Evolution of aerosol research in India and the RAWEX–GVAX: an overview

K. Krishna Moorthy[1,2,*], S. K. Satheesh[1,3] and V. R. Kotamarthi[4]

[1]Centre for Atmospheric and Oceanic Sciences, Indian Institute of Science, Bengaluru 560 012, India
[2]Indian Space Research Organization, Bengaluru 560 231, India
[3]Divecha Centre for Climate Change, Indian Institute of Science, Bengaluru 560 012, India
[4]Environmental Science Division, Argonne National Laboratory, Argonne, IL, USA

Climate change has great significance in Asia in general, and India in particular; and atmospheric aerosols have a decisive role in this. The climate forcing potential of aerosols is closely linked to their optical, microphysical and chemical properties. Systematic efforts to characterize these properties over the Indian region started about 5 decades ago, and evolved over the years through concerted efforts in the form of long-term scientific programmes as well as concerted fields experiments. All these have resulted in this activity becoming one of the most vibrant fields of climate research in India and have brought several important issues in the national and international foci. The field experiment, RAWEX–GVAX (Regional Aerosol Warming Experiment–Ganges Valley Aerosol Experiment), conducted during 2011–12 jointly by the US Department of Energy, Indian Space Research Organization and Department of Science and Technology, has emerged as a direct outcome of the above efforts. This overview provides a comprehensive account of the development of aerosol–climate research in India and south Asia, and the accomplishment and newer issues that warranted the above field campaign. Details of RAWEX–GVAX, the major outcomes and the subsequent and more recent efforts are presented, followed by the way forward in this field for the next several years to come.

Keywords: Aerosols, climate change, ICARB, RAWEX–GVAX,

Introduction

ATMOSPHERIC aerosols, tiny particles in solid/liquid phase ubiquitously dispersed in the atmosphere, are important in imparting significant forcing to weather and climate, regionally and globally, impairing the visibility, degrading the ambient air quality and the environment, and impacting human health[1–8]. Studies on atmospheric aerosols, especially their environmental and radiative implications, have evolved in India over the last few decades with the concerted efforts using ground-based network, satellite data, thematic campaigns and modelling. The implicit climate impacts of these have great significance to Asia in general, and India in particular due to diverse geographical features and high population density of this region. In the subtropical eastern Northern Hemisphere, the upper Ganges valley has some of the highest, persistently observed aerosol optical depths (AOD) values[9]. This region is also home to about 700 million people and accounts for about 20% of the Indian gross domestic product. Approximately 70% of coal-fired thermal power plants in India are located over the Indo-Gangetic basin. It houses heavy industries such as steel, cement, and power plants and is one of the ancient continuously farmed regions of the world.

Ground-based and space-borne measurements have shown that the Indo-Gangetic Plains (IGP) has some of the highest AOD/loading, persistent during the dry winter and pre-monsoon seasons, extending spatially across the IGP and over to the Bay of Bengal (BoB)[10]. This is also one of the few regions across the globe that shows a persistent steady increasing trend in AOD[11]; increasing surface dimming[12], and enhanced mid tropospheric warming[13,14]. In recent decades, this area has been heavily exposed to air pollution arising from fossil fuel and biomass burning as well as mineral dust from the Thar Desert[15]. Growth in industries such as cement factories, steel mills and the coal-fired thermal plants has added to existing regional sources of aerosols, such as agricultural residue burning. Tiny hydrophilic particles in the smoke act as hubs for water droplets to form clouds. As these particles and clouds absorb and scatter sunlight, they change the way heat is distributed in the atmosphere. The perturbations to regional radiative balance through direct, indirect and semi-direct effects of this high and increasing aerosol loading have strong climate implications. Modelling studies have shown these to significantly impact the monsoon rainfall, hydrological cycle and the snow-cover over the Himalayas (which remains the largest source of fresh water supply to the major rivers flowing through this region and act as the lifeline for its millions of population), though the exact nature and extent of this impact are not unequivocally understood or quantified[16–18]. Some studies have suggested that the haze over the Ganges Valley region will increase air temperature, which might draw

*For correspondence. (e-mail: krishnamoorthy@caos.iisc.ernet.in)

moisture from the ocean and intensify the monsoon. Other studies indicate that the increased heat will cause clouds to burn up/evaporate during early stages of their formation. The persistent winter fog in the region is another cause of much concern, and several studies have been proposed to understand the economic, scientific and societal dimensions of this problem.

Over the years, the Ganges valley has undergone rapid industrialization. Coal remains the primary energy source. The region is dotted with many large and super thermal power stations, cement factories and steel mills, all extensively using coal as fuel. Climatologically, a high-pressure ridge over the wintertime IGP confines the pollutions within the shallow atmospheric boundary layer (ABL), leading to enhanced concentrations. The synoptic regional wind is dominated by westerly flow, conducive for advection of significant quantity of mineral dust from the western Thar Desert and the vast arid regions of the Middle-East and Africa[19]. The period during late December and early January marks the traditional planting of the year's second crop in much of the Ganges valley. This is preceded by large-scale burning of the agricultural residues of the previous harvest, adding significant aerosols to the regional aerosol mix[20]. The northwesterly winds at the 850 hPa level favour long-range transport of biomass burning plumes. Aerosols during this period would better represent the anthropogenic component of the composite aerosols in this region and aid in evaluating their impacts at regional and global scales[21].

These issues have been in the centre stage of aerosol-climate research in India and several focused programmes and field experiments have been conceived and executed. A major outcome has been the establishment of a network of aerosol observatories under Aerosol Radiative Forcing over India (ARFI) project of the Indian Space Research Organization (ISRO)-Geosphere Biosphere Programme (I-GBP) and air-borne measurements of crucial aerosol parameters under Regional Aerosol Warming Experiment (RAWEX) again under I-GBP. The details of these would be appearing in the sections to follow. Aiming to take advantage of these efforts to address the basic issues of aerosol–climate interactions and aerosol–cloud interactions as well as their regional implications, by making long-term and concurrent measurements using an extensive suite of advanced ground-based instruments from a strategic location, the joint Indo-US field experiment Regional Aerosol Warming Experiment–Ganges Valley Aerosol Experiment (RAWEX–GVAX) was formulated. The rationale of conducting such a major experiment included the fact that the monsoon systems occupy and impact about 30% for the world's land mass and impact a large fraction of the world population and the aerosols have a decisive impact on monsoons. The monsoons represent a collective thermodynamic response of the atmospheric–oceanic coupled system, modelling of which demands coupled Earth system models and excellent validation. In addition, South Asia exhibits extensive cloud cover from early spring to summer, modulating the regional energy budget, the intrinsic dynamics of the system, and a potential for the local pollution to affect the sequence of events that lead to monsoon by changing regional and local energy budgets. As such, this field campaign has its focus on measurements of clouds, precipitation and complex aerosols to study their mutual interactions. The comprehensive dataset thus generated could be used to constrain convection, cloud properties and aerosols[21].

