

# Chapter 13

## Variability of Atmospheric Aerosols Over India

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### 1 Introduction

The effect of aerosols on climate through various radiative interactions (involving the atmosphere, clouds, and the cryosphere) is well recognized by the international community (e.g., Andreae et al. 2005; Satheesh and Moorthy 2005; Bollasina et al. 2011; IPCC 2013). Atmospheric aerosols play a significant role in climate change due to their ability to scatter and absorb the incoming and outgoing radiation (direct effect). In addition to this, aerosols can also impact climate through modifying cloud properties, such as droplet size distribution and cloud lifetime, a process known as “indirect effect” (Twomey 1974; Kaufman et al. 2005; Rosenfeld et al. 2014). In addition, absorption of solar radiation by aerosols can lead to changes in cloud properties known as semi-direct effect (Lohmann et al. 2001). Of late, several reports focus on the impact of carbon-containing aerosols on monsoon, even though there have been contradictions and the processes are not understood well. Further, there have been reports that aerosols might impact crop yields, stimulate tropical cyclones, cause droughts and floods, and so on, most of these not validated yet (Chameides et al. 1999; Chung and Ramanathan 2006). Carbon-containing aerosols

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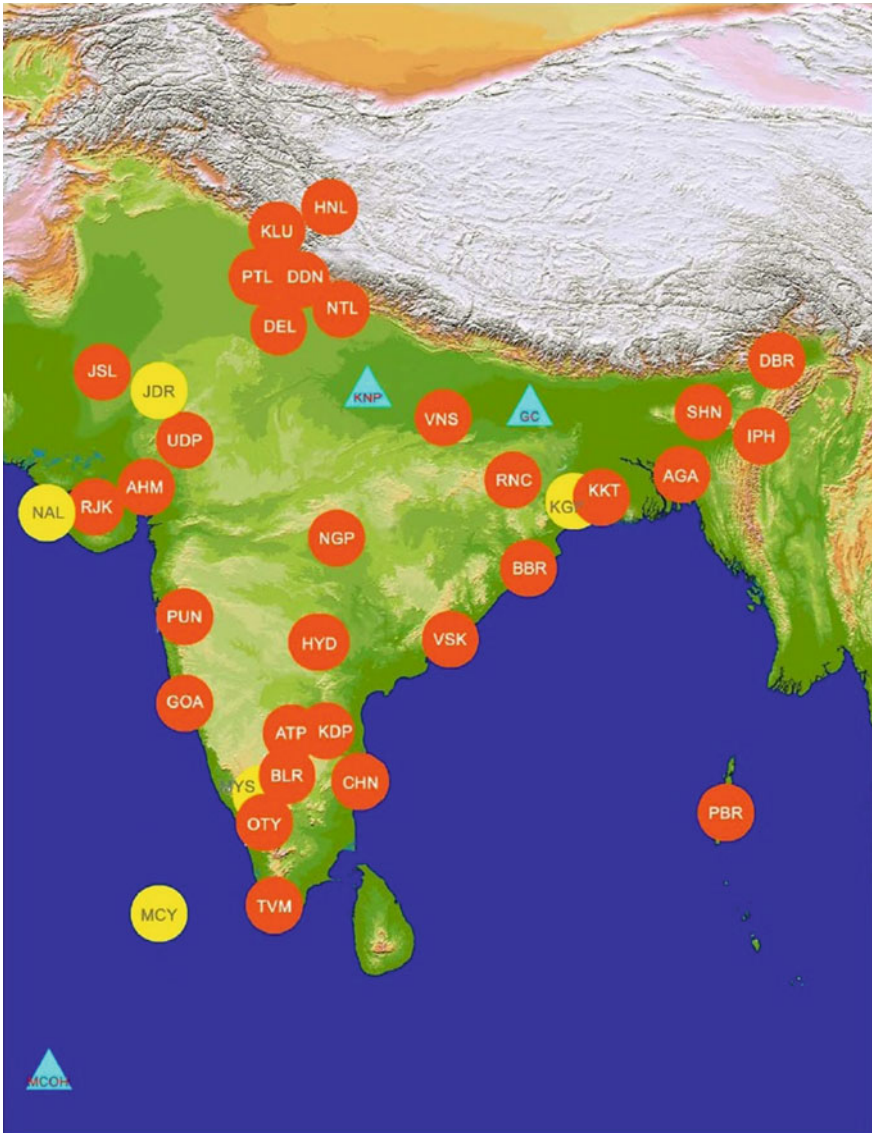
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are composed of black carbon (BC) and organic carbon (OC); however, they have different climate forcing. Major sources of both BC and OC are biomass and fossil fuel burning. While OC is of light-scattering type, the most important light-absorbing aerosol species is BC. The OC through its cloud condensation nuclei (CCN) activity also contributes to indirect forcing while BC can contribute to evaporation of clouds known as “cloud burn off” (Ackerman et al. 2000). The fact that atmospheric aerosols exhibit high degree of spatial and temporal heterogeneity which makes their role in cloud microphysics very complex. Consequently, the aerosol–cloud interactions have been a subject matter of great interest to the climate science community, and efforts are being made by making use of a wide variety of experiments onboard ground-based, airborne, and space-borne platforms (Koren et al. 2004, 2008, 2014). Thus, it is imperative that relevant aerosol quantities are being measured from ground, aircraft, and space and used to carefully answer the fundamental questions related to the aerosol–climate problem.

Systematic studies on aerosols, clouds, and radiation budget were virtually nonexistent in India till the 1980s, except a few studies based on the atmospheric turbidity measurements using Volz Sun photometers by the India Meteorological Department (IMD) (Mani 1968; Mani and Huddar 1972; Mani and Chacko 1980; Rangarajan and Mani 1982). Even though aerosol research in India dates back to 1950s, systematic studies of the aerosols commenced only in the 1980s under the Indian Middle Atmosphere Programme (I-MAP) (Murthy 1988; Moorthy et al. 1988, 1999, 2009). During the I-MAP, a project to monitor the aerosol characteristics was initiated by utilizing multi-wavelength radiometers over a few selected locations (Murthy et al. 1988; Moorthy et al. 1988, 1989, 1991, 1993), following common instrumentation, data and analysis protocols, based on a network concept involving multi-institutional collaboration. This project became operational in the mid-eighties. Realizing its scientific potential and success, this project has been continued after IMAP under ISRO’s Geosphere Biosphere Programme (ISRO-GBP) as Aerosol Climatology and Effects (ACE) having long-term objectives. With this modest beginning, networking at several geographically distinct locations, a few selected sites were equipped with a regional focus under ISRO-GBP since the 1990s (Moorthy et al. 1993, 1994, 1996; Moorthy and Satheesh 2011). Around the same time, vertical profiles of aerosols have been obtained using rocket-borne and balloon-borne payloads and Lidars (Parameswaran et al. 1984; Devara et al. 1994). These activities have enabled the increased recognition of the important role aerosols may have on climate forcing, and the ACE activity was converted to a national project namely the Aerosol Radiative Forcing over India (ARFI) project (Moorthy et al. 2009; Moorthy and Satheesh 2011) with the regional network of observatories known as ARFINET. This currently comprises of 41 aerosol observatories spread across India covering urban, remote, island, coastal, inland, semiarid, arid, and remote mountain regions over the mainland and the adjoining oceanic regions (Babu et al. 2013) (Fig. 1 for ARFINET locations). Around the same time, the global network of aerosols such as AEROSOL ROBOTIC NETWORK (AERONET) also established surface-based sites in India (Holben et al. 2001). In addition, recent years witnessed the expansion of several other regional networks also in India such as SKYNET (Kim et al. 2005).

