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Aerosol Characteristics over the Indo-Gangetic Basin: Implications to Regional Climate

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1. Introduction

The climatic and environmental effects of atmospheric aerosols are the critical issues in global science community because aerosols, derived from variety of natural and man-made (or anthropogenic) emission sources, are well known to affect the air quality, human health and radiation budget [1]. While comparing the third and fourth assessment report of the Intergovernmental Panel on Climate Change (IPCC) as shown in Figure 1, the level of scientific understanding for the role of green house gases (GHGs) in projected temperature changes is higher relative to that of aerosols [2,3]. This is because of inadequate measurements of aerosols, their microphysical and optical properties and poor understanding of their role in the Earth's radiation budget.

Aerosols influence the Earth's climate directly by scattering and absorbing the solar and terrestrial radiations and indirectly by modifying the cloud macro- and micro-physical properties [4]. The direct and indirect effects of atmospheric aerosols are shown in the schematics in Figure 2a and 2b, respectively. Varity of aerosols present in the atmosphere from natural and anthropogenic emission sources can influence our Earth's atmosphere directly by absorbing/scattering the incoming solar radiations (Figure 2a). It can also absorb and re-radiated the outgoing radiations emitted from the Earth. On the other hand, aerosols indirectly affect the climate system by acting as cloud condensation nuclei (CCN) and ice nuclei (IN) and thereby modify the cloud properties and their impacts depending upon the environment like polluted or un-polluted regions (Figure 2b). In a recent study, reported in [5], they have investigated the indirect aerosol effect during the successive contrasting monsoon seasons over Indian subcontinent. However, a different study reported in [6] was carried out to investigate the intensity and the spatial extent of the indirect effect over the Indo-Gangetic Basin (IGB) region.



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Figure 1. A comparison between III and IV assessment reports of IPCC for 2001 and 2007 (*Adopted from* [2,3]).



Figure 2. Schematics showing (a) Direct and (b) Indirect impacts of atmospheric aerosols.

The uncertainty in quantifying these impacts have no doubt improved over the years due to assimilation of observations (especially after global observations of aerosols by EOS-Terra started in 1999), but not up to the desired level, particularly at regional scale [3]. The Indian subcontinent is one such region, where heterogeneity in aerosol optical and microphysical properties over a wide range of spatial and temporal scales continues to hinder in improving the estimates of aerosol-induced climate forcing. Thus, it is important to improve aerosol characterization with high spatio-temporal resolutions; particularly over the IGB region, which supports nearly 70% of the country's population and is one of the highly polluted regions in the world. The problem is also critical due to lack of adequate long-term measurements of aerosol distribution by General Circular Model (GCM) [7].

Although, aerosol properties have been measured at many sites in India in continuous and campaign modes (the *in-situ* observations have summarized in [8]) in the last two decades,

only few of them have fairly long-term data of aerosol microphysical properties [9-11]. Satellites are proved to be a good tool to understand the broad spatio-temporal characteristics of aerosols and associated effects from global to local scales [12-15]; but they are unable to provide an in-depth view of aerosol properties on a local scale and pose higher uncertainties as compared to the ground-based instruments [16]. In the IGB, aerosols of natural and anthropogenic origins mix with each other during dust loading season [17,18]. As a result, aerosol properties change, leading to even larger uncertainty in satellite retrievals [14,19] as this is not considered in the aerosol retrieval algorithm [20]. National Aeronautics and Space Agency (NASA) has setup ground-based aerosol monitoring network under the Aerosol Robotic Network (AERONET) program [21], in which automatic sun/sky radiometers are deployed at various places around the world. As per India, particularly in the northern part, is concerned, the routine measurements of aerosols under this network were started initially by the deployment of the sun/sky radiometer at Kanpur over the IGB region in year 2001 [22]. At a later stage, it was deployed at other places in the IGB, considering the region as crucial for aerosol measurements where significant aerosol loads of pollution mostly from the combustion of biomass, bio-fuel/fossil fuel emissions and the transported mineral dust have led to one of the largest regional TOA energy losses worldwide (For Example see [11,23-25]). The measurements by individual instruments can particularly be useful for the quantification of the regional impact of aerosols on the radiative energy balance. Provided a parallel individuation of aerosol types is possible, these local studies can also help to reduce the uncertainties on the effect of individual aerosol species, which is necessary because the direct radiative forcing by individual aerosol species is less certain than the total direct radiative forcing by all aerosols [3]. The complex mixture of aerosols over the IGB has been evaluated in the literature over the last decade starting with Indian Ocean Experiment (INDOEX) [26], and the research has been continued with the AERONET data [27 and references therein].

Understanding and quantifying the aerosol effects are important in the IGB region due to several pathways have been hypothesized to explain the possible impacts of aerosols on the regional hydrological cycle. The region is of great research interest due to its unique topography surrounded by the Himalayas to the north, moderate hills to the south, Thar Desert and Arabian Sea in the west, and Bay of Bengal to the east. The IGB region is dominated by the urban/industrial aerosols [28-30], which demonstrate significant seasonal variability based on the complex mixture of these aerosols with the naturally produced aerosols, particularly during the pre-monsoon and monsoon seasons. In addition to the urban-industrial pollution, desert dust is one of the other major natural sources of the aerosols over the Ganga basin [24,27,31-36], transported frequently from the neighboring desert regions, mostly during pre-monsoon periods. Dust storms are often experienced in northern and northwestern part of India, including over different parts of the IGB region during the pre-monsoon season, when dust aerosols are transported by southwesterly summer winds from the western Thar Desert [37,38]. High dust loading over the IGB region during the pre-monsoon period has been established by remote sensing data [39,40]. These dust storms apparently deposited silty materials in the downwind directions, as observed

on the quartzite ridges in the Delhi area [41]. The wind also carries heavy metals to the IGB during the summer season [42] along with the dusts, causing severe air pollution and degradation in the visibility. On contrary, the spatial distribution of aerosols (in terms of AOD) during the winter season also revealed high aerosol loading over the IGB and its outflow to the northern Bay of Bengal due to high anthropogenic emission sources, which was observed by satellite [8,12,13,15] and ground-based measurements [22,43-46].

