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RESEARCH ARTICLE

Black carbon over tropical Indian coast during the COVID-19 lockdown: inconspicuous role of coastal meteorology

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Abstract

Black carbon (BC) aerosols critically impact the climate and hydrological cycle. The impact of anthropogenic emissions and coastal meteorology on BC dynamics, however, remains unclear over tropical India, a globally identified hotspot. In this regard, we have performed in situ measurements of BC over a megacity (Chennai, 12° 59′ 26.5″ N, 80° 13′ 51.8″ E) on the eastern coast of India during January–June 2020, comprising the period of COVID-19-induced strict lockdown. Our measurements revealed an unprecedented reduction in BC concentration by an order of magnitude as reported by other studies for various other pollutants. This was despite having stronger precipitation during pre-lockdown and lesser precipitation washout during the lockdown. Our analyses, taking mesoscale dynamics into account, unravels stronger BC depletion in the continental air than marine air. Additionally, the BC source regime also shifted from a fossil-fuel dominance to a biomass burning dominance as a result of lockdown, indicating relative reduction in fossil fuel combustion. Considering the rarity of such a low concentration of BC in a tropical megacity environment, our observations and findings under near-natural or background levels of BC may be invaluable to validate model simulations dealing with BC dynamics and its climatic impacts in the Anthropocene.

Keywords Aerosols · Black carbon · Fossil-fuel emissions · Continental air · Biomass burning · Marine air · Transport · COVID-19 lockdown · Pollution · India

Introduction

Aerosol concentrations and spectral characteristics critically influence the air quality and hence mortality rates (Apte et al. 2015), as well as profoundly impact the Earth's radiation budget and climate (Lelieveld et al. 2015; Ramanathan et al. 2001). The tropical Indian region is a global hotspot of enhanced aerosol loadings and among few regions of the world where aerosol concentration has been constantly increasing (Babu et al. 2013; Krishna Moorthy et al. 2013; Ramachandran et al. 2020). Aerosol dynamics over India manifest complex influences of strong anthropogenic and biomass burning emissions and diverse natural processes (Babu et al. 2016; Ojha et al. 2020; Singh et al. 2020a). Additionally, warm and humid tropical climatic conditions and mesoscale coastal meteorology also have considerable impacts on the spatial and temporal distributions of aerosols and their spectral characteristics (Moorthy et al. 2003; Ratnam et al. 2018). Despite numerous experimental efforts, the role of coastal meteorology and regional emissions remained

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unclear over this part of the world owing to large biases between chemical transport models and in situ observations resulting from lack of observations, which will delineate the impact of anthropogenic emissions. Further, the aerosol concentrations remain typically elevated over urban megacities and downwind, which inhibit the sampling of variations in the regional background in order to observationally constrain the effects of emissions. At times, the high-altitude sites have been recommended as a proxy to the background/natural pollutant concentrations. The differences between emission rates, boundary layer dynamics and impacts, rate of transport and removal, and very strong differences between meteorology, however, preclude such an assumption. Therefore, to understand the influence of human-induced pollution on climate, near-natural condition measurements are essential.

While the need for baseline measurements is extremely important, it is, however, very impractical to shut down anthropogenic emissions significantly under the business-as-usual scenario. Recently, an unprecedented lockdown imposed to contain the spread of the COVID-19 pandemic over various regions of the globe, including India, offered such a rare situation. Several studies reported that comprehensive lockdowns implemented for extended periods dramatically impacted the air quality globally (Kroll et al. 2020; Rodríguez-Urrego and Rodríguez-Urrego 2020; Venter et al. 2020) and regionally (Agarwal et al. 2020; Berman and Ebisu 2020; Menut et al. 2020; Sharma et al. 2020; Singh et al. 2020b). Stringent lockdown forced the public to stay indoors, resulting in a near-zeroing of the emissions from fossil-fuels and other anthropogenic burning sources over different regions of the globe. India also observed a very comprehensive and stringent lockdown restricting anthropogenic activities for an extended period: 24th March to 30th June 2020. This lockdown offered a unique opportunity to investigate the near baseline variations in various pollutants over the tropical Indian conditions with negligible modulations by anthropogenic factors. Several studies have been carried out to study the lockdown impacts on surface-level air quality as well as the tropospheric columns of pollution loadings over the Indian region (Dhaka et al. 2020; Kumari and Toshniwal 2020; Lokhandwala and Gautam; 2020; Mahato et al. 2020; Naqvi et al. 2020; Pandey and Vinoj 2021; Sharma et al. 2020; Singh and Chauhan 2020; Singh et al. 2020b). These studies showed a remarkable decline in the levels of various gas-phase and particle-phase trace species across the Indian region. Tropical megacity and India's capital, Delhi, observed a counter-intuitive enhancement in fine particulate matter, which is attributed to a remarkable interplay between meteorology and baseline emissions (Dhaka et al. 2020). This, along with few other studies (Li et al. 2020; Su et al. 2020; Wang and Zhang 2020), highlights the need to investigate other distinct geographical locations with their own peculiar meteorology in order to assess the range of effects such interactions can offer. Tropical coastal setup offers one such distinct atmosphere where the effects of coastal meteorology need to be delineated in the observed aerosol dynamics. Studies on the impacts of anthropogenic and biomass burning sources on BC with segregated continental and marine air masses have still been lacking.

