 2×10^{-12} kg/m^2/s) is the highest emitting sector followed by AWB (0.5 to 1×10^{-12} kg/m^2/s). Apart from these, the negative trends in the wind speed (-0.03 m/s/yr) in all regions (except the rural PI with significant positive trends in wind speed [Fig. S2]) might be responsible for the enhanced aerosol transport from the heavily polluted IGP.

During 2007–2019, a reversal of the BC trend is found in IGP, NWI, parts of NEI and CI, for which the significant negative trends (-0.02 μg/m^3/yr) (Fig. 4 d) correspond to an increase in wind speed (from 0.4 to 1.2 m/s, Fig. S2). The reduction in positive BC trends over the rural PI might also be attributed to the reduction in wind speed (from 0.02 m/s/yr in 2000–2006 to 0.005 m/s/yr in 2006–2019) that decreased the transport of aerosols from the northern parts of rural India (Fig. S2).

A study conducted by Kumar et al. (2022b) discovered a notable acceleration in global wind speeds post-2010, which was approximately three times the declining rate observed prior to 2010. This finding contradicted previous theories of global wind speed reduction, as proposed by Vautard et al. (2010). It is worth noting that while terrestrial roughness remained relatively constant before and after 2010, there was a discernible uptick in global wind speeds. Consequently, the fluctuation in wind speed appears to be primarily influenced by the driving forces linked to the decadal variability of large-scale ocean-atmospheric circulations, challenging the previous notion that it was solely determined by terrestrial roughness (Zeng et al., 2019). However, the influence of other climate drivers might also have contributed to the temporal change in BC concentration in rural India. The influence of these drivers is discussed in Section 3.4.1 in detail.

The seasonal anomalies in BC concentration in rural India during 2000–2019 are shown in Fig. 5. The highest negative anomaly of 1.2 μg/m^3 was observed in 2000 during winter, whereas the highest positive anomaly of 0.6 μg/m^3 was detected in 2016 during the post-monsoon season. The seasonal trends during 2000–2019 show upper IGP (0.1 μg/m^3/yr) and eastern CI as the hotbeds of BC during winter and post-monsoon, whereas the pre-monsoon has a negative trend of BC in the central IGP. The rural NEI shows the highest positive trends during post-monsoon owing to high AWB there. The monsoon season shows an overall reduction in trends (about 0.01 μg/m^3/yr, significant at the 95% confidence interval), throughout rural India. The seasonal variation in BC in the rural IGP during 2000–2019 shows the highest concentration during winter (7.2 μg/m^3), followed by post-monsoon (5 μg/m^3). The rural NEI has highest concentration during winter (3.2 μg/m^3), followed by pre-monsoon, which experiences high AWB due to shifting cultivation prevalent in this season. A continuous rise in BC was observed from 2000 to 2019 in the rural NWI during post monsoon (1–2 μg/m^3) and winter (1–3 μg/m^3), due to an increase in local emissions and enhanced downwind transport from highly polluted adjoining regions. However, the most immaculate is the rural HR, with BC concentration of between 0.4 and 0.9 μg/m^3 in winter and post-monsoon of the study period.

3.4. Why is there a reduction in BC?

3.4.1. Causal discovery of natural drivers influencing BC aerosols

Having identified the changes in BC concentrations from 2006 onwards, we now apply the causal discovery method for the identification of change in the influence of causal drivers on surface BC variability in rural India during the periods 2000–2006 and 2007–2019 (Fig. S3). Herein, a significant negative trend in BC over the rural IGP (-0.004 ± 0.011 μg/m^3/yr) and NWI (-0.005 ± 0.003 μg/m^3/yr) is observed during 2007–2019, as shown in Fig. S3. Therefore, we generate the causal network graph of the rural IGP and NWI using the Pearl causality-based method, i.e., PCMCI to examine whether surface BC variability is driven by changes in climate variables such as WS, SH, P, 2mAT and PBLH. In causal analysis, edges without arrows denote the association between drivers. During 2000–2006, a positive trend of BC in the rural IGP (0.073 ± 0.031 μg/m^3/yr) and NWI (0.057 ± 0.020 μg/m^3/yr) is observed where the causal discovery reveals the negative influence of WS on BC (maximum lag of 1 month) in both regions with a conditional cross correlation of about −0.2 and -0.4, respectively. However, the reversal in BC trend after 2006 in the rural IGP (-0.004 ± 0.011 μg/m^3/yr) and NWI (-0.005 ± 0.003 μg/m^3/yr) implies a change in the influence of drivers as revealed from the causal links. The reduction in BC can be attributed to the conjoint influence of WS and SH (influenced by P), as shown in the causal analysis and the government policies as described in Section 3.4.2. It shows a negative cross-correlation with BC in the rural IGP (-0.2 and -0.2) and the rural NWI (-0.4 and -0.4). Precipitation contributes to reduce BC in the atmosphere through wet deposition. However, there is no direct causal relationship observed between BC and P here, though an indirect link is identified through SH. This suggests that meteorology can modify the dispersion and reduction of atmospheric aerosols. However, there might be a possibility of an immediate causal relationship, contingent upon the data to establish statistical correlations between these drivers. It is essential to recognise that such connections may not always imply a definitive unequivocally causal.