2. Factors affecting aerosol characteristics over IGB

Complex nature of aerosols over the IGB is mainly because the region is very diverse in topography, population distribution, meteorology and emission sources. Figure 3 shows unique topography of the IGB region, surrounded by the variety of aerosol emission sources and thus making it hotspot for aerosol research.



Figure 3. IGB region, showing unique topography surrounded by the variety of aerosol emission sources (Adopted after modification from personal presentation of William K. M. Lau on Aerosol, Monsoon Rainfall Variability and Climate Change).

2.1. Topography

The IGB region, world's most populated river basin having more than 700 million populations, stretches from Pakistan in the west to Bangladesh in the east, encompassing most of the northern part of India. The region is bounded by the Himalayas to the north,

and by Vindhyan and Satpura range of mountains in the south. Due to its unique topography, this region can be summarized as a type of region, where, both anthropogenic and natural, aerosols show distinct seasonal characteristics and mixing [13,17,22,25,28,30,47]. General seasonal abundance shows that the winter months are dominated by the fine-mode aerosols, produced by various anthropogenic sources from the IGB region, and pre-monsoon or summer months are dominated by the coarse-mode mineral dust, primarily from the Thar Desert region in the western Rajasthan and its frequent transportation over the IGB region. Further details regarding geography, climate, regional sources and emissions of these aerosols over the IGB as well as over the other Indian region, however, can be found in [15]. This region also provides favorable climate for the agricultural activities due to its fertile soils and abundant water supply from the Southwest monsoon and the rivers originating from the Himalayan glaciers such as the Ganges. Consequently, the cultivable land forms a major fraction of the total geographical area in the IGP region (~76%) as compared to the rest of India (~50%) (http://dacnet.nic.in/).

2.2. Synoptic conditions and aerosol characteristics over IGB

Synoptic meteorology (e.g. wind pattern, air temperature and specific humidity) over the IGB region along with its surroundings is shown in Figure 4 for (a) winter and (b) summer seasons for the period of 2007-2008. The European Centre for Medium-Range Weather Forecasts (ECMWF) reanalysis monthly data of weather parameters such as wind, air temperature and specific humidity at 850 hPa pressure level were used to study the synoptic meteorological conditions over the region. In both the figures, winds are shown with arrows pointing towards the wind direction, where length of arrows defines the magnitude of wind speed (in ms⁻¹), line contour represents air temperature (in °C) and shaded color contour represents specific humidity (in kg kg-1), showing in dark blue color for low and red color for high magnitude of specific humidity. Results reveal that the IGB region during the winter period is relatively drier than during the summer. The persistence of low temperature and the westerlies (with low intensity) can be seen over the region during the winter whereas during the summer, relatively high temperature with intense southwesterly winds was observed to dominate. These winds are found to pass through arid regions of the western India (particularly from the Thar Desert) and bring dry air masses over the region [24,27,48].

The general aerosol characteristics over the entire IGB region are shown in Figure 5 as mean AOD values at 550 nm for (a) winter and (b) summer seasons for the period of 2007-2008 in color codes, obtained from the Moderate Resolution Imaging Spectroradiometer (MODIS). Large spatial heterogeneity in AOD can readily be noticed over the IGB region during both winter and summer periods, which has also been confirmed through various ground-based measurements, discussed in the later section of this chapter. Relatively large magnitude of AOD was observed throughout the IGB region during the summer, which is mainly due to frequent occurrence of dust storms over the Thar Desert region that caused large amount of dust particles to be transported over the station (showing the highest AOD). However, large

AOD during the winter is confined mostly over the eastern part of IGB. During the winter, the IGB region is often enveloped by thick fog and haze [49]. The prevailing winds over the region are westerly to northwesterly with relatively low wind speeds (<5 ms-1) as compared to the summer (as can be seen in Figure 4a) and the eastern parts of the IGB are impacted by a localized area of strong subsidence in winter [8,12,13,50]. These conditions tend to trap the pollution at low altitudes and responsible for the higher AOD along the eastern part of IGB. Results obtained, although, include the impacts of aerosol emissions from various natural and anthropogenic sources and the prevailing meteorology over the region, it also encourages to further investigate the plausible causes and impacts over the radiation budget as well as on weather and climate.



Figure 4. Synoptic meteorological conditions over the entire IGB region derived from ECMWF at 850 hPa pressure level during (a) winter and (b) summer periods (*Adopted from* [46]).



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Figure 5. General aerosol characteristics in terms of AOD (550 nm) over the entire IGB region derived from MODIS during (a) winter and (b) summer periods (*Adopted from* [46]).

2.3. Emission sources

The IGB region, apart from being a major source region for aerosols, is bordered by densely populated and industrialized areas on the west and eastern sides from where different aerosol species such as mineral dust, soot, nitrate, sulfate particles and organics are produced and transported to this region and thus making it an aerosol hotspot, as can also be seen in Figure 3. The region itself has both, rural and urban population and various kinds of emission sources such as natural and industrial. In rural areas, bio-fuels burning such as wood, dung cake and crop waste, predominantly contribute to the major aerosols loading [51]. However, in urban areas, aerosol emissions from fossil fuels burning such as coal,

petrol and diesel oil dominate [52,53]. Large fluxes of absorbing aerosol emissions (black carbon and inorganic oxidized matter, which is mostly fly ash from coal-based power plants and particles from open burning of crop waste/forest-fires) were reported over the IGB [51]. Apart from the dust emissions from the Thar Desert, predominantly during the premonsoon months, the influence of emissions from the forest-fires and open burning of crop waste from the central India were also found over IGB during these months as biomass aerosol contribution [15,27].

