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Evaluation of Background Black Carbon Concentration in India

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ABSTRACT

Air pollution levels are rapidly increasing over the Indian region in recent times impacting the health and welfare of the general population of which black carbon (BC) is an important component. Significant efforts are being made to reduce pollution levels focusing on highly polluted cities. It is qualitatively known that a significant background BC pollution load exists over the Indian region. However, no systematic studies have focused on scientifically quantifying them. In this study, an effort is made to understand and segregate BC, an important primary aerosol/air pollutant, into the background (the minimum concentration levels that exist due to regional and long-range transport) and foreground concentrations (local emissions) using a simple methodology. The method is evaluated for consistency using ground-based observations (BC from Aethalometer) and extended to the whole Indian region using chemical reanalysis datasets (MERRA-2, Modern-Era Retrospective Analysis for Research and Applications, version 2). Our analysis reveals that the background BC over India reaches its highest during the winter season, 2.32 $\mu g\ m^{-3}$ (background BC ~86%), and its lowest during the monsoon season, 0.60 $\mu g\ m^{-3}$ (background BC ~79%). Furthermore, regardless of the season, the level of background BC in India exceeds 75% consistently. An independent evaluation of foreground BC after the removal of the background shows major emissions sources similar to the HTAP_v3 (Hemispheric Transport of Air Pollution) BC emission database, indicating the strength of the analysis. The high degree of qualitative agreement between foreground BC concentration and BC emission sources provides confidence in the analysis. These findings will provide policymakers with a valuable perspective, emphasizing the significant influence of background BC aerosol pollution in India.

Keywords: Air pollution, Air quality, Atmospheric aerosols, Carbonaceous aerosols, Regional air quality

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1 INTRODUCTION

Black Carbon (BC) is a primary aerosol produced from the incomplete combustion of fossil fuel and biomass burning. Due to its strong light-absorbing properties, BC absorbs radiation thereby warming the atmosphere. This affects the atmospheric thermal structure, and potentially cloud properties and circulation (IPCC, 2013). Developing nations like India, which have a large population and are rapidly industrializing, have significant sources of carbonaceous aerosols. However, the diverse nature of emissions and long-range transport makes changing characteristics of BC concentration not always attributable to local emissions. It is well known that both local and background pollution contributes to the overall pollution levels in a region. Several studies have been conducted in the Indian region to understand the nature of BC or air pollution at various spatiotemporal scales, including long-range transport (Guha *et al.*, 2015; Kumar *et al.*, 2015; Mishra *et al.*, 2013; Pandey *et al.*, 2016; Prasad *et al.*, 2018; Vinoj and Pandey, 2022). In describing the seasonality of air pollution, most of the studies show that winter is the most polluted period, and attribute it to local emissions. However, surprisingly some studies indicate long-range transport even during winter. For example, backward trajectory analysis shows, air masses reaching Bhubaneswar (a location in the extreme east and the outflow region) via IGP from West Asia, carrying significant amount of aerosols (Norman *et al.*, 2001; Mahapatra *et al.*, 2014). Both the properties of BC aerosols and their relatively longer residence time in the atmosphere allows them to travel across continents (Bond *et al.*, 2013), contributing to background BC in receptor locations far away from their original sources. However, it is unclear what portion of overall BC aerosol concentrations are caused by regional and long-range transport versus local emissions over the entire Indian subcontinent.

The global databases such as HTAP (Hemispheric Transport of Air Pollution) have created monthly emission grid maps collaborating with regional emission inventories (North America, Europe, Asia including Japan and South Korea) and with EDGAR emission inventory which provides the information about local BC emissions (Crippa *et al.*, 2023). Thus, most scientific attempts were directed to either characterise the mean BC mass concentration, emissions, their spatiotemporal variability, long-range transport or directly their climate effects. In all these, one characteristic that is highly uncertain/unknown is the background or baseline BC concentrations.

The background concentration refers to the lowest concentration level of pollution possible at any given location/time and is present in ambient air to which the general population is continuously exposed with implications to their long-term health (Tchepel *et al.*, 2010; Gómez-Losada *et al.*, 2018). On the other hand, these baseline concentrations are also capable of influencing regional-scale weather and climate due to their large spatial scales. However, spatially the background concentrations are not a constant value as local geographical attributes, continuous local emissions and regional air quality can indirectly impact it (Han *et al.*, 2015) leading to large spatio-temporal variability (McNabola *et al.*, 2011). Despite their importance, systematic attempts in estimating them are limited over India, as it is also not possible to directly measure them.

Most importantly gaining knowledge about background pollution, especially black carbon (BC), will provide insight into its regional scale spatio-temporal distributions. Localized emission reductions for cleaner air will not be able to reduce pollution in areas with elevated background pollution levels. On the contrary, emission reduction strategies will work more efficiently where background levels are lower. Such information is required for both the government and policymakers for a comprehensive understanding of BC and develop effective and collaborative strategies for controlling BC air pollution.