These data were analysed and synthesized with model simulations and a number of publications providing deeper insights into aerosol–radiation–cloud interactions have emerged.

Evolution of aerosol–climate studies in India

The earliest scientific measurements of atmospheric aerosols over India could be traced to the airborne measurements (from 17°N; probably Hyderabad) in the nineteen fifties[22]. Subsequently, isolated efforts to quantify the atmospheric turbidity over India, using an year-round Volz sunphotometer data, have been made by Mani *et al.*[23–25]. However, systematic characterization of the spatio-temporal properties of atmospheric aerosols, their spectral optical and microphysical properties over the Indian region and assessing their implications for radiation balance and climate forcing started only more recently; in the nineteen eighties, with the efforts under ISRO[26–29]. Since then, well-focused and concerted efforts have been and are being made to generate the climatology of aerosols (optical, microphysical and chemical properties), over distinct geographical regions of Indian landmass and the oceans surrounding it, through a series of national programmes such as the Indian Middle Atmospheric Programme (IMAP), I-GBP, the Indian Ocean Experiment (INDOEX)-India (INDOEX-i) Programme, the Indian Climate Research Programme (ICRP) and the like, using ground-based network stations[27,28,30,31], co-ordinated multi-platform field experiments[16,32,33], measurements aboard balloons and rockets[34,35] and lidars[26–40], besides making extensive use of data from operational satellites[9,41–44]. These efforts, spanning over the last more than three decades, have generated a wealth of information on the physico-chemical properties of these particles[2,26,45–47], their spatio-temporal variations[48–52], responses to meso-scale and synoptic meteorology[53–55], long-range transport[49,51,56,57], vertical distribution[58–62], microphysics[63], and more importantly on their regional radiative forcing[16,64–66] with possibly large implications, especially on the monsoons, Himalayan glaciers and stratospheric ozone[6–8,13,67–72] though several of them are location/region-specific.

As on now, the ISRO maintains the largest and densest network of aerosol observatories (ARFINET) over India (in fact, over South Asia) under its ARFI project, covering almost the entire distinct landmass regions of India

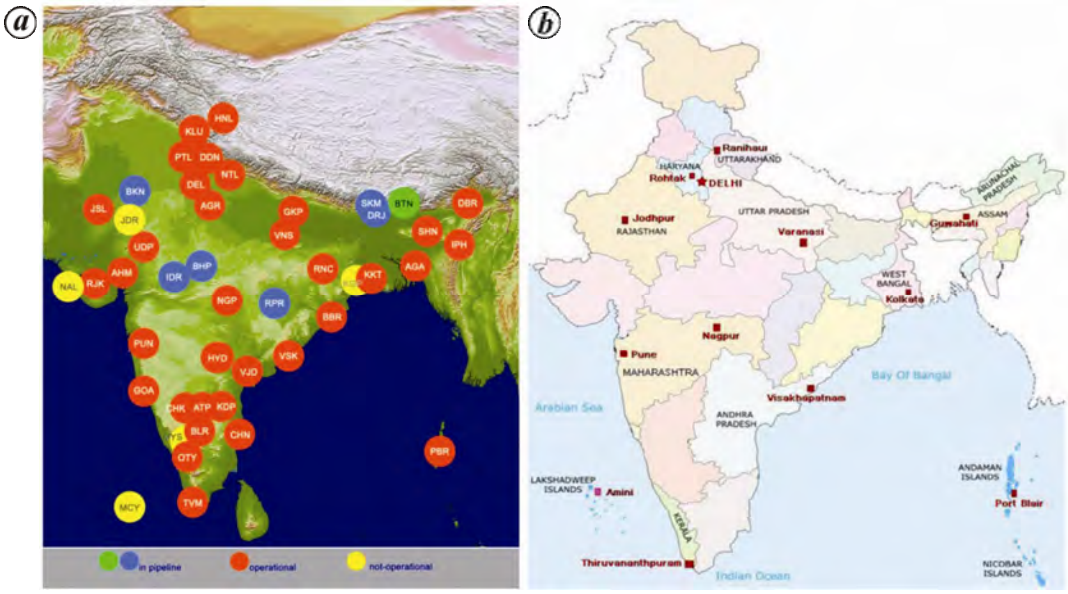

Figure 1. ***a***, The ARFINET observatories under ISRO; ***b***, the network of IMD.

and oceanic regions around it (Figure 1 *a*), under its geosphere biosphere programme (I-GBP). In recent years, the India Meteorological Department also has set up a network of 12 sky radiometers (Figure 1 *b*) under its environmental monitoring program (http://www.imd.gov.in/section/nhac/dynamic/emrc/emrc.htm). These jointly provide columnar spectral AOD information over India with a fairly good spatial resolution during clear sky period, and of other climate-sensitive aerosol species such as the Black Carbon (BC) mass concentration. Information on the scattering and absorption properties, size distribution and new particle formation are also obtained at some of the supersites.

Besides these networks, there are isolated measurements by individual research and academic institutions. The data from these measurements have been useful for providing the much needed insights into the spatio-temporal distribution of aerosols over this densely populated region, the impact of natural processes associated with synoptic and meso-scale meteorology, long-range or trans-boundary transport, as well as contributions from human activities associated with industrialization, urbanization, transport sector and energy use; besides providing the much needed primary data for policy decisions, planning mitigation strategies, health studies and so on.

One of the most significant results that has emerged from these studies, having relevance to RAWEX–GVAX, is the high absorption efficiency of Indian (Asian) dust compared to its African counterpart. Extensive studies using thermal IR brightness temperature data from METEOSAT Satellite[73–75] and synthesizing ground-based spectral AOD measurements with Infrared Differential Dust Index (IDDI) derived from satellite based thermal IR data[76], it have been shown that the Asian desert/arid dust aerosols are much more absorbing in nature than the African dust and also that the absorptive nature of Asian dust is higher for the aged dust than for the nascent dust. This increased absorbance arises partly from the higher hematite content in the Asian dust[15], which leads to higher absorption in the short wavelength regime, and the chemical processing of dust[77], besides mixing of dust with other absorbing species such as BC[78,79]. The anthropogenic aerosols, especially carbonaceous, from biomass burning in the neighbouring locations (for example burning of agricultural residue), exhaust from automobiles in the urban areas and household emissions all add to the aerosol burden significantly leading to mixed aerosols. Moorthy *et al.*[76] have further reported that the dust AOD has the annual maximum during pre-monsoon and early summer months, when the AOD at 500 nm goes well above 1, and the AOD spectra tend to become flat signifying abundance of coarse mode particles. Ground and satellite-based measurements as well as back-trajectory analyses have confirmed the prevalence of elevated dust layers over the entire north and northwest India as well as the Gangetic plains during these seasons, every year[9,15,80,81]. Model simulations have shown that such absorbing aerosol layers, when are sufficiently high and strong, can act as elevated heat pump (EHP) and enhance the upper level convections, invigorate clouds and strengthen the summer monsoon[69]. The secondary organic aerosols (SOA) and the so called brown carbon (BrC) emanating from biomass burning also contribute significantly to aerosol absorption at short wavelengths[82].