3. Measurements

3.1. Ground-based

Aerosol measurements in the Indian sub-continent started as early as the 1960s, when [54] studied Angstrom turbidity from solar radiance measurements. Later, a multi-wavelength radiometer was developed by the Indian Space Research Organization (ISRO) to monitor spectral AOD at Trivandrum in the year 1985 [55] and in the same year, aerosol vertical distribution measurement by ground-based lidar was initiated at Pune [56]. Further, NASA has setup ground-based aerosol monitoring network in India under the Aerosol Robotic Network (AERONET) program [21], in which automatic sun/sky radiometers are deployed at various places, particularly in the northern part of India, including the Himalayan foothills. The routine measurements of aerosols under this network were started initially by the deployment of the automatic sun/sky radiometer at Kanpur over the IGB region in year 2001 [22]. At a later stage, it was deployed at other places in the IGB, considering the region as crucial for aerosol measurements [36].

Using ground-based radiometric measurements, [22] have reported for the first time the seasonal characteristics of aerosol optical properties and the spectral behavior of AODs over Kanpur, an urban-industrial city, situated in the central part of the IGB. They showed pronounced seasonal influence of various aerosol properties, with maximum dust loading during the pre-monsoon season. The increase of pollution has a direct impact on climatic conditions, especially the increase of haze, fog, and cloudy conditions, which decrease the visibility especially during the winter season. On the other hand, in-situ aerosol measurements by Central Pollution Control Board (CPCB) have also showed very high annual average concentrations (>210 μ g/m³, in the critical range compared to the air quality standard in India) of particulate matter of diameter less than 10 µm (PM₁₀) in the atmosphere of the major cities of the Ganga basin like Delhi, Kanpur and Kolkata (http://www.cpcb.nic.in). These high PM10 concentrations provide an opportunity for SO4 formation on the particulate surface, leading to very high concentration of sulfate aerosols in the atmosphere, which is the case observed over the IGB and reported in [29]. Several studies indicate strong seasonal variability in aerosol loading and changes in aerosol properties over the IGB [14,22,25,31,33-36,48,57,58]. In the recent studies, [23] and [24] have demonstrated the distribution of aerosols and associated optical and radiative properties in the IGB region and its further expansion to the foothills of Himalayas during the premonsoon period. The pre-monsoon period is of particular interest because this is the key period when locally generated and regionally transported aerosol loading peaks over the IGB region and spread up to the foothills of Himalayas [23,35,59-62], which has been linked to influence the monsoon circulation in India [63,64]. Significant gradient in the magnitude of most of the aerosol characteristics was observed over the IGB, which may be due to the gradual changes in weather parameters and/or emission sources. Such gradient is, ultimately, found to impact the Earth-atmosphere system by negative radiative forcing, thus causing cooling, at the surface, and positive aerosol forcing, thus causing heating in the atmosphere for the study period. Such gradient in heating rate raises several climatic issues, and is needed to be answered on the basis of longer period investigations at several stations to improve the scientific understanding of the regional climate in inter-annual as well as intra-seasonal scale.

The first simultaneous measurements of chemical composition (carbonaceous and inorganic species) and optical properties (absorption coefficient and mass absorption efficiency) of ambient aerosols (PM2.5 and PM10) have been recently reported in [58] at an urban site (Kanpur) in the IGB region. The study provides important information on the temporal variability in the abundance of organic matter and mineral dust over the IGB region, which has large implications to the large temporal variability in the atmospheric radiative forcing due to these aerosols. Based on the measured aerosol chemical composition, other studies have been carried out to understand the characteristics of anthropogenic aerosols and their quantification to the total radiative forcing over the IGB region, which are limited only at Kanpur [11] and Delhi [25]. Figure 6 shows seasonal variability of optical properties of composite aerosols estimated over Delhi (a typical urban station at the western part of the IGB near to the Thar Desert) during the winter, summer and post-monsoon seasons; however, the same for anthropogenic aerosols are shown in Figure 7. The anthropogenic components were found to be contributing ~72% to the composite aerosol optical depth (AOD_{0.5} ~0.84) at Delhi. The contribution was found to be more during the winter (~84%) and post-monsoon (~78%) periods and less during the summer (~58%). On the other hand, mean SSA for composite aerosols was found to be ~0.70 (ranging from 0.63 to 0.79). However, SSA for anthropogenic aerosols was found to be slightly less (by $\sim 1\%$) than that for composite aerosols, and the difference may be due to the mixing of natural dusts with anthropogenic aerosols in the region (for composite aerosols).

The resultant atmospheric forcing due to composite and anthropogenic aerosols at Delhi is shown in Figure 8. The anthropogenic contributions to the composite aerosol were found to be ~93%, 54%, and 88%, respectively during the winter, summer, and post-monsoon seasons (with a mean contribution of ~75%). However, the anthropogenic fraction of ~73% is responsible for the composite aerosol atmospheric heating rate (2.42 ± 0.72 Kday⁻¹) at Delhi. On the other hand at Kanpur, another typical urban station at central part of the IGB region, the heating rate due to anthropogenic aerosols was reported to be ~65% to the heating due to composite aerosols [11]. Relatively higher heating rate at Delhi may be caused by the large contribution of transported mineral dust aerosols due to the proximity of the station to the Thar Desert region as compare to Kanpur and their probable mixing with the other absorbing aerosol species like black carbon (BC) [27].