In the discussed scenario, we have performed continuous measurements of aerosol spectral absorption and meteorological parameters at a tropical urban coastal location, Chennai (12° 59′ 26.5″ N, 80° 13′ 51.8″ E, 6 m above mean sea level) in India (Fig. 1a) from 21 January 2020 to 30 June 2020. The Chennai megacity region experiences typical mesoscale coastal dynamics, comprising land and sea breezes, transporting continental and marine air to the sampling site (see the "Methods" section for additional details). The BC measurements have been segregated into BC in the marine air mass and BC in the continental air mass by combining the air mass history and the onset of sea-breeze and land-breeze (Hari Prasad et al. 2019; Raj et al. 2002; Reddy et al. 2021). Analysis has been performed for both pre-lockdown (21st January–23rd March 2020) and lockdown (24th March–30th June 2020) periods, separately. Figure 1a shows a representative schematic of the continental and marine air masses arriving at the measurement site. The continental air masses travel and meander over more than 600 km of the landmass to reach the measurement site and therefore are well exposed to the regional emissions of peninsular India before reaching the measurement site. In contrast, the marine air masses reside entirely over the Bay of Bengal and reach the measurement site without any considerable and strong anthropogenic influences.

Methods

Measurements of BC and meteorological parameters have been conducted from the rooftop of Mechanical Science Block (MSB) in the Indian Institute of Technology, Madras campus (12° 59′ 26.5″ N 80° 13′ 51.8″ E, 6 m above mean sea level) in Chennai, India. This site is centrally located in the institute campus and is suggested to be a representative of the coastal urban background conditions of this region. The institute campus occupies an area of 2.5 km^2 (617 acre) and was formerly a part of the adjoining Guindy National Park. The sea coast is approximately 5–7 km from the measurement site. Figure 1a shows the location of Chennai (greet dot) in peninsular India on the India map.

Aerosol spectral absorption measurements have been performed employing a seven-channel dual-spot Aethalometer (Model AE-33, Magee Scientific, USA) (Drinovec et al. 2015; Hansen et al. 1984), set up in an air-conditioned laboratory in the MSB. The samples are drawn from a height of ~ 15 m above the ground and on the rooftop of the MSB. The flow rate