Field experiments and regional characterization

The network observatories are sources of continuous and long-term primary data on aerosols and are useful for

developing climatology of aerosol parameters for modelling, impact assessment, for delineating impacts of meteorology and also for examining long-term trends. The results derived from the network data are, however, mostly location-specific and have limited spatial validity (unless suitably assimilated and validated), due to the large heterogeneity in aerosol sources and sinks. Field campaigns, on the other hand, are thematic in nature and often address specific issues related to processes and interactions in a given spatial domain; which is often much larger. They also involve focused and concerted efforts, employ bunching of resources (instruments, measurements and models), and the results have better regional representation. The two decades since 1995, witnessed a number of field campaigns/experiments over the Indian region; both, big and small; over land, ocean as well as airborne. The pre-INDOEX cruise of 1996 was the first major field experiment undertaken by the Indian aerosol science community[83]. It examined the outflow of continental pollutants over the tropical Indian Ocean on the one hand and quantified the contribution of sea-salt aerosols, produced *in situ* over the oceans by the action of winds, on the other. This was followed by a series of campaigns; the ocean cruises in 1996 and 1997 associated with satellite data validation; the pre-INDOEX campaign in 1997; the first and intense field phases (FFP and IFP) of the INDOEX and its Indian component INDOEX-India programmes (1998 and 1999); several subsequent short and isolated shipboard measurements around Indian peninsula; the Arabian Sea Monsoon Experiment (ARMEX (I & II) during 2003–04) under the Indian Climate Research Programme (ICRP); and the mobile as well as fixed station Land Campaigns (LC-1 and LC-2) over landmass under the I-GBP. These campaigns established, perhaps for the first time, (i) the large atmospheric forcing of BC aerosols despite these contributing ≤ ~10% to the composite aerosol mass[67], (ii) the dependency of atmospheric forcing of aerosols more on the mass fraction of BC to the composite aerosols than on the absolute concentration[84], (iii) the large vertical heterogeneity in aerosols leading to higher aerosol abundance (in the column) away from active source regions of higher concentration near the surface[85], (iv) the internally mixed nature of aerosols during pre-monsoon and summer seasons leading to enhanced absorption[78] and (v) spatial heterogeneity in the altitude variation of aerosol single scatter albedo giving rise to a west to east gradient in aerosol heating rate in the atmosphere[48] over the peninsula.

ICARB

Despite their significance, 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 made, 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 (as on today) field experiment, integrating different observation platforms and variety of scientific instruments, has been the Integrated Campaign for Aerosols, gases and Radiation Budget (ICARB), done in two phases; the first during spring of 2006 followed by the W-ICARB during the winter of 2008 (refs 32, 33). These campaigns synergized intense and highly spatially resolved measurements of most of the aerosol parameters by deploying specially instrumented ships making measurements over >4 million km^2 of oceanic areas around Indian peninsula, aircrafts making altitude resolved measurements of key aerosol parameters over the continental outflow over oceans in tandem with the cruises, and fixed network observations of the time variation of aerosol parameters over mainland and islands as well as satellite data. It was unique in producing the first 3-D characterization (physical, chemical and optical) of aerosols over the Indian domain, delineating the various geophysical processes controlling these and assessing the radiative impacts[66]. Figure 2 shows the domain and details of activities under the ICARBs. A wealth of information has emerged (and still continues to emerge) from the ICARB data. These campaigns have been chiefly instrumental in moulding our current understanding of south Asian aerosols and their regional impacts; a mid-term account of which is available in the excellent review by Lawrence and Lelieveld[86]. However, the most important outcomes of ICARB are some of the 'discovery class' results, of significant regional implications. These include:

1. The discovery of elevated aerosol layers in the lower troposphere, with a northward gradient in its altitude, vertical extent and the atmospheric heating rate[14]. The synthesis of ground-based and airborne lidar measurements and radiative transfer models have shown that the atmospheric heating rate due to this elevated aerosol layer increased from ~0.5 K d^{-1} over northern Indian Ocean to reach as high as ~2 K d^{-1} over Central India. Furthermore, 50–70% of the aerosol abundances were above low-level clouds. These clouds act as highly reflecting background and amplify aerosol absorption through multiple reflections. Under such conditions, if the aerosols are internally mixed with BC, the aerosol-induced warming above clouds gets enhanced further by a factor of two to three. Besides, height of this aerosol layer increases from 1 km to 5 km as we move from the Indian Ocean to Central India. Radiative transfer computations revealed that strong absorption by these elevated aerosol layers and the consequent atmospheric heating at higher altitudes over northern India cause meridional gradients in temperature at around 4 km height, conducive to trigger dry convection, and drawing of