In this chapter, long-term changes and variability of aerosol concentration and its spatial vertical distribution over the Indian region are discussed, based on the results from several experiments conducted over the Indian region. Aerosol observations carried out during various experiments/campaigns are discussed, and analysis was



**Fig. 1** Locations of ARFINET sites marked on a digital elevation map of India, where each circle represents the ARFINET observatories, identified by its short name. The red circles denotes the stations which are operational and yellow circles denotes which are not operational. The triangle symbols indicate AERONET stations

made on different types of aerosols with special emphasis to black carbon aerosols. Potential linkages between the changes in aerosol, radiation and clouds are also discussed.

## 2 Variability in Aerosol Properties

During 1940s and 1950s, climate science community was focused on hygroscopic aerosols because of their ability to act as CCN. Sea salt aerosol was the most important species of interest during this period (Woodcock 1953, 1957; Monahan 1968; Tsunogai et al. 1972; Lovett 1978; Monahan et al. 1982, 1983). Stuhlman (1932) suggested that the bursting of bubbles produced jets of water, which results in the production of small seawater droplets. Kohler (1936, 1941) proposed that strong winds can cause spray at the wave crest and are responsible for the airborne sea salt particles. Kientzler et al. (1954), using high-speed photographic study of bursting bubbles, concluded the mechanism of production of seawater droplets from bubble breaking. By early 1960s onwards, attention got gradually shifted to mineral dust aerosols due to their ability to get transported to long distances from their sources. A number of reports regarding the transport of aerosols from continents to ocean and vice versa are available in the literature (Eriksson 1959, 1960; Toba 1965a, b; Junge 1972; Delany et al. 1973; Prospero 1979; d'Almeida 1986; Bergametti et al. 1989; Arimoto et al. 1995; Gong et al. 2003; Zender et al. 2003). The presence of Saharan dust even over the remote areas of the Atlantic and Pacific Oceans is an example (Carlson and Prospero 1972; Junge 1972; Prospero and Carlson 1972; Prospero 1979; d'Almeida 1986; Bergametti et al. 1989; d'Almeida et al. 1991). Bergametti et al. (1989) have carried out chemical analysis of aerosol samples from the Atlantic Ocean, which revealed that source is Africa. By late 1970s/early 1980s, investigators have started realizing the radiative effects of mineral dust aerosols especially the fact that dust aerosols have important role in the warming of lower atmosphere due to short-wave absorption (Alpert et al. 1998; Miller and Tegen 1998). Several studies have indicated that, in general, under clear-sky conditions, dust approximately doubles the shortwave radiation absorption. Tegen and Fung (1994) have shown that dust causes a net cooling at the surface, simultaneous with an increase in atmospheric warming. By late 1980s, most likely due to the enhanced presence of anthropogenic aerosols as a consequence of industrial revolution, several studies have focused on anthropogenic sulfate aerosols and their climate impacts (Charlson et al. 1991, 1992). By early 1990s, there was an increase in the interest of black carbon aerosols and their effect on climate due to their strong absorbing characteristics (Cachier et al. 1989; Satheesh et al. 1999; Satheesh and Ramanathan 2000; Babu et al. 2002).

### 2.1 Trends in Aerosol Optical Depth

Of late, long-term changes (trends) in aerosols have gained increased interest due to their importance to regional and global climate. There have been several studies over the Indian region to address trends in aerosols using ground-based measurements using

short-term, single-station data (Parameswaran et al. 1998; Moorthy et al. 1999; Satheesh et al. 2002; Kaskoutis et al. 2012) as well as long-term multi-station databases (Moorthy et al. 2013; Babu et al. 2013). Some investigators have also used satellite-derived products to study trends in aerosols (Ramachandran et al. 2012; Dey and Girolamo 2011). Aerosol optical depth (AOD) is a common data available on aerosols from several satellites regularly, and there are limitations arising from the large uncertainties involved in the satellite-retrieved AODs, especially over the landmass mainly due to the diverse surface reflectance and cloud contamination (Jethva et al. 2007). Studies by Zhang et al. (2005), Remer et al. (2005), Kahn et al. (2007), Levy et al. (2010), and several others have suggested that satellite data may have problems in long-term trend analysis due to the fact that noises and biases in the products can often be (mistakenly) interpreted as legitimate. Such studies are also beleaguered by the calibration drifts, which can often be (mistakenly) interpreted as trends in aerosols. Therefore, data from ground-based Sun photometers (preferably from a network), which can provide the most accurate AOD data, are the best choice for trend analysis. As on now, ARFINET is the only ground-based network with such long-term data.

The detailed regional synthesis of long-term data on AOD from the ARFINET has shown an increasing trend with a seasonal variability (Moorthy et al. 2013; Babu et al. 2013). The results are shown in Figs. 2, 3, and 4. Comparison of the turbidity coefficients in this study with those reported around late 1960s and early 1970s (Mani 1968) using Volz Sun photometer data indicates the extraordinary nature of the increase in aerosols during the last decades. The rate of increase is always high during the dry season (December–March) over the entire region, whereas the trends are rather inconsistent and weak during the pre-monsoon (April–May) and summer monsoon period (June–September) (Fig. 3). The variations in the spectral variation in AOD reveal the important contribution of anthropogenic sources on the increasing trend in AOD (Fig. 4, which reveals an increasing trend in the submicron aerosol abundance). The insignificant trend in AOD observed over the Indo-Gangetic Plain during the summer and pre-monsoon months is mainly ascribed to the competing effects of transport of dust and wash out of aerosols by the monsoon rainfall. Nonetheless, if this trend continues so, AOD at several locations would nearly double and approach unity in the next few decades. This can lead to an enhancement in aerosol-induced lower atmospheric warming by a factor of two. These observations indicate that trends in the aerosol forcing elements and their regional and global climate implications need to be better evaluated using global climate models.