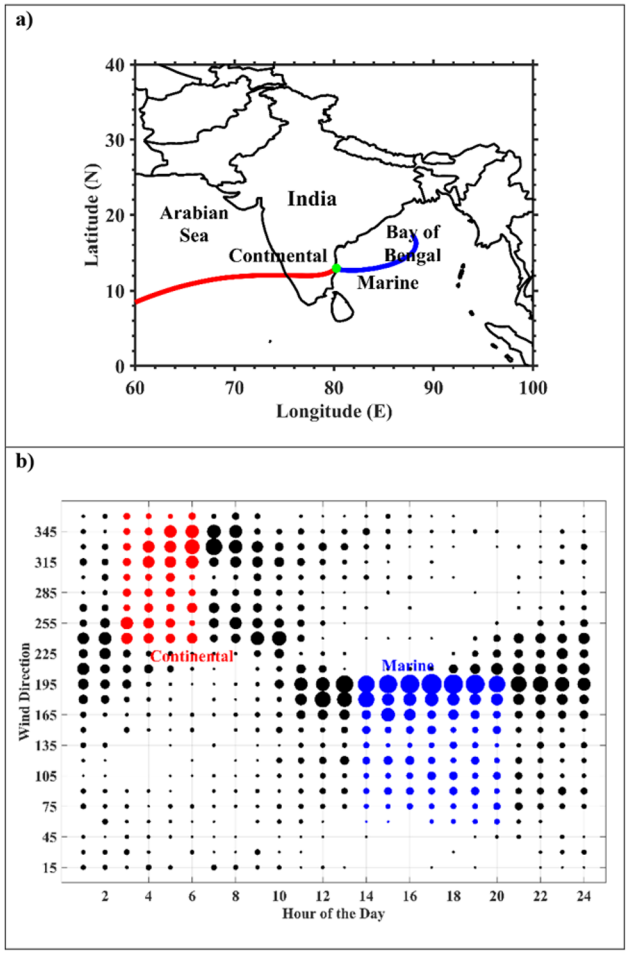


Fig. 1 **a** Schematic of the location of the measurement site (green dot) on the Eastern coast of India. Superimposed on this are two major and predominant air masses, continental (red), and marine (blue), arriving at the measurement site. The air masses were classified based on air mass history and wind direction, as shown in **b**. **b** Classification of the two air masses—(i) marine air mass arriving at the site between 14 and 20 h and with wind directions between 60 and 195° (blue dots); (ii) continental air mass arriving at the site between 3 and 6 h and with wind directions between 240 and 360° (red dots). The size of the dots indicates the frequency of occurrences in a given hour and over the entire wind direction range, with the smallest dot representing 1% and the largest dot 60%

is maintained at about 2 LPM, and the temporal resolution of measurements is 60 s. The instrument primarily measures the aerosol spectral light absorption which has been converted to BC mass concentrations using a mass absorption cross section value of 7.77 m^2/g at 880 nm (Drinovec et al. 2015; Liousse et al. 1993). Measurements by the dual-spot Aethalometer have been shown (Drinovec et al. 2015; Vaishya et al. 2017) to minimize the artifacts caused by multiple scattering and filter loadings (Collaud Coen et al. 2010; Drinovec et al. 2015; Lack et al. 2014; Müller et al. 2011; Segura et al. 2014; Vaishya et al. 2017). Uncertainties in the measurement of the aerosol light absorption coefficient by legacy Aethalometers are estimated (Lack et al. 2014) to be in the range 12–30%; however, for dual-spot Aethalometer used in the present study, the uncertainties are estimated (Vaishya et al. 2018) to be ~ 10%.

An automatic weather station (AWS) (CLIMA US SENSOR 4.9200.00.000, Thies CLIMA, Germany) has been used to measure various meteorological parameters. Meteorological measurements have been recorded at a temporal resolution of 1 min and have been converted into an hourly average for the analysis. The hours which had at least 40% valid measurements have been considered in the analysis. Figure S1 in the supplementary section shows the time series of the meteorological parameters, viz., wind direction, wind speed, relative humidity, pressure, dew point, and temperature. Also shown in figure S1 is BC mass concentrations color-coded with prevailing wind direction. Figure S2 in the supplementary section shows the time series of aerosol absorption parameters such as BC mass concentration, biomass burning percentage, and aerosol spectral absorption.

Two contrasting air masses influencing the measurement site have been defined as follows:

i) Marine air mass arriving at the site between 14 and 20 h local time with wind directions between 60 and 195° (shown with blue dots in Fig. 1b).
ii) Continental air mass arriving at the site between 3 and 6 h local time with wind directions between 240 and 360° (shown with red dots in Fig. 1b).

The size of the dots in Fig. 1b indicates the frequency of occurrences, with the smallest dot representing 1% and the largest dot representing 60%. The identification of air masses is based on long-term meteorological observations and analysis (Raj et al. 2002) and robust datasets from micro-pulse lidar, radiosonde, and surface meteorological measurements (Reddy et al. 2021). Further, it was shown using Lagrangian transport modeling and weather research and forecasting (WRF) model that marine air mass has significant inland penetration (Hari Prasad et al. 2019).

In order to contain the spread of the COVID-19 pandemic, an unprecedented, stringent, and comprehensive lockdown was imposed in India from 24th March to 30th June 2020. This resulted in minimal anthropogenic activities for an extended period of time. In the present study, measurements and data pertaining to the phase before the lockdown was imposed are termed as pre-lockdown (21st January–23rd March 2020). The phase during which COVID-19 restrictions were imposed is termed as the lockdown (24th March–30th June 2020).

Results and discussion

Time-series variations of general meteorological features and BC are shown in Figure S1. It can be seen that while there was a general increase in temperature, from 25 to 30 °C to 30

to 35 °C, over the measurement period, i.e., from January to June, 2020, pressure over the region decreased from ~ 1015 to 1020 mbar to ~ 1010 to 1015 mbar. Wind speed and relative humidity remained between 1 and 3 ms^{-1} and 60 and 80%, respectively. A general decrease in the BC mass concentration is seen from ~ 10 to 1 $\mu g/m^3$. Higher BC concentrations were mostly from the continental sector. Figure S2 shows time-series of two additional parameters, BB%, and Alfa abs., pertaining to spectral aerosol absorption properties. While BB% range changed from 5 to 15% to 15 to 30%, Alfa Abs. range changed from 0.8 to 1.0 to 1.2 to 1.4. As will be seen in the following sections, these parameters indicate a change in aerosol absorption properties and also possible changes in source type.