Figure 6. Seasonal mean spectral variation of (a) AOD and (b) SSA for composite aerosols over Delhi (*Adopted from* [25]).



Figure 7. Same as figure 6, except for anthropogenic aerosols (Adopted from [25]).

Large atmospheric heating rate of the order of more than 2 Kday⁻¹ is quite significant. Moreover, the large surface cooling due to negative forcing at the surface and strong heating due to positive forcing in the atmosphere, particularly for the anthropogenic aerosols, can strongly affect the atmospheric dynamics over the region. The warmer atmosphere close to the surface (due to high atmospheric absorption) and the colder surface during winter and post-monsoon periods over Delhi would create low-level inversions and strengthen the boundary layer stability [11], which restrict the mixing and dispersion of aerosols into the atmosphere. On the other hand, during summer, the observed large heating in the atmosphere, which is probably due to the mixing of anthropogenic aerosols with abundance of natural dusts, may supply excess energy to be trapped in the atmosphere during dry season and can have significant impact on regional climate and monsoon circulation systems [65,66]. Anomalous atmospheric heating due to absorbing aerosols (dust and BC) over the northern part of India during pre-monsoon season has been reported in [63]. A comparative study of aerosol direct radiative forcing was made with the available estimates from the literatures at various regions and given in Table 1. Various regions, characterized by different kinds of aerosol sources and prevailing meteorological conditions, are associated with different values of aerosol forcing and have provided an understanding of the aerosol radiative effect on regional scales, which are significantly different from the global mean radiative effect (indicating slightly cooling of the atmosphere).



Figure 8. Monthly variation of atmospheric forcing for composite and anthropogenic aerosols over Delhi. The corresponding heating rate values for respective aerosols are given in the parenthesis (*Adopted from* [25]).

Location	Type of Location	Period	Aerosol DRF (Wm ⁻²) at		Deferrer
			Surface	Atmosphere	References
New Delhi	Urban	Annual	-67	71	[57]
Kanpur	Urban	Annual	-32	28	[11]
Ahmedabad	Urban	Annual	-49	44	[131]
New Delhi	Urban	Jan-Nov 2007	-79	87	[25]

Location	Type of Location	Period	Aerosol DRF (Wm ⁻²) at		D (
			Surface	Atmosphere	References
New Delhi	Urban	Mar-Jun 2006	-77	80	[48]
Kanpur	Urban	Apr-Jun 2009	-26.1 to -29.2	19.5 to 16.1	[24]
Gandhi College	Rural	Apr-Jun 2009	-29.7 to -31.9	20.9 to 16.6	[24]
New Delhi	Urban	Winter (Dec 2004)	-66	67	[132]
Hissar	Urban	Winter (Dec 2004)	-10 (before fog) -20 to -25 (during fog)	15 (before fog) 25 to 40 (during fog)	[128]
Pune	Urban	Nov-Apr 2001 and 2002	-33	33	[133]
Hyderabad	Urban	Jan-May 2003	-33	42	[134]
Bangalore	Urban	Oct-Dec 2001	-23	28	[135]
Central India (multiple stations)	Urban and Rural	Winter (Feb 2004)	-15 to -40	16 to 29	[72]
Nainital	Rural (high- altitude)	Winter (Dec 2004)	-4.2	0.7	[77]
Nainital	Rural (high- altitude)	July 2006-May 2007	-14	14	[62]
Kathmandu	Urban (high- altitude)	Winter 2003	-25	25	[136]
Chennai	Urban	Feb-Mar	-19	13	[137]
Arabian Sea	Polluted Marine	Mar-Apr	-27	15	[70]
Bay of Bengal	Polluted Marine	Mar	-27	23	[138]
Indian Ocean	Polluted Urban	Feb-Mar	-29	19	[139]
Global mean	Natural and Anthropogenic			-0.5±0.4	[3]

Table 1. Aerosol direct radiative forcing (DRF) at different locations in India.

Due to high level of anthropogenic emissions, aerosol distribution in terms of type and loading undergo strong variability associated with the episodic yet strong influence of dust transport and biomass burning during the pre-monsoon period [23]. Dust was found to be one of the major components of aerosol composition (apart from other species) over the region [32], which significantly affects the region during pre-monsoon period due to enhanced surface convection activities [24,25,31,33-36,48,67], and thus essential to quantify its contribution over the region. Aerosol composition was measured with the chemical analysis method over IGB during different periods of time as reported in [44,52,53]. Retrieval of columnar black carbon and organic carbon has been carried out over IGB using the AERONET data [60,68]. Moreover, [7] have integrated AERONET and Cloud-Aerosol