Though not extensive, there are other studies on aerosol trends based on data from one or a few stations. Parameswaran et al. (1998) have reported an increasing trend in the AOD from 1989 to 1994. Satheesh et al. (2002) reported increase in AOD using two decades of ground-based Sun photometer measurements. Aerosols have an important role in determining solar radiation reaching the Earth's surface. Padmakumari et al. (2007) have made an assessment of monthly mean surface-reaching solar radiation under all-sky conditions for several stations, distributed across the Indian region, for the period 1981–2004 (see Chap. 9). They have shown that all the stations showed a decline in  $S$  ranging from  $-0.17$  to  $-1.44$   $W\ m^{-2}$  per year. The consequent annually averaged solar dimming over India for the period 1981–2004 is estimated to be  $-0.$

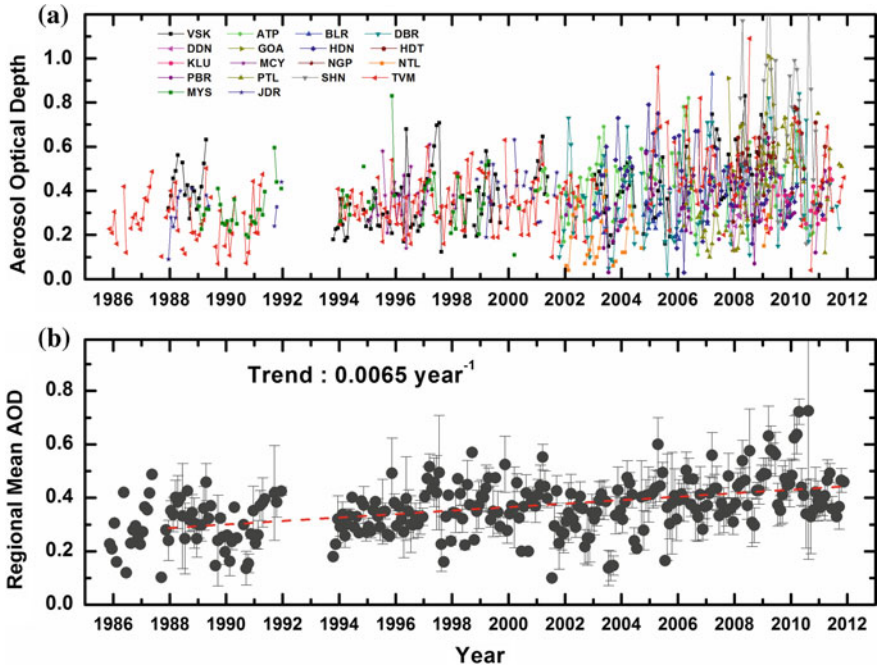


Fig. 2 a Long-term trends in aerosol optical depth at 500 nm, derived from measurements at ARFINET stations where different colors and symbols differentiate the stations. b Long-term trend in regional mean aerosol optical depth at 500 nm (color figure online)

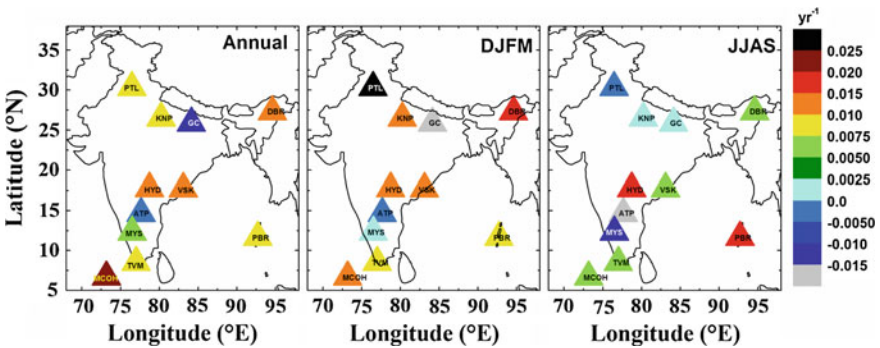
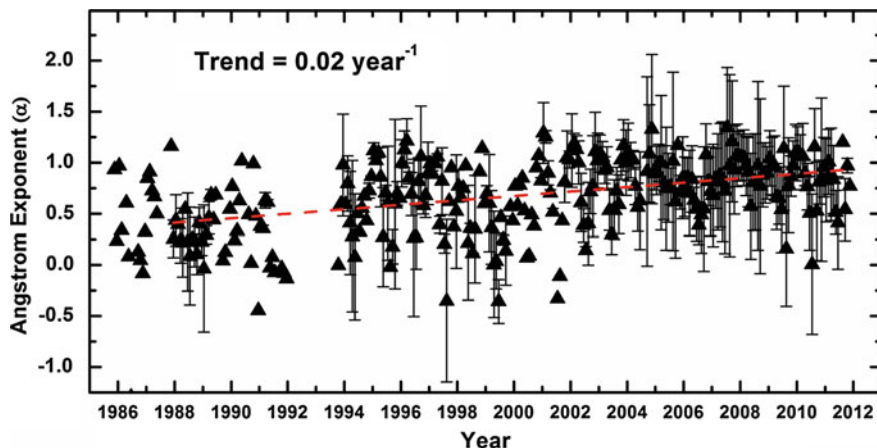


Fig. 3 Seasonal changes in the spatial variation in AOD trend over Indian region

86 W m<sup>-2</sup>, while the seasonal mean values for winter, pre-monsoon, and monsoon seasons are pegged at -0.94, -1.04, and -0.74 W m<sup>2</sup> per year, respectively. Dey and Girolamo (2011) have used ten years (2000–2010) of observations from MISR to quantify seasonal linear trends of AOD and showed that many regions have statistically significant increasing trend in the range 0.1–0.4 in the last decade. Kaskoutis et al. (2012) discussed aerosol loading variability and trends at Kanpur using AERONET



**Fig. 4** Long-term trend in the regional mean values of Angstrom wavelength exponent, an indicator of anthropogenic impact of aerosol column loading

data. Their studies show an increase in AOD and attributed to an increase in the seasonal/monthly averaged AOD during the winter and post-monsoon seasons. Aerosol sources during these seasons are dominated by anthropogenic emissions. In contrast, a weak decreasing trend is observed during pre-monsoon and monsoon seasons. Soni et al. (2012) have studied the variability in annually averaged irradiance (global and diffuse) and bright sunshine duration over 12 stations of solar radiation network of IMD for the period 1971–2005 (see Chap. 9). They have reported that the annually averaged all-sky global irradiance decreased between 0.1 % ( $0.3 \text{ W m}^{-2}$ ) and 3.6 % ( $9.0 \text{ W m}^{-2}$ ) per decade at these stations. Examining the seasonal and annual mean trends in AODs for the last decade using MODIS data over different locations in India, Ramachandran et al. 2012 have reported an increasing trend. Dani et al. (2012) have reported an increasing trend of 45 % per decade over Pune, based on Sun photometer-derived multispectral AOD measurements. Pathak et al. (2012) examined the variability in aerosol loading over the central region of the Indo-Gangetic Plains (IGP), during a decade (2001–10). An overall increase in AOD (ground-based radiometric measurements) was observed on yearly basis. Lu et al. (2013) presented the first inter-annual comparison of  $\text{SO}_2$  emissions and those retrieved from satellite data [ozone monitoring instrument (OMI)] for Indian coal-fired power plants during 2005–2012. The results show that  $\text{SO}_2$  emissions increased by 71 % during the study period.