Aerosol spectral characteristics

Figure 2 shows the spectral variation of aerosol absorption coefficient (Mm^{-1}) and compensation parameter (k) for marine and continental influenced air mass during the pre-lockdown and lockdown periods. Aerosol absorption, owing to higher BC concentrations, is significantly stronger during the pre-lockdown conditions compared to the lockdown conditions. Further, strong contrast was observed in the spectral absorption between the continental and marine air masses during the pre-lockdown. Absorption was most pronounced (0.16–0.18 Mm^{-1}) in the continental air masses, particularly in the shorter wavelengths (400–500 nm) during the pre-lockdown, indicating large absorption aerosol loading in the continental air masses. Implementation of the intense COVID-19 lockdown exhibited a remarkable impact on the absorption spectra with a dramatic reduction in the aerosol absorption during the lockdown period across all the wavelengths (370–950 nm). Additionally, in contrast with the pre-lockdown period, there was little difference in the magnitude and the spectral nature of the absorption between marine and continental regimes. This clearly indicates that the dominant absorption aerosols in the two air masses were possibly of the same origin or from a single source. The corresponding slopes of the absorption aerosol spectra for the marine and the continental sectors were ~ 1.06 during pre-lockdown, which increased to 1.15 during the lockdown phase. Absorption aerosol spectra slope indicate the possible sources of origin (Kirchstetter et al. 2004). Slope values of one indicate absorption dominated by aerosols from fossil fuel sources and values higher than one indicate increasing presence of aerosols from biomass burning sources. Thus, the slope values of 1.06 and 1.15