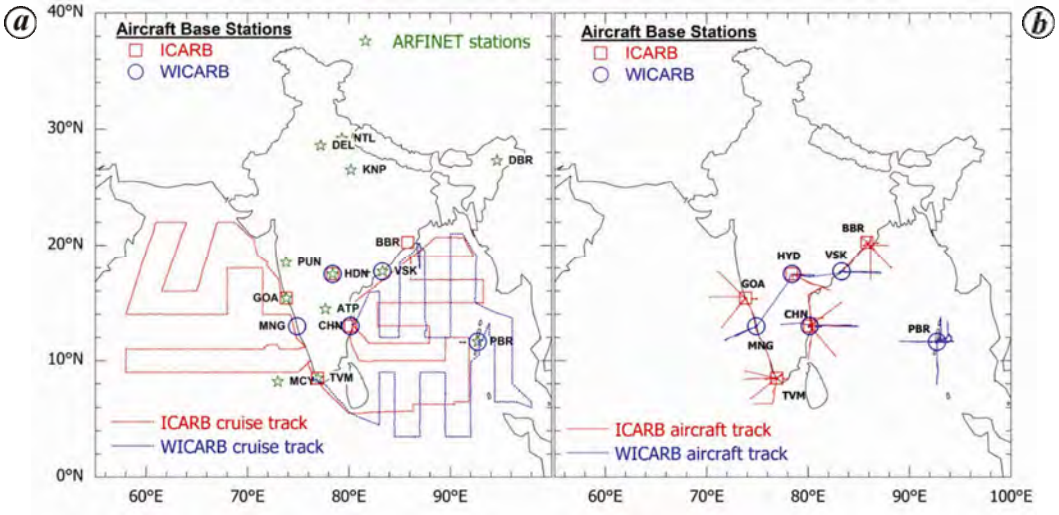

Figure 2. Configuration of the ICARB: ***a***, Represents the ocean and land segments in which, the lines over the oceanic regions around Indian peninsula depict the track of the oceanographic research vessel *Sagar Kanya* with specially instrumented laboratory for aerosol and atmospheric measurements. The red lines correspond to ICARB-2006 and blue colour the W-ICRAB (2009). The green-coloured star symbols represent the ground-based ARFINET observatories, from where continuous measurements of aerosol parameters have been carried out. ***b***, Shows the configuration of the air segment; the points were the bases from which aircrafts were operated, and the lines represent the ground track of the sorties.

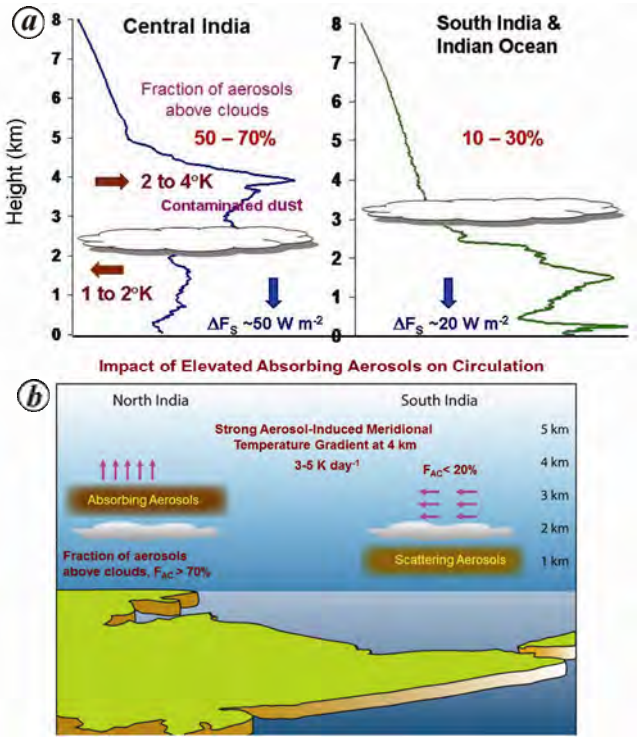

Figure 3. ***a***, Contrasting nature of the vertical distribution of aerosol extinction profile from ICARB airborne lidar measurements; with strong elevated layers over Central India. ***b***, A cartoon indicating the consequence of the elevated warming, based on the heating rates estimated using the profiles. Details are in the text.

moist air from the Indian Ocean more vigorously towards north, as shown schematically in Figure 3.

2. A first-time estimate of the 3D regional radiative forcing over South Asia, by synthesizing the ICARB data has shown a significant vertical heterogeneity[66].

3. Synthesizing the cruise measurements with those from the network and data from lidars, Nair *et al.*[71] have shown large zonal and meridional gradients in the aerosol induced lower atmospheric heating rate across Indian Peninsula; from <0.5 K d^{-1} at 55°E (over western Arabian Sea) increasing to >1.5 K d^{-1} over eastern BoB (100°E) with contrasting meridional gradient on either side of the peninsula.

4. The ICARB results revealed, for the first time, the surprisingly high anthropogenic aerosol fraction in southern BoB, than over the apparently more polluted head BoB, and also hypothesized the significant role of commercial shipping emissions to it[87].

5. First time vertical profiles of aerosol single scatter albedo, scattering and absorption coefficients and number size distributions[60,88,89], and

6. The contrasting roles of ocean biogeochemistry and anthropogenic activities in bringing about the spatial distinctiveness of ultrafine particle concentration respectively, on the west and east coasts of India[90]

Chemistry

While physical and optical characterization of aerosols evolved as above, chemical characterization has not been sidelined. Chemical characterization of the components of the complex aerosol mixture is important in source fingerprinting, inferring on their optical (scattering and absorbing) properties and hence potential radiative implications; the heterogeneous chemistry involving aerosols; as well as in understanding the impacts on environment and health. In the early days, the research was mainly dependent on analysing the composition of rainwater and

inferring the water soluble components of the atmospheric aerosols, washed out by the rain[91]. Subsequently, high-volume and cascade impactors were used to collect ambient aerosols from oceanic and continental environments and analyse them in the laboratory. Extended-period studies have revealed the seasonality in aerosol composition; with a significant signature of marine aerosols during monsoon season and sulphate aerosols during winter[92] and significant presence of soil-derived trace elements (Al, Fe, Mn and Ca) and sea-salt over the oceanic regions[93] around Indian peninsula. While the mineral components were attributed to dust transported from the Arabian Peninsula, the sea-salt aerosols were considered to be produced *in-situ*. In those days, the CI/Na ratio in aerosols had near-sea-water values, especially in the giant size (>2.5 μm dia) regime (unlike the large chlorine depletion seen in the recent period, which is attributed to photolysis reactions involving sulphates). However, these studies were rather isolated and pertained only to a small fraction of the composite aerosol mass. Perhaps, the first extensive chemical characterization to quantitatively estimate most of the constituents of aerosols in the south Asian outflow into the Indian Ocean, leading to development of an aerosol model, emerged during the INDOEX[64,94]. These have emphasized the dominant contributions of carbonaceous aerosols and mineral dust in the aerosol absorption, sulphates, particulate organic matter, nitrates, ammonia as well as sea-salt constituting the composite aerosols in the outflow. Airborne measurements by Mayol-Bracero *et al.*[95] revealed the increased share of carbonaceous aerosols in the upper residual continental air than in the marine ABL. It was also shown that anthropogenic aerosols contribute ~65% to the columnar optical depth. BC, which formed ~6% of composite aerosol mass, contributed 11% to the columnar AOD and more than 50% to the aerosol-induced atmospheric heating[66,96] and brought the carbonaceous aerosol into a major focus in aerosol–climate forcing studies.