Watson and Chow (2001) were the first to determine the regional and local contribution of BC using the Moving Mean Subtraction (MMS) method in Mexico City. They showed that 65% of the BC came from distant sources (5–50 km), 20% came from neighbourhood sources (1–5 km), and 12% came from nearby (< 1000 m) sources. Both *et al.* (2011) used the same method to infer local and urban-scale emissions to confirm that emissions are higher in low-income areas than in middle-income areas. Kumar *et al.* (2018) found that, on average, local sources contributed 8 to 12% among the three rural sites. Local contributions were highest (16–25%) during the morning peak hours. Prabhu *et al.* (2022) and Yeganeh *et al.* (2021) successfully used this method in Bangalore and Tehran, suggesting that ~78% and ~77% of BC come from regional and long-distance sources respectively. It is important to note that the studies mentioned here were specifically carried out at individual sites using high-frequency surface BC or particulate matter ($PM_{2.5}$) based measurements to understand city-scale aerosol/air pollution mass concentrations.

In the present study, an attempt is made to estimate baseline/background BC mass concentration with the following objectives,

1. What is the most appropriate averaging window to estimate local and regional/background/ baseline BC mass concentrations?
2. Possibility of applying the MMS technique to chemical reanalysis datasets such as Modern Era MERRA-2 chemical reanalysis to extend the analysis to a larger spatio-temporal scale.
3. What additional insights could be gained by the application of MMS on high-frequency MERRA-2 datasets?

The paper is organized in the following ways: data in Section 2, methodology in Section 3, results in Section 4, and discussion and conclusion in Sections 5 and 6, respectively.

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2 DATA

2.1 BC Concentration from Aethalometer

The BC mass concentration was measured using the Aethalometer model AE33 (Magee Scientific) at the Indian Institute of Technology Bhubaneswar campus in the Southern part of Bhubaneswar city during the period January 2018 to December 2021. The sampling ducts were located at a height of ~10 m above the ground. The AE33 uses a new dual spot technology to measure BC mass concentration in the sample at seven different wavelengths: 370, 470, 520, 590, 660, 880, and 950 nm. The aerosol particles are sampled continuously on the filter, and the optical attenuation is measured in a one-second or one-minute resolution time. Attenuation is measured at two points with two sample flows (ATN_1 and ATN_2) with different flow rates and at the reference point without any flow. The BC mass concentration was calculated from the variation of the optical attenuation at 880 nm in the selected time interval using a mass absorption cross-section of 7.77 m^2 g^{-1} (Drinovec *et al.*, 2015).

2.2 BC Concentration from MERRA-2

MERRA-2 is the most recent satellite-era global atmospheric chemical reanalysis produced by NASA's Global Modeling and Assimilation Office (GMAO) using the Goddard Earth Observing System Model (GEOS) version 5.12.4. The dataset spans from 1980 to the present, with a 3-week latency after the end of a month. MERRA-2 products are available with a temporal resolution of 1 hour. Hourly BC surface mass concentration data spanning the years 2018 to 2021 was utilised in this analysis.

2.3 BC Emission from HTAP v3

The HTAP v3 mosaic is a collection of monthly and sector-specific global air pollutant emission grid maps created by combining detailed regional data from recently publicly published national or regional emission inventories. HTAP_v3 incorporated data from six primary regional inventories spanning North America, Europe, and parts of Asia, such as Japan, China, India, and South Korea (Crippa *et al.*, 2023). The HTAP v3 dataset has a spatial resolution of 0.1 × 0.1° and covers a time period from 2000 to 2018. The dataset includes global emission grid maps for monthly air pollutants such as CO, NO_x, SO_2, PM_{10}, $PM_{2.5}$, NMVOCs, NH_3, BC, and OC (Kim *et al.*, 2023). The HTAP_v3 input emission grid maps include monthly temporal distributions to accurately represent the seasonal variations of sector-specific emissions such as home, power production, and agricultural activities, in contrast to the earlier HTAP emission mosaics (Crippa *et al.*, 2023).

3 METHODOLOGY

The background BC concentration in a location denotes the least loading of BC, which can originate from sources either within and beyond the region, either through continuous emission or continuous influx. The MMS method, developed by Watson and Chow (2001), was used in this study to segregate BC concentrations into two distinct groups: background BC (concentrations by eliminating short-term spikes) and local BC (comprising short-term spikes in the data above the background). The short-term spikes are regarded as the local contribution in this approach. The one-minute average BC values undergo smoothing at different time intervals (6 hours, 3 hours, 1.5 hours, 45 minutes, and 15 minutes), with the condition of retaining the lowest values at all times. The concentrations, which were smoothed after removing short-term spikes, were considered to represent the contribution of baseline (background) concentration of BC, following the methods outlined by Both *et al.* (2011), Kumar *et al.* (2018), Yeganeh *et al.* (2021), and Prabhu *et al.* (2022). More specifically, the term "background" is used to refer to the estimated baseline concentrations.