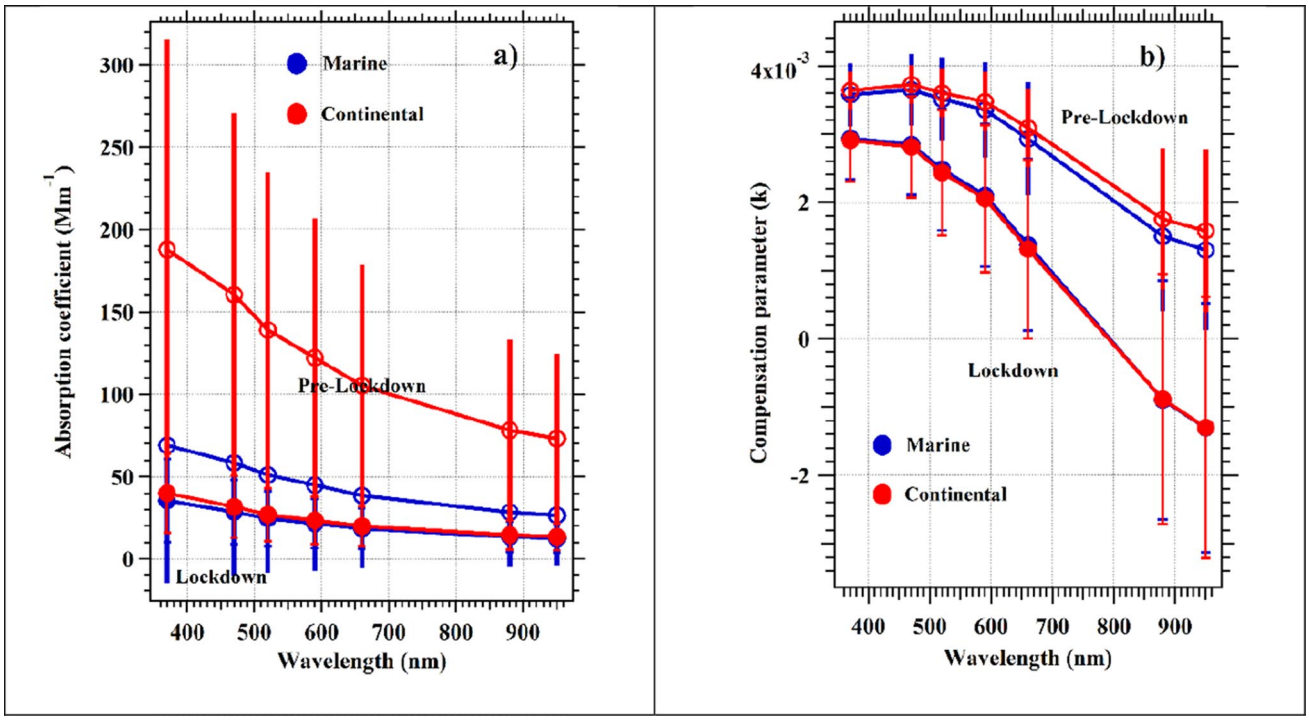


Fig. 2 Spectral variation of BC aerosol characteristics derived from AE33 measurements. **a** Aerosol absorption coefficient (Mm^{-1}) and **b** compensation parameter (k) in the marine sector (blue circle) and in the continental sector (red circle) during pre-lockdown phase (21st January–23rd March 2020) (open circle) and the lockdown phase (24th March–30th June 2020) (solid circle) of the COVID-19-induced pandemic. Thick vertical bars are standard deviation for pre-lockdown episodes, and thin vertical bars with a horizontal cap are standard deviation for lockdown episodes. Duration and other details related to pre-lockdown and lockdown phases are defined in the "Methods" section

indicate aerosols from different sources and type, in the two phases. The pre-lockdown phase appeared to be dominated by aerosols from fossil fuel and biomass burning sources, and the lockdown phase was dominated particularly by aerosols from biomass burning sources. The higher values of the compensation parameter (k), an indicator of the aging of aerosols, suggests freshly emitted and highly absorbing aerosols, whereas lower values indicate aged aerosols, which are generally highly scattering in nature. Aerosols age as they get transported over longer distances, and hence, higher k values are also indicative of locally emitted aerosols and having high absorption, versus low k values indicating transported aerosols with increasing scattering characteristics with aging and transformation. From Fig. 2b, it can be seen that k values during pre-lockdown are found to be considerably higher (mean 0.002 with a 95% confidence interval of 0.0002), suggesting that fresh emissions from local to regional emission sources dominated the aerosol build up over this part of the globe. In contrast, mean k values during the lockdown were − 0.0007 with a 95% confidence interval of 0.0003. As also in the case of absorption coefficients, there is a difference between continental and marine air during the pre-lockdown phase wherein aerosols are more aged over the marine sector; in contrast, transported and aged aerosols dominated in both continental as well as marine sectors during the lockdown phase. An interesting inference from the spectral nature of k is that biomass burning remained a stable local source during both periods. However, BC from biomass burning sources were present during the pre-lockdown period albeit the aerosol population was dominated by aerosols from fossil fuel sources. In over 50% of the cases, during pre-lockdown, biomass burning percentage was in the range 5–15%. BC emitted from fossil fuel sources appeared to be dominated by long-range transport during the lockdown phase as local emissions due to fossil fuel combustion were nearly absent from the scenario.

Black carbon (BC) dynamics and sources

Figure 3 shows the time series of daily averaged BC mass concentration (μg/m^3) during marine (blue) and continental (red) air masses (and also the overall period in yellow) presented for the entire study period. Measurements reveal an unprecedented reduction in BC mass concentration almost by order of magnitude during the COVID-19 lockdown. Such reductions in BC concentrations have also been observed earlier at specific locations (Kompalli et al. 2014), albeit during a national strike. BC concentration in the continental air masses dropped more abruptly as compared to the marine sector. Nevertheless, the reductions of BC in both air masses are found to be significant, with BC reductions of an order of magnitude from about 10 to 2 μg/m^3. Welch's unequal variance t-test on the BC data indicates that the reductions in the BC from pre-lockdown to lockdown are statistically significant, with the highest p value of 0.0003 for the continental sector. It is to be noted that such strong contrast in BC mass concentration was observed despite more frequent heavy rain-induced significant BC washout during the pre-lockdown period (cumulative rain of ~ 3000 mm). Rainfall events were fewer in numbers and less intense, with cumulative rainfall of ~ 1500 mm, during the lockdown phase. The reduction in the intensity and amount of rain during the lockdown is puzzling. It is likely linked with a reduction in cloud precursors (Andreae and Rosenfeld 2008), i.e., aerosols, which are the potential