In parallel, and independently, long-term and spatially resolved measurements of BC over India were initiated under I-GBP, based on the findings from the network measurements[97]. Currently regular measurements of BC are being carried out from a large number (>60) of locations in India. BC, emitted during all incomplete and low-temperature combustion (mainly of fossil fuels and biomass), is an efficient tracer of anthropogenic activities. The strong absorption over a wide spectral range, fine size and chemically inert nature makes this species of special interest in aerosol–climate studies. There exist a large number of papers, characterizing BC from different locations in India starting with Babu *et al.*[98]; though regional syntheses are still to emerge in a major way[99]. Also these measurements did not address the major components of the carbonaceous species – the BrC and the SOA.

All the above efforts, though have provided deeper insights into some of the chemical properties of aerosols and their possible radiative implications, detailed analyses enabling source fingerprinting, evidences of heterogeneous chemical interactions leading to modification of species while in the atmosphere, as well as the biogeochemical implications over Indian region began only more recently, with the extensive chemical analysis of aerosols over IGP[77,100]; the continental outflows into Arabian Sea and BoB and the organic carbon (OC) and EC components of the carbonaceous aerosols[101,102]; the BrC, having large specific absorption in the blue and shorter wavelengths and originate mainly from biomass burning[82] and their optical properties[103,104]; the chemical properties of aerosols pertaining to the Himalayan regions[105] as well as the oceanic regions around the subcontinent[106] under different campaigns of ISRO-GBP. These studies have revealed (i) acid processing of mineral dust over the IGP during its atmospheric transport leading to reduced fractional solubility of aerosol Fe compared to the anthropogenically impacted dust transported from the Southeast Asia with enhanced fractional Fe solubility, (ii) observation of significant amount of trace metals of anthropogenic origin in the mineral dust deposited over BoB and its seasonality, (iii) higher crustal ratio of Fe in the long-range transported mineral dust during spring, leading to the increased absorbance in short wavelengths, (iv) The first field measurements of BrC in the continental outflow to the BoB and the Arabian Sea, showing an inverse hyperbolic wavelength dependence and having a mean mass absorption efficiency of 0.45 $m^2\ g^{-1}$ and identification of the effluents from agricultural waste burning as a major contributor to this, and (v) delineation of the outflows from distinct regions (such as the middle east to the northern Arabian Sea, the IGP to northern BoB, and from the Southeast Asia to the southern BoB) and the role of emissions from the commercial ship traffic to the enhanced anthropogenic fraction in the southern BoB/ northern Indian Ocean. Synthesis of ICARB measurements with model and satellite-based measurements of chlorophyll over oceans has brought out the contrasting roles of ocean biogeochemistry and anthropogenic activities in producing ultrafine particles from gas to particle conversion, respectively over the Arabian Sea and BoB[90].

Extensive information on the aerosol chemical composition over the IGP also emerged as a result of the Land Campaign II (LC-II) organized under ISRO-GBP during December 2004. The campaign provided a comprehensive database on the optical, microphysical and chemical properties of aerosols over the Indo-Gangetic belt[48,49,51,107–110]. Most of these studies have shown the persistence of high AOD and BC concentrations near the surface. Potential Source Contribution Function (PSCF) analysis was employed to identify regional sources of secondary aerosol formation at Allahabad[111] while measurements were made of the atmospheric abundances of

BC, organic carbon (OC) and water-soluble organic carbon (WSOC) during winter from urban and rural locations in northern India and brought out the dominant signatures of biomass burning sources (wood-fuel and agriculture waste) at urban sites with OC/BC ratios in the range from 2.4 to 14.5 in contrast to the OC/BC ratios at the rural site (2.1–4.0) influenced by emissions from coal-fired industries. This emphasized that comprehensive measurements of BC, OC and WSOC/OC ratios over northern India are crucial in modelling climate impact of carbonaceous aerosols on a regional scale. Behera and Sharma[112] analysed aerosol samples collected from Kanpur for ionic species (NH_4, SO_4, NO_3, and Cl), carbon contents (EC and OC) and elemental contents. Based on a mass reconstruction approach they differentiated primary and secondary components of measured aerosol and established that secondary aerosol formation (inorganic and organic) was responsible for significant mass of aerosol (approximately 50%); a similar observation was made by Ram *et al.*[113].

Spatial synthesis, observations and models

During the evolutionary phase of aerosol–climate research in India, most of the measurements and forcing estimates remained location-specific, despite several of them, especially those from the ARFINET stations, using long-period data[28,114–117] to delineate the seasonality, spatiality, and the role of prevailing meteorology. Regional synthesis of these data to yield a regional picture and its large seasonality associated with the contrasting monsoon circulations, rainfall and mesoscale processes over this region, as well as modeling were attempted basically only in the campaign mode[17,49,85,118–120]. Satellite data, though available, suffer from large uncertainties, arising from the use of unrealistic land-surface reflectance values[121], over the heterogeneous landmass of India, in the retrieval techniques.

The first-time major effort for a spatial synthesis (of multi-station and multi-platform experimental data) has been by Beegum *et al.*[119,120] for columnar and near-surface aerosols. They have shown the spatial distinctiveness and the seasonality of aerosol types (anthropogenic, dust and sea-salt) over the different parts of Indian mainland and delineated the importance of long-range transport (especially of mineral dust from the Middle East, eastern Africa and West Asian deserts, and of sea-salt from the Arabian Sea and Indian Ocean). Syntheses at sub-regional scales have established and quantified, to a certain extent, the significant impacts of convective mixing and associated meso-scale dynamics of the local boundary layer and local human activities; all modulated by synoptic meteorology, biomass burning and urban activities in producing the observed spatiality and seasonality[45,48,51,79,85,107,108,122–125].

Modelling

The surface network observations being long-term but 'point' measurements, each with limited spatial representativeness, and the satellite data providing only a 'snap shot' despite their global nature, modelling efforts are essential for prescribing aerosols for assessing the regional and global radiative and climate impacts. Most of these models rely on emission inventories, chemical interaction between the species, and meteorology to simulate the aerosol fields in space and time and hence are bound by the accuracies and resolution of those primary parameters. As such, they differ significantly in the simulated outputs and most of the models under-predict the surface and columnar aerosol abundance, chemistry and their seasonality due to a variety of reasons[99,126–128]. However, they are still being used for regional and global impact assessment studies, their seasonality and vertical distribution as well as in providing inputs for global climate models. The widely used chemical transport models for simulating aerosols include GOCART (Goddard Global Ozone Chemistry Aerosol Radiation and Transport)[129], CHIMERE[130], WRF-Chem (Weather Research and Forecast coupled with Chemistry)[131], SPRINTARS[132,133] (Spectral Radiation-Transport model for Aerosol Species). MOZART[134], MATCH (Multiple-scale Atmospheric Transport and Chemistry model)[135] and GEOS-Chem. There are several other models used by different scientific groups in research and development mode. Despite the developments and improvements over the period, the models differ among themselves in their simulations, even over a given region and season. Thus, extensive validation efforts are needed to ensure their applicability. In the recent years, performances of several of these models over the Indian/South Asian region have been evaluated in simulating the surface concentration, altitude distribution and columnar loading of aerosols (composite and species specific) by comparing with extensive ground-based and/or satellite measurements[99,127–129,136,137]. Of these, Moorthy *et al.*[99] and Pan *et al.*[128] have examined the performance of multiple models; the latter being the most exhaustive. All these have unequivocally revealed the large under-estimation of surface concentrations as well as columnar loading by the models, by factors ranging from 2 to as much as 10 or more, depending on the location, model, season and species. The reasons for the underestimations also varied; and included several factors such as inaccurate emission inventories, incorrect meteorology, inadequate representation of boundary layer dynamics especially under stable conditions (winter and nighttime), insufficient accuracy of dry-deposition schemes, etc. and have been univocal in emphasizing the need to improve the models[99,126,128]. Performing 3-D chemical transport model (GEOS-Chem) simulations over the Himalayan and sub-Himalayan regions, driven by GEOS-5 assimilated meteorological fields, *in situ* measured BC