3.4.2. Government policies and regulations

To mitigate air pollution several regulations and actions were implemented through various provisions like the Air (Prevention and Control of Pollution) Act, 1981, and the Environment (Protection) Act, 1985, which outline the mechanisms and authorities responsible for addressing the issues. After the inception of Mass Emission Regulation in 1991, the Indian government has enacted laws aimed at controlling and monitoring industrial and vehicular emissions. The adoption of progressively stringent standards such as Bharat Stage II (BS II) in 2000, BS III in 2005, BS IV in 2017, and BS VI in 2020 followed the implementation of the India 2000 emission standard as depicted in Fig. S1. Simultaneously, efforts have been made to improve fuel quality. Recent initiatives to adhere to BS VI standards, notably reducing sulfur, lead and benzene content, have also enhanced the octane/cetane numbers for petrol/diesel. The sale of BS IV vehicles was officially prohibited as of April 2020, consistent with the nationwide enforcement of BS VI standards, to reinforce these measures (Manoj et al., 2019).

Despite an overall increase in fuel consumption, coal's contribution to total energy remained stable from 2000 to 2012 (Manoj et al., 2019), indicating that the emission control measures across various sectors have effectively managed particulate emissions, including BC. A study by Wang et al. (2014) suggests that advancements in combustion technology and fuel quality have increased efficiency, resulting in lower BC emissions from industries, residential areas and transportation in India. By 31 March 2016, 98.1% of inhabited villages in India had access to electricity, with approximately 24% of their electricity consumption allocated for domestic purposes (Ministry of Statistics and Programme Implementation, 2017b). The Ministry of Petroleum and Natural Gas (MOPNG) launched the Pradhan Mantri Ujjwala Yojana (PMUY) in 2016, a flagship initiative aimed at providing clean cooking fuel such as LPG to rural and underprivileged households that previously relied on traditional fuels like firewood, coal and cow-dung cakes. In addition, as of April 2017, electrification had been completed for 45% of India's railway network (route kilometres) (IR, 2017).

Burning of paddy stubble is predominantly practised in IGP, particularly in Punjab, Haryana, and Uttar Pradesh, to prepare fields for sowing Rabi crops. To support pollution control efforts of the governments of these states and Delhi, and to subsidise the machinery needed for managing crop residue in situ, a scheme called 'Promotion of Agricultural Mechanization for In-Situ Management of Crop Residue' has been implemented since 2018 (Ministry of Agriculture & Farmers Welfare, 2018). Promoting of electric vehicles and compressed natural gas