Lidar with Orthogonal Polarization (CALIOP) data into atmospheric GCM to infer aerosol types at two AERONET sites in the IGB. However, in a recent study, [27] have discriminated the major aerosol types over the IGB region during pre-monsoon period using multi-year AERONET measured aerosol products associated with the size of aerosols (mainly fine mode fraction, FMF) and radiation absorptivity (mainly single scattering albedo, SSA). Figure 9a shows density plot of SSA versus FMF at Kanpur (KNP, a typical urban AERONET site over the central IGB region) and Gandhi College (GC, a typical rural AERONET site over the central IGB region) for different aerosol types. High dust enriched aerosols (i.e. polluted dust, PD) were found to contribute more over the central IGB station at Kanpur (~62%) as compared to the eastern IGB station at Gandhi College (~31%) whereas vice-versa was observed for polluted continental (PC) aerosols, which contain high anthropogenic and less dust aerosols. Contributions of carbonaceous particles having high absorbing (mostly black carbon, MBC) and low absorbing (mostly organic carbon, MOC) aerosols were found to be 11% and 10%, respectively at Gandhi College, which was ~46% and 62% higher than the observed contributions at Kanpur; however, very less contribution of non-absorbing (NA) aerosols was observed only at Gandhi College (2%). The mean SSA and FMF based on cluster analysis of daily-averaged data at Kanpur and Gandhi College, associated with the different aerosol categories is also shown in Figure 9b. The horizontal and vertical lines indicate the standard deviations of SSA and FMF from their respective means, indicating the variability of these parameters for different aerosol types. Although similar magnitude of SSA was observed for PD, PC and MBC type aerosols, they are further distinguished based on FMF thresholds following Lee et al. (2010), i.e. FMF<0.4 indicates dominantly coarse-mode and hence is assigned to PD aerosols, FMF>0.6 indicates dominantly fine-mode and hence is assigned to MBC aerosols, and PC aerosols are considered for $0.4 \le FMF \le 0.6$. MOC and NA type aerosols have similar FMF, but higher scattering relative to the other aerosol types.



Figure 9. (a) Density plot and corresponding (b) cluster plot of AERONET-derived SSA vs. FMF for two stations over the IGB region (showing different aerosol types) during pre-monsoon period (*Adopted from* [27]).

Spectral information of SSA for each aerosol type was also shown and discussed in [27], which clearly discriminates the dominance of natural dust (SSA increases with increasing wavelength) with anthropogenic aerosols (SSA decreases with increasing wavelength) at Kanpur and Gandhi College over the IGB. As expected, SSA for PD and PC aerosols was found to have spectrally increased, suggesting relative importance of dust. PD has higher spectral trend relative to PC due to larger fraction of dust, which was found to be dominated at Kanpur as compared to Gandhi College. On the contrary to PD and PC type aerosols at Gandhi College, SSA for NA aerosols was found to have spectrally decreased with relatively larger magnitude at all the wavelengths. However, relatively less spectral dependence in SSA was seen for MBC and MOC aerosols, which shows slight decrease in spectral SSA at Gandhi College and opposite at Kanpur.

Further, the absorption aerosol optical depth (AOD_{*abs*}) at different wavelengths (λ) can be obtained, suggested in [69] as

$$AOD_{abs}(\lambda) = \left[1 - SSA(\lambda)\right] \times AOD(\lambda)$$
(1)

The absorption Ångström exponent (AAE) has been computed as negative of the slope of fitted line of the natural logarithm of AODabs vs. natural logarithm of the respective wavelengths and used to substantiate the inferred aerosol types over IGB, as shown in Figure 10. The magnitude of AAE near to 1.0 (marked by dotted line in Figure 10) represents a theoretical AAE value for black carbon as reported in [69]. AAE values for PD and PC aerosol types were found to be 1.70 and 1.43, respectively at Kanpur and 1.30 and 1.18, respectively at Gandhi College. However, for MBC and MOC type aerosols, AAE values were relatively higher at Kanpur (~20%) than at Gandhi College, where values were found to be closer to the theoretical AAE value for black carbon (i.e. AAE≈1.0), thus indicating the presence of fresh BC at Gandhi College, which can be expected from the potential source of combustion of fossil fuel and biomass burning used for domestic purposes. On the other hand, aged BC or mixed BC can be expected at Kanpur (mostly from biomass burning and urban/industrial sources), which is favorable scenario during the summer periods [17-36]. The estimated AAE values over the IGB thus suggest relative dominance of absorbing type aerosols over the central part of IGB (due to dominant dust mixed with other absorbing aerosols) as compared to the eastern part during pre-monsoon period.

Apart from these continuous measurements, various field campaigns have also been conducted regionally to study and improve the aerosol remote sensing measurements as well as provide data for atmospheric prediction over the past decade. Campaigns conducted in or near India, which used space-based, airborne, and surface-based instrumentations to observe high aerosol loading over the Indian subcontinent and the surrounding Oceanic regions, included the Indian Ocean Experiment (INDOEX) reported in [26], Arabian Sea Monsoon Experiment (ARMEX) reported in [70], Indian Space Research Organization Geosphere Biosphere Programme (ISRO-GBP) Land Campaign reported in [43-45]. The first phase of Land Campaign (LC-I) was conducted during February to March 2004, to understand the spatial distribution of aerosols and trace gases

over central/peninsular India. The details of these campaigns and the major findings have been reported in literatures in [71-74]. As a continuation of this experiment, second phase of land campaign (LC-II) was conducted during December 2004, to characterize the regional aerosol properties and trace gases across the entire IGB region including the Himalayan foothills. This phase of the campaign provided a comprehensive database on the optical, microphysical and chemical properties of aerosols over the IGB and the foothills of Himalayas and reported in [44,45,75-80]. All these studies showed the persistence of high aerosol loading (in terms of high AOD) and black carbon mass concentrations over the region.



Figure 10. Mean AAE values for each aerosol type at Kanpur and Gandhi College over the IGB (*Adopted from* [27]).

Further, an International TIGERZ experiment was conducted by the NASA AERONET group within the IGB region around the industrial city of Kanpur during the pre-monsoon period [36]. The major objectives of TIGERZ include the spatial and temporal characterization of columnar aerosol optical, microphysical and absorption properties; the identification of aerosol particle types/mixtures; and the validation of remotely sensed aerosol properties from satellites. In a recent past, Ramanathan group from the Scripps Institution of Oceanography, University of California, USA conducted a field measurement (from November 2009–September 2010), called *Project Surya*, in a rural area over the IGB region. Studies were focused on to establish the role of both solid biomass based cooking in traditional stoves and diesel vehicles in contributing to high BC and organic carbon (OC),

and solar absorption [81,82]. In continuation to this, [83] have studied the link between local scale aerosol properties and column averaged regional aerosol optical properties and atmospheric radiative forcing.