in air, in snow, and AERONET-derived absorption AOD and using improved anthropogenic BC emission inventories for Asia that account for rapid technology renewal and energy consumption growth, and global biomass burning emission inventories, He *et al.*[136] have reported statistically good agreement between simulations and measurements, at locations far from urban centres. The simulations, though captured the seasonality except in winter, were again an underestimate by typically a factor of 2, the simulated BC on snow being higher by factors of 2–4 and modelled absorption AOD to be lower by a factor of >2. They attributed this partly to the meteorology over Himalayan terrain, partly to the underestimation of emissions, and the assumption of external mixing of BC aerosols in the model.

Impact assessment

Notwithstanding the above limitations, the regional impacts of aerosols over south Asia have been examined by several researchers by prescribing different aerosol fields in climate models but with highly contrasting results. For example, Lau *et al.*[69] have hypothesized that the heating of the atmosphere by the accumulation of absorbing aerosols (including dust from the western arid regions, mixed with other anthropogenic emissions) in the free troposphere on the southern slopes of the Tibetan Plateau would act as an 'elevated heat pump', which could provide a diabatic heating source that leads to additional heating in the upper and middle atmosphere as well as strengthening of the monsoon and rainfall. On the other hand, synthesizing the INDOEX data, Ramanathan *et al.*[68] have hypothesized that the large reduction of solar radiation at the Earth's surface, simultaneous with lower atmospheric warming, increases the atmospheric stability, slows down hydrological cycle and reduces rainfall during monsoon; a totally contrasting scenario. Analysing the trends in tropospheric temperatures, derived from longest available record of microwave satellite measurements over the Himalayan and upper Ganga Basin regions, Gautam *et al.*[17] have shown a significant warming to the extent of 2.7°C in the 29-year record (1979–2007), in the pre-monsoon season, in the altitude region 4–7 km. This trend has been shown to be highest when this region is strongly influenced by dust aerosols at elevated altitudes, lining up with the observations and deductions of Satheesh *et al.*[14]. The resulting enhanced tropospheric warming would strengthen the land–sea temperature gradient, and would support the EHP. Hu *et al.*[138] have shown that the increased aerosol absorption tends to increase the surface temperature, which in turn could intensify the summer pressure gradient between land and ocean and hence strengthen the monsoon rainfall. A recent study[139] has suggested that the continuous increase of human-influenced aerosol emissions over the Indian subcontinent would reduce the mean monsoon precipitation, due to reduction in monsoon cloud cover arising from the warming of the atmosphere in response to aerosol absorption. Other studies[6] have also indicated similar weakening of Indian monsoon rains from increased aerosol concentration, but due to the outcome of the slowdown in tropical meridional circulation that must be driven to counteract the energy imbalance forced by aerosols between the Northern and Southern Hemispheres. Analysing aerosol cloud interactions, Rosenfeld *et al.*[140] have addressed the apparent paradox of aerosol increasing and decreasing rainfall and observed that while the near-source impacts on cloud generation and hydrologic cycles remain relatively unknown, high concentrations of fine aerosols lead to decrease in precipitation in hilly areas during the Chinese monsoon. On the other hand a recent analysis of monsoon rainfall patterns showed a trend of increasingly extreme rainfall events over the Indian subcontinent[141].

Evolution of RAWEX

The above significant, but diverse understanding of aerosol forcing and climate implication for Indian region emerged from the sustained efforts of several decades, and the findings of the persistence of elevated layers (in the altitude range 2–5 km) of enhanced aerosol concentration during pre-monsoon season, the aerosol extinction coefficient within which often exceeding the values close to the surface, and the consequent large atmospheric warming, which could reach peak values of 3–5 K during local noon at 3–5 km altitudes, all indicate possible large implications for monsoon circulation and rainfall, though the exact impact remained inconclusive. With these issues in focus, a field experiment RAWEX has been formulated under the I-GBP, aiming at (i) characterizing the elevated layers of aerosols over the Indian region, their amplitude, frequency of occurrence, seasonality and gradients; (ii) estimating the contribution of absorbing aerosols to this; and (iii) assessing the regional climate implications using multiplatform measurements and modelling. It also envisaged establishment of high-altitude observatories in the Himalayan region, and conducting altitude resolved measurements across the Indian mainland to identify the spatial gradients in the vertical structure and its seasonality.

Formulation of RAWEX–GVAX

While the above scenario has been evolving over the Indian region, international interest in this region has also been on the raise (as mentioned in the initial pages of this article). The large and persistently increasing aerosol abundance over this region, increasing industrial and urban activities, energy demand and agricultural practices as well as the natural scenario set by arid regions adjoining

India as well as the vast expanse due west of it with its characteristically absorbing soil type, which generate abundant absorbing mineral dust aerosols (through winds), which are lofted to mid and upper troposphere by the strong summer time convective eddies and transported by the synoptic winds; all aided by the peculiar orography of this region[142] and the contrasting meteorology (intense cold and hot conditions) make this region a cauldron of complex aerosol types. To understand the regional and global climate implications of this, the Department of Energy, USA mooted a field experiment GVAX aiming at deployment of the Atmospheric Radiation Measurement (ARM) mobile facility (AMF) at the northern IGP over the high-altitude site of Manora Peak, Nainital (29°21′33.84″N, 79°27′29.27″E), for an year-round measurement of all aerosol and climate-related atmospheric parameters[21] during 2011–12. The measurement site, located about 280 km northeast of New Delhi and at an altitude of 1980 m above mean sea level (amsl) in the central Himalayas, is far from major pollution sources, and for most of the time is above the regional ABL. While due north of the station are the pristine Himalayan ranges, due south and southwest of it are regions of low elevation that merge with the vast Ganges Basin (Figure 4). Thus, the site is exposed seasonally to the pollutants lofted from the IGP and dust advected from the western deserts, to rather pristine-free tropospheric conditions, and to extensive monsoon rainfall. The main objectives of the GVAX included:

(a) Obtaining a rigorous dataset of chemical, physical and optical characteristics of aerosols, atmospheric state, radiation, cloud, and convection that can be used as a baseline dataset to evaluate radiative transfer models, aerosol parameterizations, and regional-scale impacts, and to constrain parameterizations of convection in climate models over the Indian region.