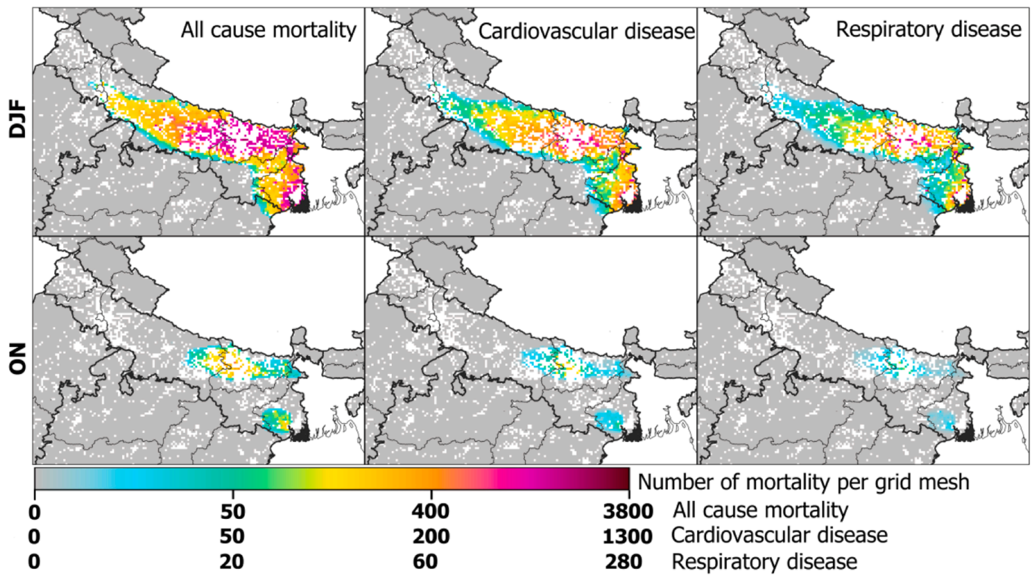

Fig. 6. Spatial distribution of seasonal all-cause, cardiopulmonary and respiratory mortality burden (number of mortalities per grid of 0.5° × 0.625°) associated with BC in IGP (Indo-Gangetic Plain) during winter and post monsoon seasons of 2019. The seasons are DJF: winter (December, January and February) and ON: post-monsoon (October and November).

(CNG) fuelled vehicles is also encouraged to reduce air pollution. Though these initiatives have contributed to control the concentration of various pollutants, assessing their impacts remains challenging.

3.5. Cause specific mortality assessment in rural India

The rural IGP is among the densely populated regions in India. The population residing in this region is exposed to high winter and post-monsoon mean BC concentration (> baseline concentration of 5 μg/m^3). Other regions (Fig. S4) as well as seasons show less BC concentration with respect to the baseline and thus, subside the health risk to the population residing in rural India. However, in winter alone, more than 248 million people (about 75%) of the rural IGP are exposed to dangerous levels of BC. We assess the health impact across the rural IGP for ACM, CVM and RM attributable to wintertime and post monsoon BC exposure. The spatial distribution of the number of mortalities per grid (resolution, 0.5° × 0.625°) during 2019 is shown in Fig. 6. Areas of large BC-attributable mortality (> 50 inhabitant mortality per grid) are found spatially distributed over the BC hotspots, with mortality > 400, > 200 and > 60 inhabitants per grid in parts of Uttar Pradesh, Bihar, West Bengal and Jharkhand attributed to ACM, CVM and RM, respectively, in winter. The overall BC-attributable burden is estimated to be around 5,17,651 and 34,082 inhabitants during the winter and post monsoon seasons, respectively, in the rural IGP due to ACM in 2019. We observe a reduction in BC over the rural IGP and NWI after 2006 (Fig. S3). However, the associated health burden is still crucial and demands the implementation of improved mitigation plans to further reduce BC concentrations in India.

Our BC-associated mortality analysis has some limitations, as we have considered all ages and sexes while assessing the mortality due to the three health endpoints i.e. ACM, CVM and RM. The effects of short and long-term smoking, drinking, effect of exercise and other confounding factors on individuals are not considered in this study, which might influence the BC-induced mortality rates. Therefore, more data on BC measurements, health impacts, climate parameters and mobility from rural regions of India would further improve the assessment of health burden. However, this study provides the first estimates of BC-associated mortality in rural India.

4. Conclusions

We present the first comprehensive analysis of BC pollution in rural India and its associated health impacts. Rural areas within IGP, NWI and CI often experience high BC exceeding its baseline concentrations (5 μg/m^3). However, the highest levels are observed during winter (> 6.5 μg/m^3) and post-monsoon (> 5.5 μg/m^3) seasons in the rural IGP, whereas the rural HR (< 1 μg/m^3) maintains BC concentrations below the baseline throughout the year. We have observed a significant negative trend in BC in the rural IGP and NWI with regional average reductions of about -0.004 and -0.005 μg/m^3/yr, respectively. Causal analysis suggests that natural drivers, including wind speed and specific humidity, might have influenced the changes in BC. Apart from this, the policies associated with BC and other co-pollutants might have played a significant role in this reduction, but that cannot be quantified due to the dearth of data in rural India, making it a discussion for future studies. The health impacts of BC still make it a severe threat to the lives of a large rural population in India. This research provides the first assessment of the disease burden linked to BC exposure in rural India in validation of the WHO recommendation to include BC as an indicator, alongside $PM_{2.5}$, for gauging human exposure to air pollution. The highest mortality rates associated with BC are observed in the rural IGP, with over 400, 200, and 60 deaths per grid attributed to ACM, CVM and RM, respectively, particularly during winter. By highlighting BC-related mortality in rural India, this study prompts policymakers to prioritise emission mitigation efforts while considering the sources of BC, and thus, provide health benefits to around 248 million individuals residing in the rural IGP exposed to high BC concentrations.

CRediT authorship contribution statement

Mansi Pathak: Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Jayanarayanan Kuttippurath:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Conceptualization. **Rahul Kumar:** Writing – review & editing, Visualization, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.hazadv.2024.100519.

Data availability

Data will be made available on request.

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