Horizontal surface visibility is one of the simplest measures of local atmospheric pollution. Jaswal et al. (2013) have studied trends of morning poor visibility days (PVD, visibility <4 km) and afternoon good visibility days (GVD, visibility >10 km) based on 279 surface meteorological stations, well distributed over India, for the period 1961–2008. Their studies show that during the last 5 decades, all India averaged range of annual morning PVD has increased from 6.7 to 27.3 % days, while the range of afternoon GVD has decreased from 76.1 to 30.6 % days. Sreekanth (2013) has used the MODIS data to study trends in AOD over Bangalore.

They have shown an overall increasing trend, due to sustained increase in the seasonal averaged AOD during summer. Ramachandran and Kedia (2013) have used satellite-derived AOD to study inter-annual and regional variations in aerosols over six homogeneous rainfall zones in India for the month of July from 2000 to 2010 and reported that AOD over India in a drought year (2002) was higher when compared to normal monsoon years. Investigations using MODIS data for the period 2003–2012 over Delhi region show an increase in AODs by more than 25 % (Kumar 2014). Yearly mean Terra/Aqua AOD values have shown an increasing trend at a rate of 0.005/0.009 per year, respectively, with higher rates during winter (0.012/0.007 per year, respectively). An increasing trend in AOD is also reported by Soni et al. (2014) over Raniganj (7.31 %) in eastern and Korba (5.0 %) in south-east, and Godavari Valley (32 %) in the south coalfield region in India using MODIS data from 2000 to 2012 during winter and post-monsoon periods. Thus, overall, AOD over India shows an increasing trend, which is steady and stronger during dry season, and weak and inconsistent during the monsoon season.

## ***2.2 Black Carbon Aerosols: Spatial and Temporal Variability***

Major sources of black carbon (BC) are fossil fuels, biofuel, and biomass burning. Due to its ability to absorb, BC aerosols warm the atmosphere. In addition, BC, when deposited on snow and ice, can reduce the albedo. BC can contaminate other aerosols substantially, thereby altering the radiative properties of the entire aerosol system. Though there have been several reports on the climate impact of BC, several of them are contradicting to each other, for example impact of aerosols on monsoon. Recently, brown carbon, resulting from biomass burning, has attracted the attention of global climate change community because of its strong absorption characteristics in blue and UV region, with nearly no absorption in the mid-visible, unlike BC.

The ISRO-GBP annual review meeting in 1998 recognized the importance of BC aerosols on climate system, and it was decided to pursue studies of BC in subsequent years (Moorthy et al. 1999). Details of this research activity is also available in “IGBP In India 2000 A status report on projects,” edited by Narasimha et al. (2000) and published on behalf of the Indian National Science academy (INSA). Later, the Indian Ocean Experiment (INDOEX), an Indo-US project, takes measurements of BC over the Indian Ocean. During the INDOEX field campaign, extensive measurements of BC were taken over the Indian Ocean, probably the first BC measurements over the Indian Ocean region. Based on these and other associated measurements, Satheesh et al. (1999) have developed an aerosol model for tropical Indian Ocean, which demonstrated that BC contributes 11 % to composite AOD. Later, using several calibrated satellite radiation measurements and five independent surface radiometers, Satheesh and Ramanathan (2000) quantified that even though BC contributes 11 % to optical depth, its contribution to radiative forcing can be as much as 60 %. Over the Indian mainland, continuous and long-term measurements of BC were initiated under GBP. Babu and Moorthy (2001) published the first report of the anthropogenic impact on aerosol black



carbon mass concentration based on a case study from the coastal station, Thiruvananthapuram. Thereafter, several investigators reported BC measurements at various locations in India (Babu et al. 2002, 2004; Vinoj et al. 2010; Padithurai et al. 2004; Moorthy et al. 2004; Gangulay et al. 2005; Parashar et al. 2005; Dey et al. 2006; Satheesh et al. 2006, 2008, 2013; Pant et al. 2006; Dumka et al. 2006; Ramachandran et al. 2006; Safai et al. 2007; Sreekanth et al. 2007; Niranjana et al. 2007; Rengarajan et al. 2007; Beegum et al. 2009; Rastogi and Sarin 2009; Gogoi et al. 2014), and extensive station-specific as well regional synthesis of BC is available.

In a first of the kind attempt to generate a regional synthesis of aerosol properties, a mobile land campaign (LC-I) was conducted during February to March 2004 under the support of the ISRO-GBP, where simultaneous aerosol measurements were taken over spatially separated locations, using identical instruments, mounted on a mobile van, and operated following a common protocol. The LC-I has generated a wealth of information on black carbon as well as important aerosol parameters and covered an area of more than a million square kilometers (Moorthy et al. 2004, 2005; Ganguly et al. 2005). During this campaign, aircraft-based measurements were taken on the vertical profiles of BC for the first time in India, over Hyderabad. Using these data, Moorthy et al. (2004) have shown a rapid decrease in BC concentration within the atmospheric boundary layer (ABL) up to about 500 m and a nearly steady level above. As a continuation of this experiment, land campaign II (LC-II) was organized under ISRO-GBP during December 2004 by concurrent measurements from a number of observatories across the IGP. The analysis of the comprehensive database thus generated provided deeper insights on the microphysical and optical properties of aerosols over the IGP, the roles of mesoscale and synoptic meteorology in modulating them as well the significant role of the dynamics of the ABL in controlling the aerosol concentration near the surface as well as the role of long-range transport (Tare 2006; Ganguly et al. 2006; Niranjana et al. 2006, 2007; Nair et al. 2007; Rengarajan et al. 2007). Results from LC-II showed extremely high BC concentration, often exceeding similar to  $20 \mu\text{g m}^{-3}$ , prevailing during December over the Indo-Gangetic basin. Simultaneous measurements of the local ABL height and wind fields revealed a very close association between the BC concentration and the ventilation coefficient (defined as the product of the boundary layer height and the transport wind) (Nair et al. 2007). Despite their significances, all the above field campaigns had several limitations. They did not provide adequately spatially resolved data over the oceans around India. Measurements were limited to only a few of the aerosol parameters and mostly to one season or even less, simultaneous measurements over land and oceans were not taken, and vertical profiles of aerosol characteristics over the land and ocean (especially on the continental outflows) were not made. The first and perhaps the only field experiment, integrating different observation platforms and variety of scientific instruments, has been the Integrated Campaign for Aerosols, gases and Radiation Budget (ICARB). This was a multi-platform field campaign with participation from numerous institutions, where integrated observation and measurements of aerosols, trace gases, and radiation were taken simultaneously. The details of these campaigns and the major findings have been reported in the literature

(Moorthy et al. 2008, 2009; Babu et al. 2008; Vinoj et al. 2008; Beegum et al. 2008; Nair et al. 2008; Satheesh et al. 2008, 2009, 2010). All these studies showed the persistence of high AOD and black carbon concentrations near the surface.