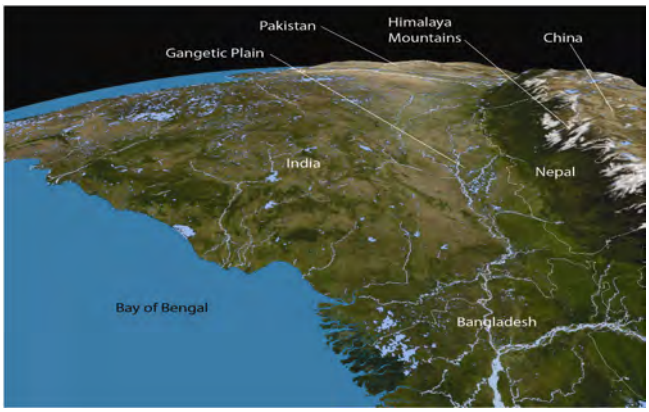

Figure 4. The domain of interest of RAWEX–GVAX experiment including the IGP, Himalayas and adjoining regions.

(b) Probe changes in the vertical distribution of aerosols as a function of day and location.
(c) Measure the fraction of SOA, which – emission profiles in India suggest – are likely a significant portion of the measured aerosol mass.
(d) Measure planetary boundary layer evolution and associated heat and moisture fluxes as a function of time of day and season.
(e) During the early spring period, which is marked by increased convective activity and clouds, evaluate the compositions of mid tropospheric aerosols and their contributions to local heating rates, in order to test current theories on the effects of aerosols on monsoon and cloud–aerosol interactions in the Ganges valley.
(f) Obtain the first measurements of the shallow convective clouds that are common during the monsoon and are typically embedded with deep convective regions.

The long-term AMF deployment enabled measurements under different meteorological regimes and aerosol abundance and types, in the wet monsoon period with low aerosol loading; in the dry, hot summer with aerosols dispersed throughout the atmospheric column; and in the cool, dry winter with aerosols mostly confined to the boundary layer and mid troposphere. Each regime, in addition, has its own distinct radiative and atmospheric dynamic drivers. The high concentration of aerosols in the upper Ganges valley, together with hypotheses involving several possible mechanisms with direct impacts on the hydrologic cycle of the region, made the experiment a unique opportunity to generate data sets that will be useful both in understanding the processes at work and providing answers regarding the effects of aerosols on climate in a region where the perturbation is the highest. The outcome from GVAX, aimed at answering some of the issues such as the reason for the discrepancy between models and measurements of absorbing aerosols; the reason for an underestimate of the contribution of absorbing aerosols from agriculture residue burning; (c) not accounting for absorbing aerosols of sizes >1 μm in the model calculation of absorption, and (d) the presence of BrC as a significant contributor to absorption.

Realizing the overlapping scientific objectives of these two experiments, the domain of the experiments and the temporal overlap, infrastructure needs and expertise involved, and the other complementary experiments existing in this region, a joint experiment RAWEX–GVAX has been formulated. The GVAX component of it was executed by the Department of Energy USA, by deploying the AMF for a period of 10 months (from June 2011 to March 2012) over Nainital, while drawing strength from the existing RAWEX measurements of the Indian side.

RAWEX–GVAX experimental phase

This complex field study utilized the ARM Mobile Facility (AMF) to measure radiative, cloud, convection and aerosol characteristics. The RAWEX–GVAX campaign was conducted under a Science and Technology Cooperation Agreement between the Government of the United States of America and The Government of India. The entire AMF instrumentation was atop a building and operated from the Aryabhatta Research Institute of Observational Sciences (ARIES) Observatory at Nainital. The data set from AMF was used to capture pre-monsoon to post-monsoon conditions to establish a comprehensive baseline for advancements in the study of the effects of atmospheric conditions of the Ganges Valley. The rationale for selecting Nainital also included the knowledge that the aerosols in this region have complex sources, including burning of coal, biomass and biofuels; automobile emissions; and dust.

The AMF (Figure 5) started operation in Nainital on 21 June 2011 and collected data until 31 March 2012. The aerosol observations performed from the Aerosol Observing System (AOS), included a 3-wavelength particle soot absorption photometer (PSAP; 470, 528 and 660 nm) to measure the particle absorption coefficient, a 3-wavelength nephelometer (450, 550 and 700 nm) to measure the total particle scattering and hemispheric backscattering coefficients, a condensation particle counter to measure the total number concentration of particles in the size range of 10 nm to 3 μm diameter, a single column CCN counter to measure total number concentration of particles in the size range of 1 μm to 10 μm diameter. The PSAP and nephelometer switched between aerosol particles of size <10 μm and <1 μm every 30 minutes.

Other measurements of interest for this study included radiosondes launched every 6 hours, a state-of-the-art passive Atmospheric Emitted Radiance Interferometer (AERI), a Multi-filter Rotating Shadow-band Radiometer (MFRSR), and a Micro Pulse Lidar (MPL) that operated between February and March 2012. In addition, various *in situ* sensors to obtain 1 min statistics of surface meteorology such as temperature, relative humidity, precipitation and wind (speed and direction) were also used. Figure 5 shows the AMF system deployed at Nainital. The AMF used a full suite of instruments for measuring short-wave and long-wave radiative transfer through the atmospheric column. While the ground radiation system collected upwelling short-wave and long-wave radiation, the sky radiation system measured the downwelling short-wave, long-wave and ultraviolet components, all at a high temporal resolution of 1 min. CO_2 and water vapour fluxes were also measured at 30 min averages. A total sky imager estimated the cloud cover over the site, while the MFRSR measured radiative properties, providing diffuse spectral radiance, direct spectral radiance, optical depth and surface albedo. The micropulse lidar (MPL) provided aerosol extinction profiles, while the radiosonde ascents provided water vapour profiles at 6 h intervals. A W-band (75–110 GHz) vertically pointing radar was used for information on cloud boundaries; a scanning cloud radar in the Ka band (26.5–40 GHz) for cloud height measurements; the narrow field-of-view radiometer to measure down-welling spectral radiance at 869 nm for determination of cloud internal structure; and a polarizing micropulse lidar to characterize droplet distribution within the clouds. The surface and lower tropospheric meteorological conditions were measured using the meteorological instruments. The surface meteorological instrumentation provided continuous data on wind speed, direction, temperature, relative humidity, surface pressure, and rain rate. The 1290-MHz radar wind profiler and the microwave radiometer profiler provided vertical profiles of temperature, relative humidity, wind velocity, and direction and the cloud liquid content. These were also supported with an optical rain gauge, and a total sky imager. The RAWEX component primarily utilized the ARFINET (Figure 1 *a*) observations.