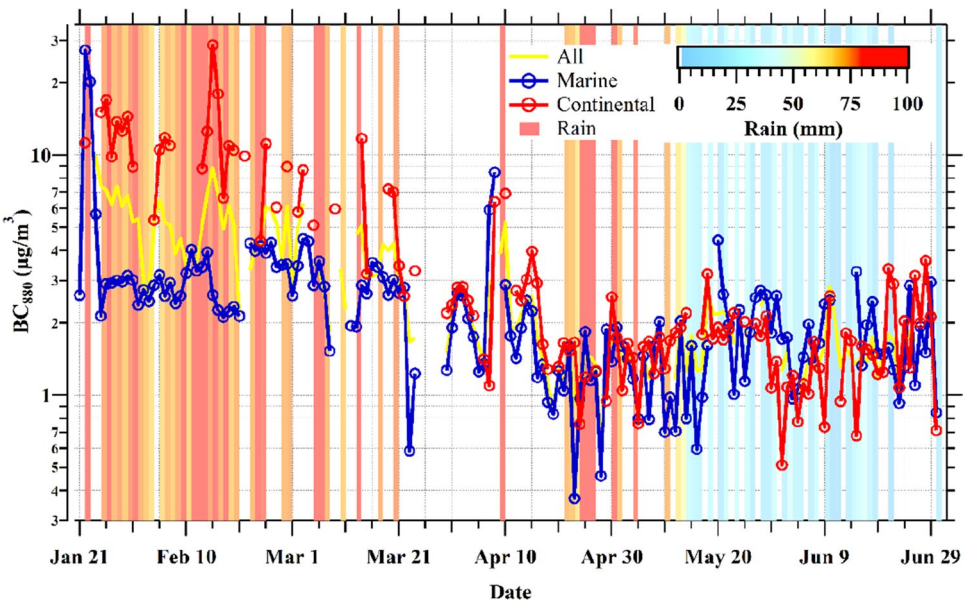


Fig. 3 The time series of black carbon (BC) mass concentration derived for the characteristic wavelength of 880 nm. The BC mass concentration is segregated by the dominance marine and continental air mass, which are separated as mentioned in Fig. 1 caption. The blue color indicates the marine sector, whereas continental sector is indicated by red. The predominant air masses which did not satisfy the criteria mentioned in Fig. 1 are termed as overall and denoted by yellow. The vertical lines in the panel indicate the rainfall amount in mm as scaled by the color bar shown in the figure

source of the cloud condensation nuclei. However, such changes in the rainfall pattern and amount might also be due to lockdown-imposed changes in surface energy balance (Diffenbaugh et al. 2020; Schwarz et al. 2020; Yang et al. 2020) or perturbed boundary layer stability.

Spectral aerosol absorption index (α), a parameter connecting the absorption coefficient and wavelength, is indicative of fossil fuel combustion aerosols as primary contributor to absorption aerosols (for $\alpha \sim 1$) and aerosol from biomass burning sources (for $\alpha > 1$) (Kirchstetter et al. 2004). The peak of α frequency distribution is observed to be shifted from 1.0 to 1.2 from pre-lockdown to the lockdown phase, with a decline in occurrences from ~ 40 to ~ 25% (Fig. 4a). This finding reveals a change in the aerosol source regime from fossil-fuel dominance to biomass burning dominance. We suggest/hypothesize that this change in α distribution is due to the cessation of fossil-fuel sources during the lockdown, whereas the biomass burning sources remained to their normal strength. This is further elaborated in Fig. 4b, showing the relative increase in the biomass burning contribution from ~ 10% during the pre-lockdown to ~ 15% during the lockdown. Assuming that absorbing aerosols over this region were from only two sources, i.e., fossil-fuel and biomass burning, we bifurcated the BC concentrations into these sources (Sandradewi et al. 2008; Vaishya et al. 2017). BC from fossil-fuel sources decreased from 4.5 ± 1.8 to 1.5 ± 0.6 μg/m^3, wherein those from biomass burning sources decreased from 0.5 ± 0.2 to 0.3 ± 0.4 μg/m^3. The detailed differences are listed in Tab. S1.

During the pre-lockdown phase, the peak in BC mass concentrations reached up to ~ 50 μg/m^3, in the continental sector, with α predominantly being around 1.0, indicating absorbing types of aerosols from fossil-fuel sources (Fig. 5a). A closer look at the polar plot during the pre-lockdown phase reveals that higher BC mass concentrations in the continental sector and higher α values indicated relatively higher contributions from the biomass burning sources. On the contrary, marine sector BC values are well within the 50 μg/m^3 range and dominated by fossil-fuel sources. The lockdown phase saw reductions in BC mass concentrations by up to an order of magnitude in the continental sector and ~ 50% in the marine sector (Fig. 5b). Also, the absorbing aerosol sources, which were dominated by fossil-fuel sources during the pre-lockdown phase, saw a relative enhancement in contribution from biomass burning sources during the lockdown phase. This relative enhancement in aerosols, from the biomass burning sources, was observed in both the sectors, marine as well as continental, but was more prominent for the continental sector, where the strict lockdown meant minimal fossil-fuel dependent aerosol sources (cars, heavy vehicles, bikes, etc.) were still active.