The local BC domain varies location-wise and seasonally, as wind speed is different in all locations and seasons. During the study period, the mean wind speed in India was 3.5 m s^{-1}. In a given area, the time it takes for a spike in wind speed to develop can range from approximately

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5 minutes to 2 hours. Hence, the approximate range of the local domain can vary from ~1 km to ~30 km. For more clarity on this, a discussion has been added in the supplementary section of this paper.

4 RESULTS

4.1 Sensitivity Analysis of the MMS Method

Both *et al.* (2011) and Kumar *et al.* (2018) previously employed the MMS method and noted that this approach primarily measures a relative proportion of background concentration, as it underestimates local emissions. In addition, it is not clear what should be the optimum averaging window to obtain reliable estimates of background concentrations.

A sensitivity analysis was carried out to address this issue. Fig. 1(a) shows the effect of applying different moving average window to the data varying from 1 hour to 168 hours. This analysis was performed to understand both the most optimal averaging window and potentially the usability of averaging beyond 1 day (as most satellite aerosol retrievals and model outputs have a repeat period or output frequency of about a day or higher unless otherwise specifically requisitioned). In every instance, the minimum values (either the observation or the moving average within the specific window) are selected to establish a baseline that represents background BC. The moving average data for the Aethalometer remains relatively stable ($<$ 5%) only after using an averaging window of approximately 48 hours or more. The background/regional concentration of BC was then determined by averaging all values of baseline data for the particular window. A diurnal representation of the segregation of BC into background and local using this approach is shown in Fig. 1(b).

4.2 The Baseline BC for Bhubaneswar

The modified MMS method mentioned in the previous section was applied to BC data over Bhubaneswar. As this study aims to apply this method on a regional scale using reanalysis data, a comparison is made between MERRA-2 BC and Aethalometer BC (both total and background). The BC data covers the period from 2018 to 2021 and is obtained from both the Aethalometer and MERRA-2. Fig. 2(a) shows the mean monthly four-year climatology of total and background BC over Bhubaneswar for the MERRA-2 reanalysis (solid orange lines) and surface-based observation datasets (solid black lines). In both datasets (MERRA-2 and Aethalometer), January has the highest concentration (4.70 µg m^{-3} and 8.16 µg m^{-3}), similar to coastal towns like Trivandrum and Kolkata (Babu *et al.*, 2002; Talukdar *et al.*, 2015). Aethalometer data shows a minimum BC concentration

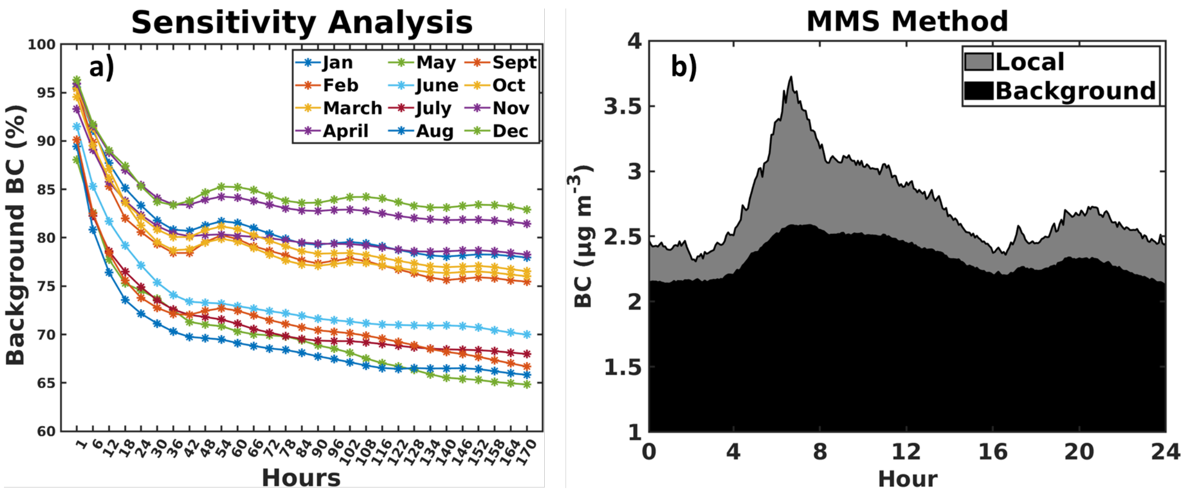

Fig. 1. The background BC mass fraction obtained after applying the MMS method to the ground-based BC measurements at Bhubaneswar, (a) Sensitivity of background mass fraction using different moving mean windows for different months, and (b) The diurnal segregation of background and local BC mass concentration.