The only long-term data on BC aerosols are from ARFINET observatories, which is now a network spanning the entire country and surrounding oceans. Some of its stations have databases of more than a decade for BC, enabling quantifying of its long-term trends. Data from Thiruvananthapuram, a remote coastal location in the southern peninsula, on the concentration of BC, normally considered as a tracer for human impact, show a decreasing trend of  $\sim 250 \text{ ng m}^{-3}$  per year. This is particularly perceptible after 2004. Decreasing trend in BC is not a unique feature of Thiruvananthapuram. Another site, Bangalore, showed decreasing trend of  $\sim 330 \text{ ng m}^{-3}$  per year. This finding has several implications. The reduction in surface BC could be considered as an indicator of the impact of stringent emission control strategies, while the increase in AOD indicates an overall increase in columnar abundance. It remains to be seen whether this increase is occurring at higher levels, above the ABL. An extensive analysis of seasonal trends in BC concentration over India is available in Babu et al. (2013). Wang et al. (2014) estimated global BC emissions from 1960 to 2007 and reported that the BC emission intensity in power plants, the residential sector, and transportation shows decreasing trend for all the regions, especially China and India. Improvements in combustion technology and changes in fuel composition are attributed to the decline of BC emission intensities.

### 2.3 *Evolution of Size Distributions*

Aerosol size distribution is one of the key characteristics required for estimating the spectral optical and CCN properties of aerosols. It is also important in determining the scattering phase function, which in turn determines the up-scatter fraction and eventually the radiative forcing. In early 1960s, Angstrom wavelength exponent was used extensively to infer on aerosol size distributions (Ångström 1961), especially when they followed a simple inverse power law relation. Using spectral variation in AOD, following the inverse power law representation, Angstrom wavelength exponent can be derived. The value of Angstrom wavelength exponent depends on the ratio of the concentration of large to small aerosols. Early measurements of Angstrom wavelength exponent was mostly taken over the continental locations where spectral variation in AOD was best represented following Angstrom power law (Mani and Huddar 1972; Mani and Chacko 1980; Rangarajan and Mani 1982). It is known that aerosol spectral optical depths contain information pertaining to their size distribution (King et al. 1978). Making use of this property, it is possible to infer the columnar size distribution of aerosols by numerically inverting the spectral optical depth measurements, following linear inversion technique by King (1982), which involves the numerical inversion of the Mie integral equation. Adapting this technique, it has been observed that over the coastal regions influenced by both land and ocean environments, the aerosol size

distributions were shown to depict changes associated with synoptic meteorology, showing the signatures of advected sea salt aerosols during marine air mass periods (Moorthy et al. 1991, 1993). By 1990s, several measurements were taken over oceanic regions where bimodal distributions are observed (Satheesh et al. 1998). Recent measurements show that a simple power law (even at two different wavelength ranges) or even bimodal distribution is not sufficient to get a best fit to spectral optical depths indicating multimodal size distributions indicating the influence of several sources (Kompalli et al. 2014).

Though near-surface aerosol size distribution measurements are being taken as part of various field campaigns, such information is still not adequate. There have been only a few attempts to develop climatology of aerosol size distribution (e.g., Gogoi et al. 2014; Kompalli et al. 2014; Kanawade et al. 2014). There have been improvements in the algorithms to derive column-averaged volume size distribution from Sun photometer measurements (Dubovik et al. 2006). Limited validation studies show agreement for the derived size distribution against in situ (surface as well as aircraft-based) measurements (Haywood et al. 2011; Smirnov et al. 2011; Gogoi et al. 2014; Kanawade et al. 2014), but these inversion products have not been comprehensively validated. There has also been some progress in measuring the aerosol size distribution down to a few nanometer size (e.g., Feldpausch et al. 2006) which enabled studies of new particle formation.

Khemani et al. (1982) have taken aerosol size distribution measurements based in Pune, and their report is among one of the earliest reports of size distribution over India. Measurements of size-distributed aerosol mass concentrations were taken at Bombay using a quartz crystal microbalance cascade impactor (Sharma and Patil 1992), and a power-function fit was applied to the size distributions. Extensive characterization of the seasonality of near-surface aerosol size distributions and their association with columnar AOD over the coastal regions is found in Pillai and Moorthy (2004). Dhanorkar and Kamra (1993) have reported that size distribution of aerosol particles is bimodal in shape based on measurements taken in Pune. Niranjana et al. (1997) have used spectral optical depths following the constrained linear inversion algorithms to derive aerosol size distributions and yielded a bimodal aerosol size distribution in Visakhapatnam. Size-differentiated concentrations of atmospheric aerosols measured in a suburban area of Agra city during 1992–1993 have revealed a bimodal distribution (Kulshrestha et al. 1998). Pandithurai et al. (1997) have approximated aerosol size spectrum measured in Pune with a composite power law distribution function.

New particle formation from precursor gases has been proposed as a source of nucleation aerosols under different environments, from polluted urban to pristine high-altitude Himalayas (Chate and Devara 2005; Moorthy et al. 2011; Kompalli et al. 2014). Several mechanisms for new particle formation have been proposed. Condensation of a binary mixture of sulfuric acid and water; secondary organic aerosol formation involving condensation of low- or nonvolatile organic compounds; and homogeneous nucleation of iodine oxides are examples. Laboratory studies have shown these mechanisms are significantly affected by temperature, humidity, and the surface area of preexisting particles. Many investigators

measured high concentrations of ultra-fine particles, which indicate efficient new particle formation in the troposphere under low condensation sinks (Chate and Devara 2005; Kompalli et al. 2014). This is in contrast to the prevailing assumption that aerosol nucleation takes place only during the daytime and typically from sulfuric acid. Carefully examining the changes in the size distribution associated with new particle bursts, estimates of the growth rate of aerosol sizes due to coagulation and condensation processes were obtained under contrasting environments (Kompalli et al. 2014). Since not many such observations are available in the literature, more future studies are required to understand the nucleation mechanisms especially during nighttime. One of the major problems hampering our current understanding of the process of new particle formation is that these new particles are smaller than the lower size detection limit of the most instruments available as of today. They are only observed after some particle growth has occurred.