Apart from ground-based aerosol measurements, vertical distribution of aerosols were carried out for the first time over Kanpur in the IGB region during the winter and summer of the year 2005, reported in [84-85]. Vertical measurements of aerosols up to 1.5 km provided useful information during the winter because aerosols were mostly confined to the boundary layer; however, during summer, aerosols get convected and reached up to the higher altitudes. The Integrated Campaign for Aerosols, gases, and Radiation Budget (ICARB) was initiated to address these issues with multi-institutional, multi-instrumental, multi-platform field campaign, where integrated observations and measurements of aerosols with special emphasis on black carbon, radiation and trace gases along with other complementary measurements on boundary layers and meteorological parameters were made simultaneously [86]. The ICARB was conducted during February-May period of 2006 as an integrated campaign, comprising three segments namely the land, ocean and air, to assess the regional radiative impact of aerosols and trace gases, and to quantify the effect of the long-range transport of aerosols and trace gases over the Indian mainland, Arabian Sea, Bay of Bengal and the tropical Indian Ocean. The details of this campaign and the major findings have been reported in different literatures in [86-90]. ICARB was covered only the eastern part of the IGB (Bhubaneswar) [91] while focusing mostly on the peninsular India and surrounding oceans. Continental Tropical Convergence Zone (CTCZ) campaign, focused on the aerosol distribution in the pre-monsoon and monsoon (June-September) seasons was initiated in the year 2008, and covering the continental part of the more common tropical convergence zone over India, including the IGB [92]. During the campaign, Aircraft and ground-based measurements together were carried out over the IGB and Central part of India to quantify the aerosol indirect effect. The details of the campaign and the major findings have been reported in a recent publication in [61].

Even though all these national and international field experiments and campaigns have greatly improved our understanding on aerosol optical, physical as well as chemical properties and have indeed reduced the uncertainty in regional aerosol direct radiative forcing at various parts of India including the IGB region, they are limited to a certain period or location due to their specific goals. In this perspective, long-term experiments with a high spatio-temporal scale can add advantages of understanding aerosol influences on a longer time scale, thereby helping to infer the signs of anthropogenic impacts. This is where satellite data become very useful and can complement the ground-based and/or *in-situ* measurements.

3.2. Space-borne

Satellite retrievals of aerosol properties over land have only been available in recent years and a few studies have been done using these data over the Indian subcontinent, focusing on the IGB region. In reference [12], they were the first to study the spatial distribution of AOD over India using Multiangle Imaging Spectroradiometer (MISR) during the winter period from 2001 to 2004, where they were able to explain the enormous pollution observed over the IGB based on meteorology, topography and potential aerosol emission sources. Further, subsequent studies using Moderate Resolution Imaging Spectroradiometer (MODIS) data have confirmed this observation [13,15,93] with additional information on the seasonal variability of AOD and fine mode fraction, to some extent. In continuation to that recently in reference [8], they have presented a detailed analysis of a 9 year (2000–2008) seasonal climatology of size- and shape-segregated aerosol properties over the Indian subcontinent derived from the MISR. The spatial heterogeneity of the aerosol parameters are shown in Figure 11 for each season.

The spatial distribution of AOD in the winter season reveals high AOD over the IGB and its outflow to the northern Bay of Bengal because of high anthropogenic emission sources, as previously observed from satellite [12,13,15,93] and ground-based [22,43,45] measurements. It is well known that the IGB region is often enveloped by thick fog/haze during this period, which is typically associated with high aerosol loading over the region [43,49,94-96]. AOD averaged over eight winter seasons is highest (>0.4) over the eastern part of IGB, which was referred to as the 'Bihar pollution pool' in [12]. As pointed out in [12], this observation has strong implications for the large population residing in this area and thus calls for further work. In [97], they have used CO retrieved from MOPITT (version 3 data) and found a corresponding pool of high CO mixing ratios at 850 hPa level in the same area in winter. In continuation of this, in [50], they have further demonstrated the extensive pollution along the eastern parts of the IGB during winter months using the improved version 4 CO data from MOPITT and the new version 3 height resolved aerosol data from CALIPSO as well as the tropospheric column ozone from two different data products. Both the CO and aerosol data from this study confirm the trapping of pollution at low altitudes by subsidence. Aerosols across the IGB was found to be transported from west to east by northwesterly winds, encounter a narrowing valley floor and are trapped efficiently within the atmospheric column in the eastern part of the IGB by subsiding air [8]. Relatively low AE (<0.8) in the eastern IGB than the other parts, suggests high concentration of coarse dust particles emitted possibly by rural activities (e.g., agriculture, etc.) from the densely populated rural population.





Figure 11. Spatial distribution of climatological mean mid-visible AOD (first panel) and AE (second panel) during the winter, pre-monsoon, monsoon and post-monsoon seasons over the Indian subcontinent (including IGB region) for the period from March 2000 to November 2008 (*Adopted from* [8]).

In the pre-monsoon season, aerosol spectral optical properties change significantly from the preceding winter season because of enhancement in dust loading, particularly over the IGB region [15,31,24]. Large emission of small particles from open biomass burning compensates the relative influence of dust on spectral AOD in the eastern part of the IGB [27] as indicated by an increase of AE during pre-monsoon season compared to the preceding winter season (Figure 11). This also leads to an overall increase in AOD in this region compared to the winter season. Thus, winter to pre-monsoon changes in aerosol properties are not just dominated by an increase in dust, as previously thought, but also by an increase in anthropogenic components, particularly in the regions where biomass combustion is in the common practice during this period. Changing atmospheric aerosol properties caused by anthropogenic activities carries serious implications for climate change and human health [98].