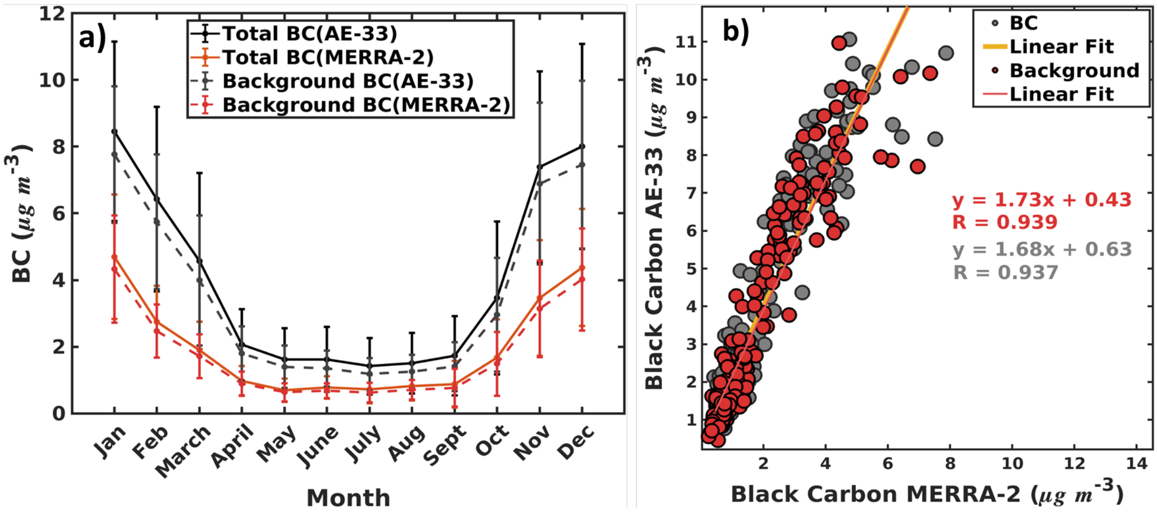

Fig. 2. (a) Seasonality of the relationship between MERRA-2 and Aethalometer-derived BC and background BC and (b) Scatter plot between the BC and background BC derived from Aethalometer and MERRA-2 analysis.

in July (1.44 µg m^{-3}), but MERRA-2 data shows the lowest BC concentration in May i.e., 0.70 µg m^{-3}, which is in the pre-monsoon season (March–May), as recorded in Bhubaneswar. Seasonally in winter months (December–February), BC reaches its highest recorded value of 7.63 µg m^{-3} in the Aethalometer and 3.94 µg m^{-3} in MERRA-2. Conversely, during the monsoon season (June–August), BC levels drop to a minimum of 1.57 µg m^{-3} and 0.78 µg m^{-3}, respectively. The higher atmospheric boundary layer height and precipitation-related wet scavenging may be the primary factors contributing to these low concentrations during the monsoon season. On the other hand, larger local fire-related emissions including biomass burning, and low atmospheric boundary layer height with reduced wet scavenging due to dry conditions lead to elevated levels of winter and sometimes post-monsoon (September–November) concentrations (Mahapatra *et al.*, 2014). Overall, high total BC was observed during winter and low during monsoon. Pre- and post-monsoon seasons exhibited large transitions between these high and low periods. It may be mentioned that we interchangeably use either specific months or seasons while discussing the results in this paper. Fig. 2(b) displays the correlation between MERRA-2 and Aethalometer total BC (grey circles) data showing a significant association, with a correlation coefficient of 0.94 for Bhubaneswar. Though there is a good correlation, there is a systematic low bias in MERRA-2 BC. In Fig. 2(b), the time frame of both datasets was averaged every week and then compared.

Overall, despite the low bias, MERRA-2 effectively captures the seasonality of BC over Bhubaneswar. The strong correlation between the Aethalometer and the reanalysis BC indicates the usability of MERRA-2 for understanding short-term variability and possibly seasonality on larger spatial scales. So, the MMS method was applied to MERRA-2 BC data to calculate background contributions in Bhubaneswar.

The monthly variation of background BC over Bhubaneswar calculated using MERRA-2 (dashed black line) and Aethalometer (dashed orange line) BC is shown in Fig. 2(a). For each of the two datasets, the concentration is consistently low from April to September and substantially higher in November, December, January, and February. The background BC for both Aethalometer and MERRA-2 follows their respective climatologies. In general, the winter season in Bhubaneswar is associated with the highest total (background) BC levels, measuring 3.94 (3.61) µg m^{-3} (MERRA-2) and 7.63 (6.99) µg m^{-3} (Aethalometer), while the monsoon season is characterised by the lowest levels, measuring 0.78 (0.68) µg m^{-3} (MERRA-2) and 1.57 (1.26) µg m^{-3} (Aethalometer). Fig. 2(b) illustrates the scatter plot between the Aethalometer and MERRA-2 total (grey circles) and background BC (orange circle). The background BC also follows a similar pattern as BC indicating that MERRA-2 is able to capture short-term variations (with a correlation coefficient of 0.94) in BC at the site quite remarkably.

Our study shows that the background BC fraction (in percentage) contributes more than ~90%

in the case of the Aethalometer and ~88% for MERRA-2 on an annual basis. Our analysis has shown that despite the differences/bias in the BC mass concentrations, both datasets exhibit similar seasonality and maintain good overall agreement, indicating that MERRA-2 can capture both the total and background BC. Apart from this study, only a few earlier studies attempted to estimate background/regional BC. Prabhu *et al.* (2022) estimated that the background BC in Bangalore is ~78% annually. Also, Yeganeh *et al.* (2021) in a West Asian city (Tehran) calculated background BC i.e., ~77%. We estimated the background BC for Bangalore and Tehran using MERRA-2 and found an exact match (~78%) for Bangalore and a close match (~79%) for Tehran indicating that the MMS method using MERRA-2 is able to capture background BC with remarkable accuracy (see Fig. S2 in supplementary file).