## 2.4 Vertical Distribution

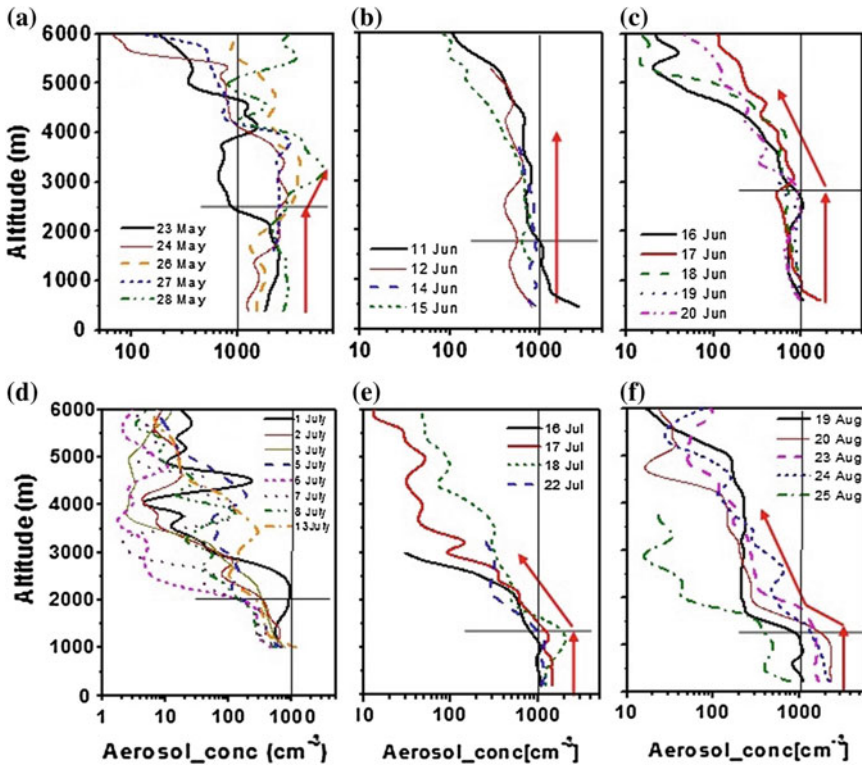
When the amounts of absorbing aerosols such as BC are significant, AOD and chemical composition are not the only determinants of aerosol radiative effects, but the altitude of the aerosol layer relative to clouds (if present) is also important. Aerosols that are transported to higher altitudes are much more likely to travel long distances. Thus, as compared to AODs, aerosol vertical profile retrievals are very important as they provide more insights on the aerosol impacts on climate such as warming of the atmosphere and the impact on the thermal structure and stability of the atmosphere. Over India, aerosol vertical distribution was studied using rocket- and balloon-borne instruments (Jayaraman et al. 1987; Ramachandran and Jayaraman 2003), ground-based lidar (Parameswaran et al. 1984; Devara et al. 1995; Jayaraman et al. 1995; Satheesh et al. 2006, 2009; Niranjana et al. 2007; Raj et al. 2008), twilight photometry (Ashok et al. 1984; Padmakumari et al. 2003, 2005, 2006), airborne lidar (Gadhavi and Jayaraman 2006; Satheesh et al. 2008), and space-borne lidar (Padmakumari et al. 2012; Prabha et al. 2012; Gautam et al. 2011). There also exist a few studies using aircraft (Moorthy et al. 2004; Tripathi et al. 2005; Babu et al. 2008; Safai et al. 2012; Padmakumari et al. 2013a). The advent of CALIPSO (McGill et al. 2007; Huang et al. 2008) also improved our knowledge about vertical as well as spatial distribution of aerosols, which enabled to study the 3D structure of desert dust transport and biomass burning aerosols (Liu et al. 2008; Uno et al. 2008). ICARB aircraft campaign has shown the presence of elevated aerosol layers during the pre-monsoon season (Satheesh et al. 2008). A substantial fraction (as much as 50–70 %) of aerosols were found above (reflecting) clouds (Moorthy et al. 2009; Satheesh et al. 2008, 2009). Aerosol extinction coefficient at higher atmospheric layers (>2 km) was found much larger compared to that near the surface (by as much as a factor of 2–3). The aerosol-induced warming was mostly confined below 2 km over the southern Indian region, whereas it is found up to 4 km over the central Indian regions. The results showed a strong meridional gradient in warming ( $\sim 4$  K) at atmospheric

levels above 2 km (Satheesh et al. 2008, 2009, 2010). The data from CALIPSO also paved way to extensive investigations of the assessment of the impact of aerosols above clouds (Chand et al. 2009).

Ground-based and satellites are proven to be suitable platforms for the measurements of atmospheric aerosol. Recently, aircraft is also proven to be an effective platform for aerosol measurements over a reasonably large spatial domain. Over India, measurements of aerosol vertical distribution during the monsoon season are very sparse to understand aerosol–cloud interactions. Aerosols are found to inhibit the cloud growth (Albrecht 1989) and also found, under suitable conditions, to invigorate deep convection (Koren et al. 2005). Most of the earlier airborne measurements were taken during the pre-monsoon season. CAIPEEX, a national experiment, conducted during the pre-monsoon and monsoon seasons with an instrumented research aircraft led a pathway to understand the vertical distribution of aerosols and their interaction with monsoon clouds (Kulkarni et al. 2012). CAIPEEX was conducted over different parts of India in 2009, while over central India in 2010 and 2011. Aerosol vertical variability and spatial distribution over different locations in continental India was studied using the airborne observations during CAIPEEX from May to September 2009 (Padmakumari et al. 2013a). The instruments used onboard for aerosol measurements were Passive Cavity Aerosol Spectrometer Probe (PCASP) and Aethalometer (see Kulkarni et al. 2012 for more details). CAIPEEX data during 2009 revealed the presence of elevated aerosol layers during the pre-monsoon as well as during the monsoon period (Padmakumari et al. 2013a, b). These elevated layers are mostly influenced by the depth of ABL, the origin of air mass trajectories, and also the presence of clouds (Padmakumari et al. 2013a). Figure 5 reveals that during the monsoon, aerosol number concentration showed strong vertical gradient and a transition is clearly observed between the boundary layer and the free troposphere as compared to that during the pre-monsoon (Fig. 5a) and just before the monsoon onset (Fig. 5b). It is also noted that during pre-monsoon, aerosols showed significant variation spatially at the elevated layers as compared to that in the boundary layer, while during monsoon high variability is observed only in the boundary layer. The surface-level aerosol number concentration and the height of boundary layer were found to influence the AODs significantly.

Vertical profiles of BC mass loading over Hyderabad and Bengaluru during monsoon 2009 showed decreasing trend from surface to an altitude of 7 km, but anomalously high BC loadings were encountered at different altitudes (Safai et al. 2012). CAIPEEX measurements over northeast India also revealed the formation of a second layer of BC in the upper atmosphere, which generates heating of  $\sim 2$  K/day (Rahul et al. 2014).

During the pre-monsoon, near the foot hills of west of the Himalayas, fine mode as well as coarse mode aerosols showed elevated layers. Characteristics of aerosols in the elevated layer and their interaction with clouds were also studied. During CAIPEEX, clouds profiled above the elevated aerosol layers showed increase in droplet number concentration ( $200\text{--}1400\text{ cm}^{-3}$ ) and small effective radius ( $3.5\text{--}5\text{ }\mu\text{m}$ ) at the cloud base indicating the observational evidence of aerosol–cloud interactions. Figure 6 shows the presence of ice phase above 6 km at temperatures



**Fig. 5** Aerosol vertical distribution at different regions during different phases of monsoon. **a** Pathankot (pre-monsoon). **b** Hyderabad (just before monsoon onset at Hyderabad). **c** Hyderabad (after monsoon onset at Hyderabad). **d** Bengaluru (monsoon). **e** Bareilly (peak monsoon in IGP). **f** Bareilly (peak monsoon in IGP). The horizontal lines represent mean height of boundary layer obtained from radiosonde profiles. The vertical and slant thick lines represent the aerosol gradient and transition from boundary layer to free troposphere during different phases of monsoon

lower than  $-15\text{ }^{\circ}\text{C}$ , representing the presence of dust aerosol acting as a potential ice-forming nucleus (Padmakumari et al. 2013b). The presence of dust aerosol in the elevated layers affects the ice microphysics. The vertical profiles of aerosols and cloud parameters obtained from aircraft were also compared with the simultaneous satellite measurements of CALIPSO and CloudSat (Padmakumari et al. 2012).