The spatial occurrences of sea breeze and land breeze were sharper during the lockdown period (see supplementary figure S3). While sea breeze occurrences were uniformly spread over the wind direction from 60 to 195° during pre-lockdown, during lockdown, majority of its occurrences were confined to 180–195°. This is also found to be concurrent with a modest increase in the frequency of occurrences of the sea breeze, which varied from 6 (pre-lockdown) to 9% (lockdown). The confinement of sea breeze to narrower spatial dimensions along with an increase in its frequency of occurrence indicates a potential perturbation of mesoscale dynamics triggered by the effects of lockdown on BC, which in turn might have perturbed heating profiles of the boundary layer (Yang et al. 2020).

The South Asian region in general and its megacities in particular experience strong BC emissions throughout the year. Therefore, assessing the impacts of regional emissions and coastal dynamics on BC concentrations has remained hindered by the lack of baseline measurements

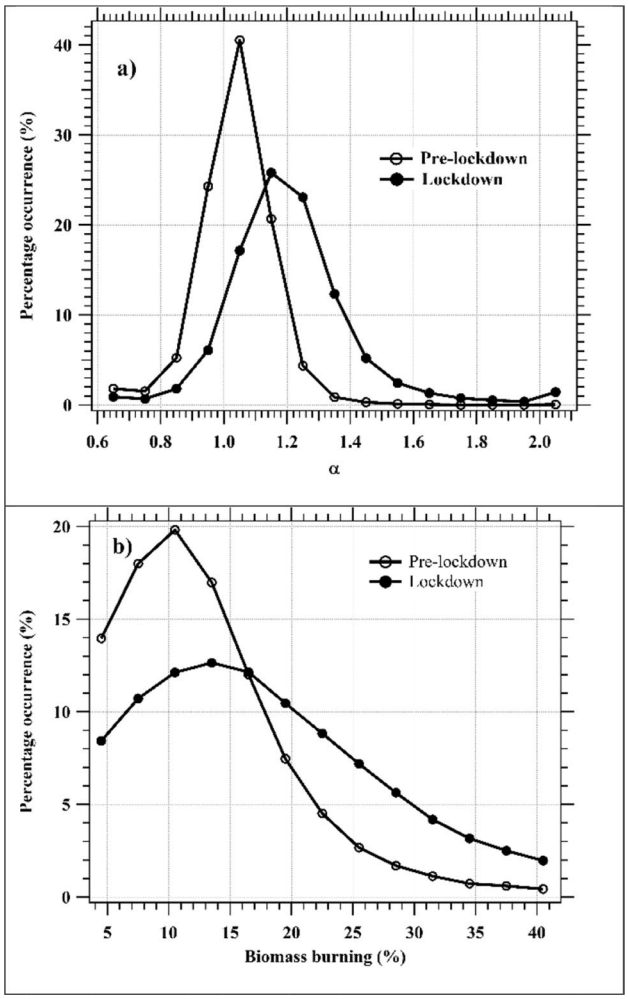


Fig. 4 Percentage occurrences of **a** spectral aerosol absorption index (α) and **b** biomass burning (%) during the pre-lockdown phase (open circle) and the lockdown phase (solid circle), respectively. Pre-lockdown and lockdown phases are defined in the "Methods" section