It may be noted that the objective of this comparison is only to evaluate the strength of the MMS method in using MERRA-2 to derive background BC and not to evaluate or validate the site-specific background BC. However, such close agreement in three different locations with significant differences in emissions and meteorology provides the confidence to extend the same analysis across the whole Indian region using MERRA-2 BC concentrations, as high spatial coverage of quality Aethalometer BC observations are unavailable in the public domain.

4.3 Background BC Concentration over India

Fig. 3(a) shows the background BC concentration over the whole Indian region. As discussed in Section 4.2, the highest spatial background BC is also observed during winter, followed by post-monsoon, pre-monsoon, and the lowest during monsoon. The most aerosol-laden region in India, the Indo-Gangetic Plain, is located in Northern India, and its background BC concentration is consistently high throughout the year potentially due to continuous emissions from biomass burning in Punjab and Haryana and the transportation of those and other emissions throughout the IGP (Fig. 3). When compared to the rest of India, the South is relatively cleaner. In the winter season, IGP and its neighbouring regions have extremely high concentrations of background BC (> 2.5 µg m^{-3}), while the West has intermediate concentrations ranging from 1.5 to 2 µg m^{-3}. Conversely, The Southern Indian states of Karnataka, Kerala, and Tamil Nadu exhibit notably reduced background BC levels, which range from 1.25 to 2 µg m^{-3}, and the central Indian region and specific states, such as Maharashtra (specifically Western Maharashtra), Telangana, and Northern Karnataka, exhibit exceptionally high levels of background BC ranging from 2.2 to 2.5 µg m^{-3} exclusively during the winter season. Also, Eastern India, Odisha, and Central India show comparatively lower background BC during the winter season. In the post-monsoon season relatively less background BC is observed over IGP and surrounding areas compared to winter. However, IGP (2 to > 2.5 µg m^{-3}) and states in central India such as Madhya Pradesh, Chhattisgarh, eastern Maharashtra, and eastern Gujarat (1.25–2 µg m^{-3}) exhibit a high background concentration in the post-monsoon season. In the summer season (MAM), the background is less compared to winter and post-monsoon; only IGP (1–1.75 µg m^{-3}), central India (Western Maharashtra, Telangana, i.e., 1–1.25 µg m^{-3}), Odisha (1–1.5 µg m^{-3}), West Bengal (2–2.5 µg m^{-3}), and Eastern India show the highest amount of background BC. During the monsoon season, background BC concentration is less than 1 µg m^{-3} in the Indian region except IGP (1–1.75 µg m^{-3}). This is because of the washout of BC due to continuous rainfall during this season.

Fig. 3(e) shows the seasonal mean BC from MERRA-2 and background BC over India, both in terms of absolute value and percentage. winter season has the highest background BC (2.32 µg m^{-3}), followed by the post-monsoon season (1.54 µg m^{-3}), pre-monsoon season (0.94 µg m^{-3}), and monsoon season (0.60 µg m^{-3}). But in terms of percentage, winter has the highest (~87%), then post-monsoon (~85%), pre-monsoon (81%), and monsoon (~79%). Annually, background BC is ~83% in the Indian region. Overall background BC is very high in the Indian region, which shows the spreading of BC throughout India, which depends on the prevailing meteorological conditions.

Overall, the results described here appear promising. In the present analysis, as the whole Indian region has been taken into consideration, which has varying geographical settings with complicated spatiotemporal BC variabilities, high-resolution datasets are needed for the validation of background BC concentrations at each grid location. However, except for a few studies (five, to the best of our knowledge), the literature lacks similar studies for comparing and contrasting. However, as an experiment, the possibility of similarity between the foreground (total minus