## 2.5 Aerosol Direct Radiative Forcing: Past and Present

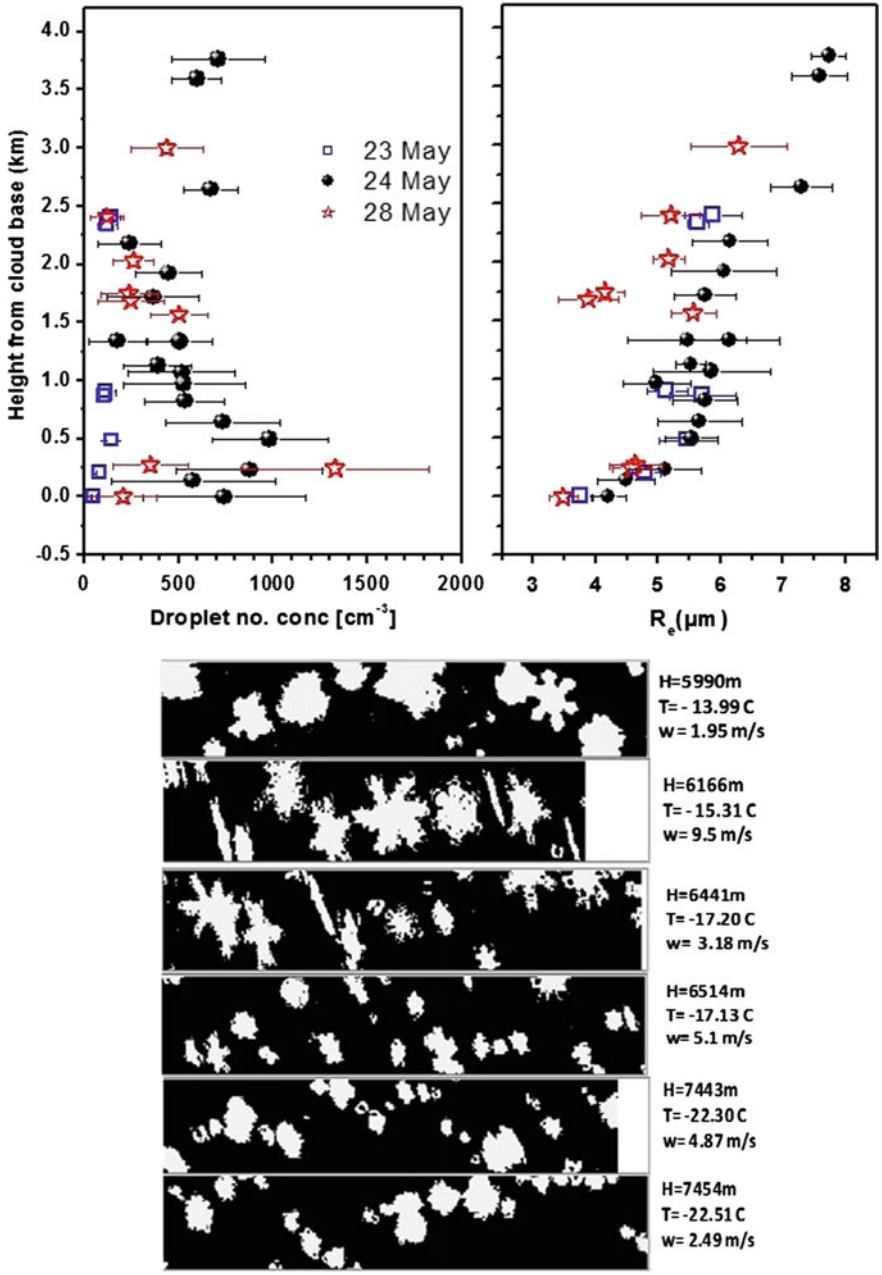
There have been several efforts in India to estimate aerosol radiative forcing under clear-sky conditions (Satheesh and Ramanathan 2000; Babu et al. 2002; Pandithurai et al. 2004; Ganguly et al. 2005; Pant et al. 2006; Ramachandran et al. 2006 and so on). Over the Arabian Sea and the Indian Ocean, using simultaneous measurements of aerosols and surface-reaching solar fluxes, Jayaraman et al. (1998) have estimated with

the change in surface solar fluxes with AOD. Satheesh and Ramanathan (2000) quantified the aerosol forcing simultaneously at the Earth's surface and at the top of the atmosphere over the tropical northern Indian Ocean. They reported that aerosols are responsible for the decrease in shortwave radiation at the ocean surface by 12–30  $\text{W m}^{-2}$  and increase in that top of the atmosphere by 4–10  $\text{W m}^{-2}$ . They argued that this threefold difference and the large magnitude of the observed surface forcing are largely due to the absorption of solar radiation by black carbon.

Babu et al. (2002) made simultaneous measurements of aerosols, black carbon, size-segregated aerosol mass concentrations over a continental urban location. Their estimated aerosol forcing was  $-23 \text{ W m}^{-2}$  (at the surface) and  $+5 \text{ W m}^{-2}$  [at the top of the atmosphere (TOA)] even during relatively cleaner winter months. The consequent atmospheric heating was  $0.8 \text{ K day}^{-1}$  for cleaner period and  $1.5 \text{ K day}^{-1}$  for moderately turbid conditions. Pandithurai et al. (2004) estimated that during the dry season, aerosol forcing at the surface, the TOA, and the atmosphere over Pune (an urban site) were  $-33$ ,  $0$ , and  $33 \text{ W m}^{-2}$ , respectively.

Since detailed chemical composition was not available during 1980s, often a hybrid approach is being used to estimate aerosol radiative forcing from spectral optical depths and other available measurements (Satheesh et al. 1999, 2002; Babu et al. 2002). We follow a modified version of this approach by Satheesh and Srinivasan (2006). This method can be used to derive simple aerosol models that are “optically equivalent” and can simulate the observed aerosol optical properties and radiative fluxes, from spectral optical depth measurements. Thus, aerosol single-scattering albedo and hence aerosol radiative forcing can be estimated. The purpose of the proposed method is to estimate clear-sky aerosol radiative forcing and not to obtain the exact chemical composition of the aerosols. By incorporating this information in radiative transfer models, aerosol radiative forcing can be estimated. This method is not a substitute for the measurement of aerosol chemical composition, but a method to estimate aerosol radiative forcing over regions where aerosol chemical composition data are not available. The spectral AOD and Angstrom wavelength exponent data from ARFINET observations are used as input, and derived aerosol composition for 1985 and 2012 is shown in Fig. 7. Aerosol single-scattering albedo thus estimated for 1985 is  $\sim 0.97$  and for 2012 is  $\sim 0.84$ .