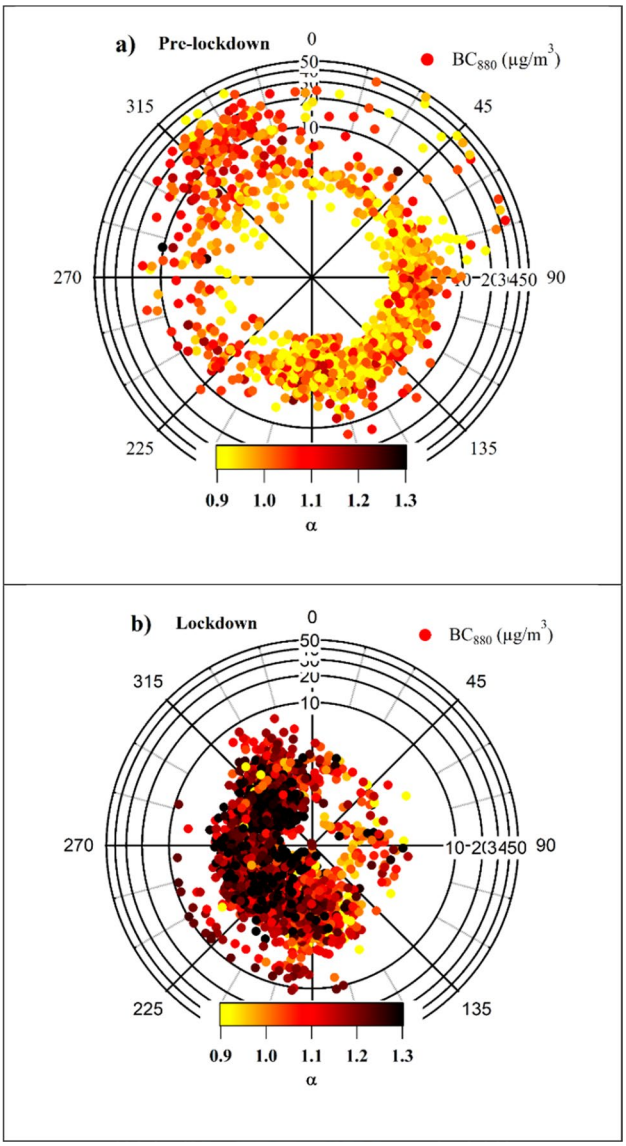


Fig. 5 Directional distribution of the hourly averaged BC mass concentration at the wavelength of 880 nm during **a** pre-lockdown phase and **b** lockdown phase. Black carbon (BC) mass concentrations are color-coded with the corresponding hourly averaged spectral aerosol absorption index (α). The color axis shows the range of α. Pre-lockdown and lockdown phases are defined in the "Methods" section

with minimal anthropogenic influences. This is especially critical since chemical-transport models also have stronger biases in reproducing BC concentrations over this part of the world. Further, studies using BC as an indicator of impact of COVID-19 lockdown impact remain limited. Such studies are of special importance because BC is a climate forcer. Estimating reductions in BC due to lower regional emissions is a key scientific input needed to mitigate climate warming. Nevertheless, most such assessments have relied on model simulations so far, and in this context, our study provides invaluable regional input in terms of BC concentrations in the marine and continental air mass. Our measurements over megacity Chennai at the coast of Bay of Bengal covering very rare episode of stringent COVID-19 lockdown allowed us to observationally evaluate the impacts of regional emissions and coastal meteorology on BC and aerosol spectral characteristics. BC dynamics in both the continental and in marine air mass was governed by the anthropogenic and biomass burning emissions in the tropical Indian region, in agreement with measurements of trace gasses and chemistry climate modeling (Girach et al. 2017; Lawrence and Lelieveld 2010). It is observed here that mesoscale dynamics have been perturbed during the lockdown period with a greater dominance of sea breeze laden air masses. The feedbacks of atmospheric composition change on meteorology during lockdown has also been reported in another recent study (Yang et al. 2020). In this regard, our observations and findings form an invaluable dataset covering a near-natural experiment with very low and very high BC concentrations. It is anticipated to be of specific value in evaluation of climate models while addressing the questions of wider scientific and public interests including potentially on rainfall trends (Ghosh et al. 2016) and intensifications of tropical cyclones (Evan et al. 2011).

Conclusions

Measurements of black carbon aerosol mass concentration was carried out over a megacity (Chennai, 12° 59′ 26.5″ N, 80° 13′ 51.8″ E), on the eastern coast of India, during January–June 2020, comprising the period of COVID-19-induced strict lockdown. The analysis focused on quantifying the impact of COVID-19-induced lockdown on aerosol absorption properties and the role of coastal meteorology. Following are the major highlights from this study:

- An unprecedented reduction in BC concentration was observed, from pre-lockdown to lockdown, in the continental sector, by an order of magnitude, and in the marine sector, by ~ 50%.
- Absorbing aerosols during the pre-lockdown appear to be dominated by local sources (fresh emissions) and that during the lockdown are dominated by aerosols from long-range transport (aged aerosols).
- BC source regime shifted from a fossil-fuel dominance to a biomass burning dominance as a result of lockdown, indicating cessation of fossil-fuel sources during the lockdown, whereas the biomass burning sources remained to their normal strength.

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Author contribution SSG and NO conceived the idea. AV, NO, and SSG conceptualized the study and designed the research. SSR, AS, and SS worked on the AE33 together and performed the long-term measurements. AV did the detailed analysis of the AE data with inputs from NO and SSG. AV, NO, and SSG performed the AE33 data interpretation. AV wrote the first draft of the manuscript with critical inputs from NO and SSG, with specific inputs from SKS and RR. All authors discussed the results and commented on the manuscript.

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Data availability All the data used in the manuscript including the code can be made available upon reasonable request with the corresponding author.

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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