The anthropogenic emissions, particularly BC and sulfate aerosols are present throughout the year in northern India over the IGB [99,100]. Such aerosols form thick layers of haze in winter, termed as Atmospheric Brown Clouds (ABC), which block the solar radiation reaching to the surface [101]. In [102], they have reported in their study over India that the AODs derived from TOMS data from 1979 to 2000 increased by ~11% per decade during the winter with large values over the IGB region, which consequently affect the surface reaching solar radiation, known as "solar dimming" [103-105]. The average solar dimming observed over India is about -0.86 W m⁻² yr⁻¹, while during winter, pre-monsoon and monsoon seasons the same was observed to be about -0.94, -1.04 and -0.74 Wm⁻², respectively [104]. The significant reduction in ground-reaching solar radiation can directly be correlated with the increased aerosol loading in the atmosphere due to enhancement in industrialization, vehicular pollution, biomass burning and dust storm activities over the region [106,107]. Apart from solar dimming effect due to variety of aerosols in terms of haze/fog conditions, our understanding about the role of secondary organic aerosols (particulate organic matters produced by gas-to-particle conversion process), particularly in climate change and its connection to health effects is very limited by numerous uncertainties. In a recent study in [108], they have observed an enhanced production of secondary organic aerosols over Kanpur in the IGB during the foggy periods of winter, which was hypothesized that the aqueous phase chemistry in fog drops is responsible for increased production of secondary organic aerosols.

During the monsoon, stronger westerly winds were found to be transport greater components of dust from the Arabian Peninsula to the Indian subcontinent [8]. In general, the spatial distribution of AOD (Figure 11) in this season is largely influenced by monsoon precipitation. Suppressed precipitation in the monsoon break phase allows for a rapid buildup of aerosols in the high anthropogenic source regions (e.g. IGB), while particles are being washed out by the precipitation in the active monsoon phase. This also leads to very high intra-seasonal and inter-annual variability in aerosol characteristics. Aerosol regional mean climatology in the post-monsoon season is very similar to that for the winter season (Figure 11), but the spatial distribution differs in several regions. For example, the wintertime high AOD zone in the IGB shows a larger spread and higher inter-annual variability across the basin in this season, owing to a stronger peak in crop waste burning in the western part of IGB than the eastern part [109] and weaker subsidence in the eastern part of IGB compared to the winter season. As a result, IGB is the region with highest aerosol absorption and thus occurred large discrepancy in MISR and MODIS derived AODs [20].

4. Coupling of IGB aerosols to the Himalayan region and their possible impacts

Due to combined effects of IGB topography and the Himalayan orography, aerosols over the IGB region are lifted up quite often and found to be extended up to the Himalayan foothills and also to the other high-altitude regions [23,61,110-113]. Absorbing aerosols in the elevated regions heat the mid-troposphere by absorbing solar radiation, and produce an atmospheric dynamical feedback called as elevated heat pump (EHP) effect. Consequently, this can lead to an increase in the summer monsoon rainfall over India [63] and enhancement in the rate of snow melting in the Himalayan regions [64], which is one of the potential themes for global scientific community and need to be addressed to improve scientific understanding of the regional climate on inter-annual as well as intra-seasonal scales. In particular, the main emphasis of the IGB region coupled with the Himalayan foothills is due to the highest AOD values in this region among the South Asia regions, which are persistent throughout the winter and spring seasons [114].

In a recent study in [115], they have shown a possible influence of desert dust aerosols originated and transported from the Thar Desert region to the high-altitude station at Manora Peak, Nainital in the central Himalayas (Figure 12). The high values of aerosol index (AI) derived from the Ozone Monitoring Instrument (OMI) attest to the presence of absorbing aerosol particles over the region; however, air mass back-trajectory analysis over the station shows different pathways for the transport of air masses from the source region to the experimental site over different time periods (Figure 12). In this study, [115] observed a thick aerosol layer at ~1500 m altitude (Figure 13), above the station level, which was substantiated by the air mass back-trajectory analysis (Figure 14).





Figure 12. OMI AI images showing the source and the progressive movement of the dust aerosols occurred in June 2006. Five-day air mass back-trajectories at Manora Peak for different time periods are superimposed on respective days AI images (*Adopted from* [115]).



Figure 13. Altitude versus time variations in BSR on a dust day (12 June 2006, with color contour) along with the variations in lidar-derived AOD at 532 nm (with solid line, average AOD = 0.83 ± 0.12) (*Adopted from* [115]).

Apart from dust transport from the Desert regions, recent study in [116], they have also demonstrated significant impact of north Indian biomass burning on aerosols and trace gases and the resultant radiation budget over the central Himalayas during the spring period through air mass back-trajectory analysis coupled with fire counts (Figure 15). The same has also been reported in [117] to be one of the major sources of BC over the same station in the central Himalayas, which was observed to be much lower (in terms of

magnitude) as compared to the urban location in the IGB, but was found to have significant contribution to the total aerosol optical depth (~17%) and the resultant atmospheric forcing (~70%) at Manora Peak [62]. Based on BC measured at two different wavelengths at ultraviolet (370nm) and near-infrared (880nm), [117] have distinguished the potential sources of BC at Delhi (one of the densely populated and industrialized urban megacities in Asia and typically represents the plains of Ganga basin) and Manora Peak (one of the high-altitude and sparsely inhabited clean site in the Indian Himalayan foothills situated in the central Himalayas). Based on the analysis, [117] have found the major contribution of BC at Manora Peak is from biomass burning while fossil fuel is found to be the dominating contributor at Delhi.