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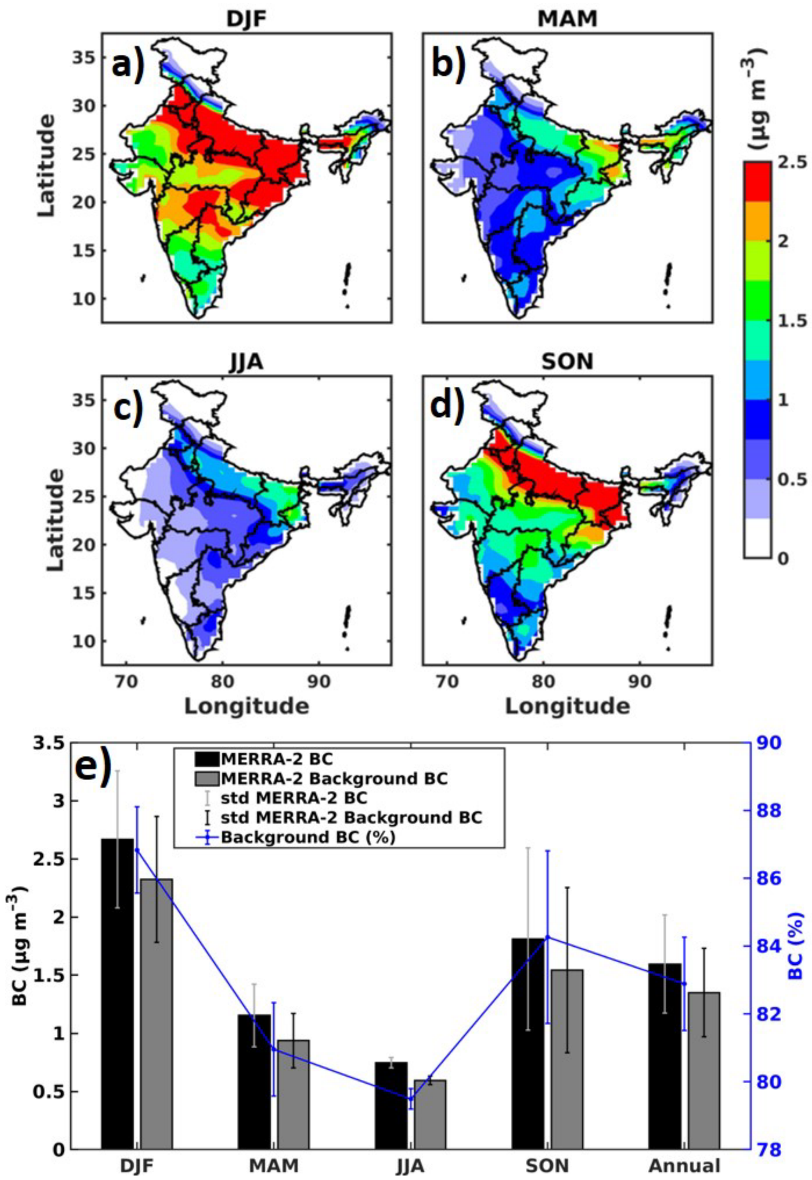

Fig. 3. Background BC concentration over India during (a) DJF (winter), (b) MAM (pre-monsoon), (c) JJA (monsoon), (d) SON (post-monsoon) season, and (e) bar plot showing the seasonal and annual total and background BC over India.

background BC concentration and known BC emissions from a widely used emission inventory is used to qualitatively evaluate our findings in the next section.

4.4 Qualitative Comparison of Foreground BC with HTAP v_3 Emissions

Fig. 4 shows the seasonal foreground BC concentration and emissions from the HTAP version 3 emission inventory from 2013 to 2018. As mentioned in the earlier section, the foreground BC concentrations during different seasons have been estimated by subtracting the respective background BC concentrations from the total BC loading during the respective season from 2013 to 2018. The foreground BC concentration is expected to show what is left after the removal of the background and therefore the concentrations from local emissions. Whereas, the emission inventories, on the other hand, give local emissions based on their temporal frequency (monthly in the case of HTAP v3). Thus, the comparison of foreground BC concentrations (after removal of background) with the emission inventories done here in this section is expected to indirectly show the strength of MMS method in capturing background BC at large spatial scales.