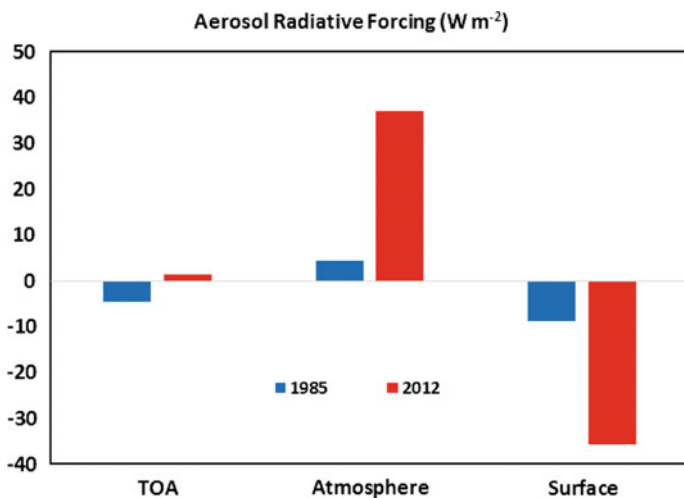
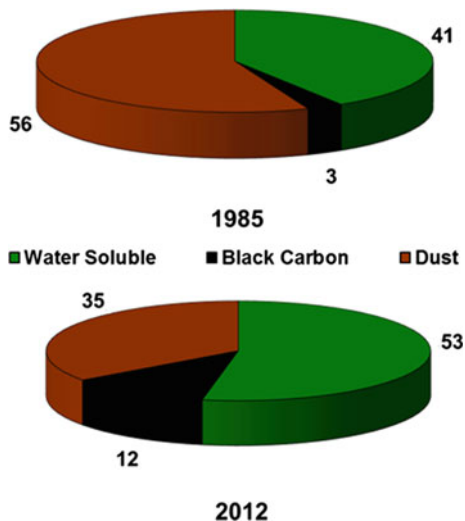
The aerosol spectral optical depth, Angstrom wavelength exponent, and single-scattering albedo as obtained above are incorporated in a Discrete Ordinate Radiative Transfer code, which was designed and developed by the University of California, Santa Barbara (Ricchiuzzi et al. 1998). A radiative forcing at the surface, TOA, and atmosphere for 1985 and 2012 is shown in Fig. 8. The TOA forcing changed sign from  $-4.6$  to  $+1.4 \text{ W m}^{-2}$  from 1985 to 2012 due to the increased presence of absorbing aerosols. The estimated surface and atmospheric forcing changed from  $-8.8$  and  $+4.2 \text{ W m}^{-2}$ , respectively, in 1985 to  $-35.7$  and  $+37.1 \text{ W m}^{-2}$  in 2012, respectively.



**Fig. 6** Upper panel shows the droplet number concentration and effective radii of the clouds profiled above the elevated pollution layers. May 24 and 28 are more polluted as compared to May 23. Lower panel shows the ice phase of the cloud observed above 6 km on May 28 measured by Cloud Imaging Probe. These measurements are first of its kind near the foot hills of the Himalayas during the pre-monsoon season



**Fig. 7** Aerosol composition for 1985 and 2012 derived using spectral AOD and Angstrom wavelength exponent data from ARFINET observations as input



**Fig. 8** Radiative forcing at the surface, TOA, and atmosphere for 1985 and 2012

### 3 Conclusions

The first regional synthesis of long-term primary data from the ARFINET has revealed a statistically significant increasing trend with a significant seasonal variability. Comparison of the data for 50 years reveals the phenomenal increase in aerosol loading. The rate of increase is consistently high during the dry months (December to March) over the entire region. However, the trends are inconsistent and weak during the pre-monsoon and summer monsoon period. The trends in the spectral variation of AOD reveal the significance of anthropogenic activities on the

increasing trend in AOD. If this trend persists, AOD at several locations will nearly double and approach unity in the next few decades. This can lead to an amplification of aerosol-induced lower atmospheric warming by a factor of two. However, a regionally averaged scenario can be ascertained only in the coming years, when longer and denser data would become available. These observations indicate that regional and global climate implications of such trends in the forcing elements need to be better assessed using GCMs.

The only long-term data on BC aerosols are from ARFINET observatories, which is now a network spanning the entire country and the surrounding oceans. Some of its stations have databases of more than a decade for BC, enabling quantification of its long-term trends. Data from Thiruvananthapuram, a remote coastal location in the southern peninsula, on the concentration of BC, normally considered as a tracer for human impact, show a decreasing trend of  $\sim 250 \text{ ng m}^{-3}$  per year. This is particularly perceptible after 2004. Decreasing trend in BC is not a unique feature of Thiruvananthapuram. Another site, Bengaluru, showed decreasing trend of  $\sim 330 \text{ ng m}^{-3}$  per year. This finding has several implications. The reduction in surface BC could be considered as an indicator of the impact of stringent emission control strategies, while the increase in AOD indicates an overall increase in columnar abundance. It remains to be seen whether this increase is occurring at higher levels, above the ABL. Improvements in combustion technology and changes in fuel composition are attributed to the decline of BC emission intensities.

Aerosol size distribution is one of the key characteristics required for estimating the spectral optical and CCN properties of aerosols. It is also important in determining the scattering phase function, which in turn determines the up-scatter fraction and eventually the radiative forcing. Early 1960s, Angstrom wavelength exponent was used extensively to infer on aerosol size distributions (Ångström 1961), especially when they followed a simple inverse power law relation. New particle formation from precursor gases has been proposed as a source of nucleation aerosols under different environments, from polluted urban to pristine high-altitude Himalayas. Laboratory studies have shown these mechanisms can occur in the atmosphere and are significantly affected by temperature, humidity, and the surface area of preexisting particles. Carefully examining the changes in the size distribution associated with new particle bursts, estimates of the growth rate of aerosol sizes due to coagulation and condensation processes were obtained under contrasting environments. Since not many such observations are available in the literature, more future studies are required to understand the nucleation mechanisms especially nighttime. One of the major problems hampering our current understanding the process of new particle formation is that these new particles are smaller than the lower size detection limit of most instruments available as of today and are only observed after some particle growth has occurred.

Aircraft is also proven to be an effective platform for in situ measurements of atmospheric aerosol, apart from ground-based and satellite measurements, over a reasonably large spatial domain. In situ measurements are very much essential as aerosols are found to inhibit the cloud growth and also found to invigorate deep

convection under suitable conditions. Most of the earlier airborne measurements were taken during the pre-monsoon. A national experiment called CAIPEEX conducted during pre-monsoon and monsoon seasons with an instrumented aircraft led a pathway to understand the aerosol vertical distribution and their interaction with monsoon clouds in India. CAIPEEX data revealed the presence of elevated aerosol layers during pre-monsoon as well as during monsoon and mostly influenced by the depth of ABL, origin of air mass trajectories and the presence of clouds. Such studies also revealed that during the monsoon period, aerosol number concentrations exhibit strong vertical gradient and a transition between the ABL and the free troposphere. The spatial distribution shows significant variation at the elevated layers as compared to that in the ABL during pre-monsoon, while high variability in the ABL during monsoon. The surface-level number concentration and the height of ABL were found to influence AOD significantly.

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