Figure 14. Temporal evolution of air masses at 1500 m altitude for three different time intervals on 12 and 13 June 2006 (*Adopted from* [115]).



Figure 15. Three day back-air trajectories arriving at Manora Peak during the fire-impacted periods in 2007, 2008 and 2009 (triangle represents the observation site) (*Adopted from* [116]).

5. Summary and future directions

The study over IGB region revealed different aerosol characteristics over the region from western to central and to the eastern parts, which show significant gradient in magnitude of most of the aerosol characteristics. Such gradient can be explained due to the gradual changes in weather parameters and/or emission sources apart from geographical heterogeneity. Such gradient is, ultimately, found to have impact on the Earth-atmosphere system by negative radiative forcing, thus causing cooling, at the surface, and positive aerosol forcing, thus causing heating in the atmosphere. Such gradient in heating rate raises several climatic issues, and is needed to be answered on the basis of longer period investigations at several stations to improve the scientific understanding of the regional climate in inter-annual as well as intra-seasonal scale.

Due to large uncertainty in satellite derived aerosol products over the IGB during premonsoon dust periods, long-term ground-based measurements during different seasons can indeed provide useful information of the characteristics of aerosol types over the region on seasonal and inter-annual basis, which are meager and crucial for the regional climate models. Further, the mixing of natural dust with anthropogenically produced aerosol particles, has been hypothesized in [17] over the IGB region, mostly during the pre-monsoon period and corroborated with the AERONET data [36], suggested the complication of the satellite retrieval of aerosol characteristics and quantifying the climatic effects [118]. Hence, it is also one of the important research areas in understanding aerosol characteristics over the IGB region to make realistic assessments of aerosol-hydro-climate interplay.

The issue of black carbon or soot particles and its relationship with climate change has gained enormous scientific and popular interest over the last few years. The knowledge and understanding on aspects such as vertical distribution and mixing of black carbon with other aerosols, effects of cloud cover and monsoon still remains uncertain and incomplete. Few studies have shown that when sulphate or organics is coated over black carbon aerosols, its absorption effects are enhanced by 50% [119]. In case of black carbon mixed with large dust particles, absorption of the composite dust-black carbon aerosol system is enhanced by a factor of two to three compared to sum of black carbon and dust absorption [120]. However, we have no information on the state of mixing of black carbon. The proper assessment of mixing and/or coating of various aerosol species and their impacts on various aerosol characteristics have not been well quantified [121], which makes the investigation a real challenge [122]. IGB, being in proximity to the Thar Desert region, is found to be affected predominantly by the enhanced dust aerosols, mostly during the pre-monsoon period. As a result, the probability of this interaction (i.e. mixing) was suggested to be more over the region during this period [17,36] and is one of the future perspectives. To better understand these crucial issues, National Carbonaceous Aerosol Program (NCAP) was recently launched in India, focusing on the measurement of black carbon; their role in atmospheric stability and the consequent effect on cloud formation, monsoon and retreating of Himalayan glacier [123].

Based on recent observations using aircraft [61] and satellite measurements ([34,46], it has been reported that during pre-monsoon season, IGB region is characterized by the elevated aerosol layers extended up to the altitude from about 3 to 5 km. When the amount of

absorbing aerosols such as black carbon and dust, are significant in the atmosphere, the aerosol optical depth and chemical composition are not the only determinants of aerosol radiative effects, but the altitude of the aerosol layer and its altitude relative to clouds (if present) are also essential. Thus, it is also essential to gather information on vertical distribution of aerosols over this region.

Further, fog over the IGB region is observed to be a common feature, occurs mostly during the winter period. The number of foggy days has been increasing in recent years as compared to earlier decades [124], with strong increasing trends of anthropogenic pollution in the IG plains [125]. Fog formation usually begins in the latter half of December and continues till the end of January, thus blanketing some regions for more than a month [126]. The low topography of the IGB, adjacent to the Himalayan range, favors formation of fog and provides high concentration of air pollutants in the plains which serve as additional CCN for nucleation. Fog affects day to day lives of millions of people living in this region, resulting in poor visibility down to less than 100 meters causing frequent flight and train delays and even a significant number of deaths from vehicular accidents in many severe events [127]. Though few studies were done focusing on fog-induced aerosol characteristics over the IGB region and their impacts to the aerosol radiative forcing [49,128], detailed studies of aerosol composition and inter-annual variation of aerosols are required to better understand the interaction of winter haze with the formation of fog over the IGB.

Apart from the measurements for various aerosol characteristics through different groundbased and space-born instrumentations, a 1-D aerosol optical model named as optical properties of aerosols and clouds (OPAC) has been developed by [129], estimating crucial optical properties of aerosols such as AOD and SSA, under the assumption of spherical aerosol particles and external mixing. In [130], they have shown that the optical depth and SSA of aerosol particles have strongest sensitivity on the direct radiative forcing, and these optical properties have found to be large deviation with shape and composition [18]. Further, with the model studies reported in [47,118], the particle composition (i.e. mixing state) and shape (i.e. morphology) attributes to more cooling at both top of the atmosphere and surface, and the combined effect is ~6% more warming than the spherical particles. The significance of consideration of particle shape is more in the regions where black carbon mixes with pure mineral dust, which are the most probable case over the IGB in northern India, because enhancement in the atmospheric warming will be under-estimated if particle morphology is not considered [47]. Thus, there is an urgent need for modeling studies over the IGB region to examine quantitatively the influence of particle morphology along with their mixing states on optical and radiative characteristics of aerosols with their size distribution.

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