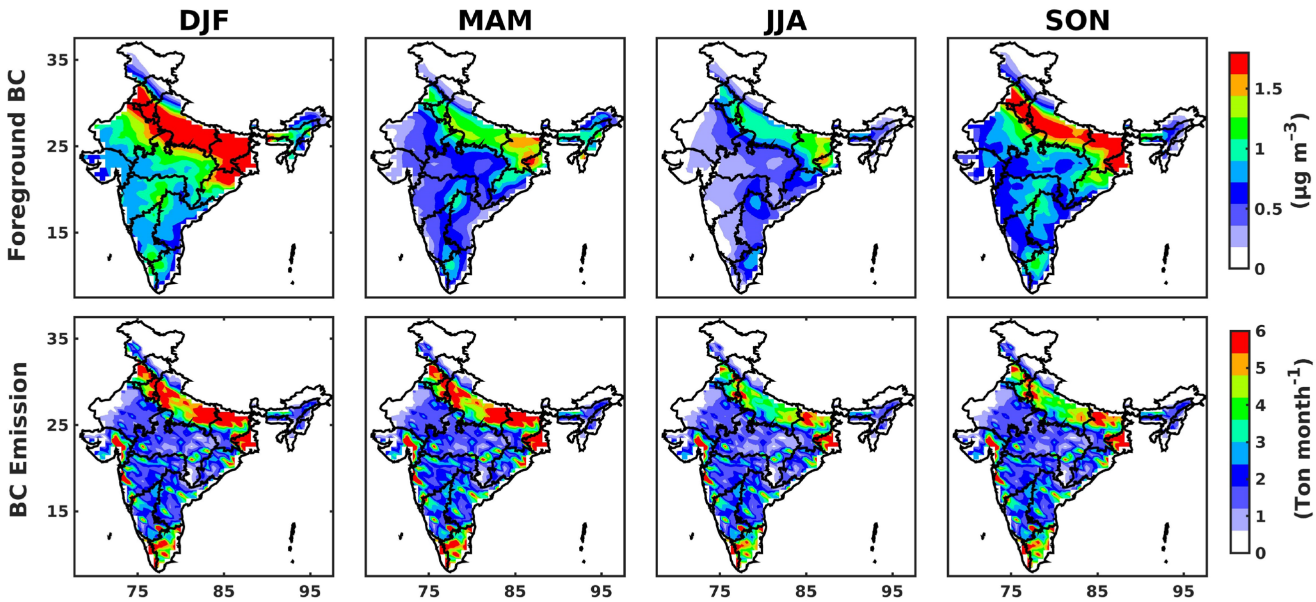

Fig. 4. The foreground (a–d) BC concentration and (e–h) BC emission over India in DJF (winter), MAM (pre-monsoon), JJA (monsoon), and SON (post-monsoon) seasons.

Fig. 4 shows the qualitative correspondence between the foreground BC and emission inventories for all seasons. The striking spatial similarity between foreground BC concentration and seasonal emissions from HTAP v3 shows that the MMS method employed in this study is able to capture and remove background BC thereby enabling the extraction of information about local emissions and therefore the concentrations. However, it may also be noted that there are some minor differences between the foreground and emission inventory maps over Western, Eastern, Central India, and North Indian (IGP) regions (discussed in the discussion section). The foreground BC is very high throughout the IGP, and the major sources like Delhi, Haryana, Punjab, and parts of West Bengal are very prominent. Other than the IGP, the Eastern part of Maharashtra, and the Northern part of Telangana, which includes Hyderabad and Bangalore, the Western part of Tamil Nadu can be consistently seen as a prominent source. Gautam *et al.* (2011) and Rana *et al.* (2019) also showed that the IGP is the primary location for BC emissions in India. The emissions shown in coastal regions are also not well captured in the foreground BC. Detailed analysis in the future using such methods as described could provide indications of either strengths and/or weaknesses of models/inventories.

5 DISCUSSION

The MMS method employed in this study is unique and offers multiple advantages. Firstly, the method is both computationally efficient and simple to implement. Another advantage is that the MMS method provides consistent results with emissions derived from a combination of local emission inventory and air quality models, which are difficult to achieve by routine monitoring and source apportionment methods (Kumar *et al.*, 2018). On the other hand, the MMS approach also has significant drawbacks. Firstly, datasets with very high temporal resolution, at least sub-hourly, ground-based measurements are required to apply the MMS method. High-resolution observational datasets are lacking in many regions around the globe, prompting the utilization of reanalysis products such as MERRA-2. Another constraint is that the MMS technique underestimates local emissions, as reported by Both *et al.* (2011) and Kumar *et al.* (2018) using an averaging window of less than 360 minutes. Hence a modified MMS method is utilized in both studies that incorporate an underwriting function. Our study clearly shows that an averaging window of at least two days is required to stabilize the background concentrations within ± 5% thereby avoiding underestimation of local emissions/concentrations. Further drawbacks of the MMS method include its failure to account for boundary layer dynamics, meteorological conditions, and topographical effects, particularly in this study given that it has never been used in a large area with such a wide range of topography and meteorological conditions. However, despite all these limitations, our study using the MMS approach effectively captures the variability of background and foreground emissions reasonably well. However, more investigations as follow-up studies will reveal the strengths and potential weaknesses if any in the future. Past studies such as Apte *et al.* (2011), Both *et al.* (2011), and Kumar *et al.* (2018) focused on $PM_{2.5}$ in their studies. They indicated that regional contributions were about 9 times higher than local contributions, varying between 80 and 90% in their respective study areas. Our study indicates that this may be a slight overestimation. Such insights are possible without high-frequency ground-based measurements along with freely available datasets such as MERRA-2 BC can be a boon for researchers limited by the lack of ground-based measurements in the developing world.

In addition, the foreground BC, obtained by subtracting the background BC from the mean BC data in Section 4.4, consistently reveals the Indo-Gangetic Plain (IGP) as the predominant source of BC throughout the year, aligning with HTAP v3 emission data. This indicates that this method in conjunction with the availability of local knowledge could be used to even identify missing sources in large-scale emission inventories. Numerous prior studies have consistently identified the IGP as a significant contributor to BC emissions. This emanates from diverse anthropogenic activities, including the burning of crop residue and biomass, forest fires, vehicular emissions, brick kilns, and coal-based power plants (Arif *et al.*, 2018; Chauhan and Singh, 2017; Vinoj and Pandey, 2016; Gadhavi *et al.*, 2015; Kaskaoutis *et al.*, 2014; Praveen *et al.*, 2012; Kharol *et al.*, 2012; Paliwal *et al.*, 2016; Pandey and Venkataraman, 2014; Prasad *et al.*, 2018; Ram and Sarin, 2015; Ramachandran and Cherian, 2008; Rehman *et al.*, 2011; Sahu *et al.*, 2008; Sarkar *et al.*, 2018; Saud *et al.*, 2012). In the foreground BC maps (Fig. 4), major metropolitan cities such as Kolkata, Delhi, Kanpur, Hyderabad, Bangalore, and Chennai prominently emerge as significant contributors to BC across all seasons. Consistent with earlier studies by Apte *et al.* (2011), Aruna *et al.* (2013), Dumka *et al.* (2013), Prabhu *et al.* (2022), Talukdar *et al.* (2015), and Thamban *et al.* (2017), these urban areas consistently exhibit high BC concentrations. However, there are some visible misses too. For example, the major metropolitan city of Mumbai does not appear as a significant source in the foreground BC discussed in Section 4.4, despite its presence in the emission inventory and reported high BC concentrations in a study by Sandeep *et al.* (2013). This is true for the whole coastal regions across Western and Eastern India and hence requires additional studies to explore the reason for the limitation of the MMS method. During the post-monsoon season, major biomass-burning sources in Punjab, Haryana, and Western Uttar Pradesh, as reported by Venkataraman *et al.* (2006), are evident in both datasets. Notably, the Northwestern part of Telangana and the Western part of Maharashtra emerge as prominent BC sources in Central India in MERRA-2 foreground BC but are not observed in the HTAP v3 database, highlighting potentially either a limitation of the emission database or the MMS method over the central Indian region. Recent studies have indicated the increasing strength of aerosol emissions over the Central Indian region (Thomas *et al.*, 2019). Thus, are these missing foreground BC hotspots a limitation of the MMS method or the emission database? That needs to be further explored.

In summary, our analysis demonstrates the usefulness of MERRA-2 reanalysis data in accurately capturing the spatial and temporal fluctuations of BC. This data exhibits comparable characteristics of background BC to those observed on the ground. Furthermore, this study determined that a 48-hour averaging period is suitable for determining the most consistent level of BC in a specific area. This finding is particularly relevant for reanalysis datasets such as MERRA-2, as it provides a practical and computationally efficient method for obtaining the baseline BC concentration in any given location. Utilising this approach reveals that background BC constitutes a substantial proportion of India's total BC pollution, comprising over 75%. Providing a broader understanding of BC loading in the Indian region allows us to effectively tackle BC pollution in India. The high background BC also reveals that reducing local emissions alone cannot effectively decrease pollution in a location if the background concentrations are already very high. It necessitates a collaborative and coordinated effort from a broader area, including the entire region. This understanding can prove highly advantageous for India's ambitious National Clean Air Program (NCAP), which aims to mitigate pollution in cities with elevated pollution levels. BC being an important component of particulate air pollution and emitted by some of the same anthropogenic sources points to the possibility of being used to understand particulate pollution dynamics over the Indian region. The findings and method used in this study could be innovatively used for providing insights into potential strengths and weaknesses in the widely used emission inventories, reanalysis, and model simulations for both BC and other particulate matter air pollutants.

6 CONCLUSIONS

I. The MMS method is applied to determine the background contribution of BC over the Indian region. It is found that using an averaging window of 48 hours or more results in the estimation of consistent background concentration within ± 5%.

II. MERRA-2 dataset shows an absolute value for background BC of ~3.6 µg m^{-3}, while the Aethalometer dataset shows a level of ~7.0 µg m^{-3} during winter (DJF). On the other hand, the monsoon (JJA) season exhibits the lowest background BC levels, with the MERRA-2 (0.68 µg m^{-3}) and the Aethalometer (1.26 µg m^{-3} showing significant underestimations in absolute values. However, both datasets show similar background BC in terms of percentage of over 85% annually demonstrating a significant degree of agreement over Bhubaneswar.

III. On a regional scale MERRA-2 data showed high levels of background BC over the Indian region throughout the year, with values exceeding 75%. The background percentage exhibited a comparable seasonal pattern to that of background BC, with peak levels observed during winter and the lowest levels during the monsoon season. Black carbon (BC) consistently has background concentrations over 80% throughout the year in most areas, indicating its wide distribution across a large geographic region.

IV. A qualitative comparison between the black carbon (BC) after the removal of background BC (referred to as foreground BC) using the HTAP v3 emission database demonstrates a notable regional similarity. This similarity is particularly evident over the Indo-Gangetic Plains and Southern India, highlighting the effectiveness of the MMS method in isolating background BC. Nevertheless, the approach also uncovers disparities in the central and coastal regions of India, suggesting potential inconsistencies in either the MMS method or the emission inventories. Future research should investigate these differences.

The findings of the study reveals that India experiences significantly high levels of background BC. Steps to mitigate BC emissions should consider synergistic strategies that address emission sources at a regional scale and not over isolated pockets such as individual cities or states. In addition, long-range transport of these aerosols needs to be studied systematically using innovative measurement and modeling tools for enacting targeted mitigation strategies having the largest benefit on a regional scale. Such systematic efforts focusing not just on BC, but other air pollutants are expected to help scientifically driven strategies for clean air under India's ambitious National Clean Air Program.

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SUPPLEMENTARY MATERIAL

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