Major outcomes from RAWEX–GVAX

RAWEX and GVAX together have brought out a number of significant results; some of these have been published recently and a few more appear in this special section. Besides, there are results, which have strong relevance to RAWEX–GVAX, but have emerged from longer-term databases and experiments (under RAWEX), falling beyond the period of the field experiment, and also from experiments and campaigns carried out subsequent to RAWEX–GVAX. The highlights, especially focusing on the vertical distribution, trends and gradients, are briefly mentioned below.

Figure 5. The AMF deployment at ARIES Nainital during the GVAX phase.

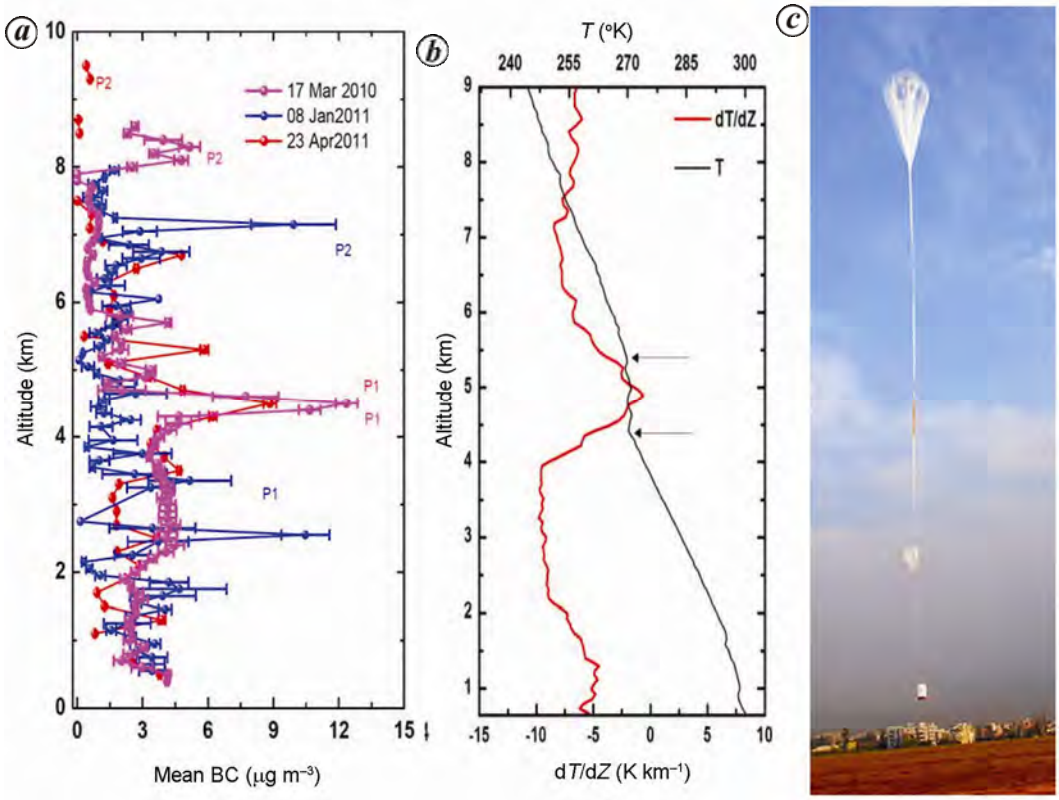

Figure 6. ***a***, Altitude distribution of BC mass concentration, deduced from high-altitude balloon ascents from Hyderabad revealing the seasonality. The elevated peaks are identified by P1, P2, etc. besides them, using the same colour code as the mother curves. Note the consistency of the elevated peaks in pre-monsoon season, and their amplitude. During winter (blue profile) the peaks occur at much lower altitudes. ***b***, The vertical profile of ambient air temperature (black curve) and the environmental lapse rate (red curve) for the spring ascent (when the effect was most conspicuous), clearly revealing the large reduction at regions around the BC peaks. ***c***, The high altitude balloon during its flight with the payload attached to the parachute.

Vertical distribution: winter versus spring

As a part of RAWEX, extensive measurements of the vertical distribution of aerosols were carried out over Central India, the IGP and the Himalayan regions, using instrumented aircrafts and high-altitude balloons, besides long-term measurements from very high altitude Himalayan locations. The gists of the findings from these are given below.

Elevated BC layers and impact on environmental lapse rate

One of the main hypotheses of RAWEX was the presence of elevated aerosol layers over the mainland of India during pre-monsoon season, as evidenced in the aircraft finding of ICARB, and that these elevated layers are capable of producing higher atmospheric warming. To study this, concurrent and *in situ* measurements of BC mass concentration and atmospheric thermo-dynamical parameters were made using instruments aboard high-altitude balloons. The balloon ascents were made from Hyderabad, during which an Aethalometer and instruments for measuring ambient meteorological parameters such as winds, temperature and relative humidity were taken aboard ~110000 m^3 zero-pressure balloons[35]. The instruments measured BC mass concentration and concurrent meteorological parameters during ascending and descending phases, and the payloads, landing with the help of parachutes, were recovered and re-used. Three such flights were made respectively during 17 March 2010, 8 January 2011 and 23 April 2011 to examine the seasonality of the profiles as well. In Figure 6 *a* we have shown the composite plot of the vertical profiles of BC mass concentrations (averaged over the ascending and descending phases of each flight) as detailed in its caption. The figure clearly reveals: (i) The consistent occurrence of sharp elevated layers of enhanced BC concentration (identified by p1, p2, etc.) in all the three profiles; (ii) A clear seasonality, with the peaks occurring at higher altitudes in spring/ premonsoon seasons and (iii) A sharp decrease in the environmental lapse rate (at times reaching close to zero) around locations of the peaks in BC (Figure 6 *b*).

Radiative forcing estimates[35] have shown that the enhanced absorption of solar radiation by the BC layer around 4 km heats the ambient air at the rate of ~2.8 K d^{-1}. Such elevated warming impacts the environmental lapse rate.

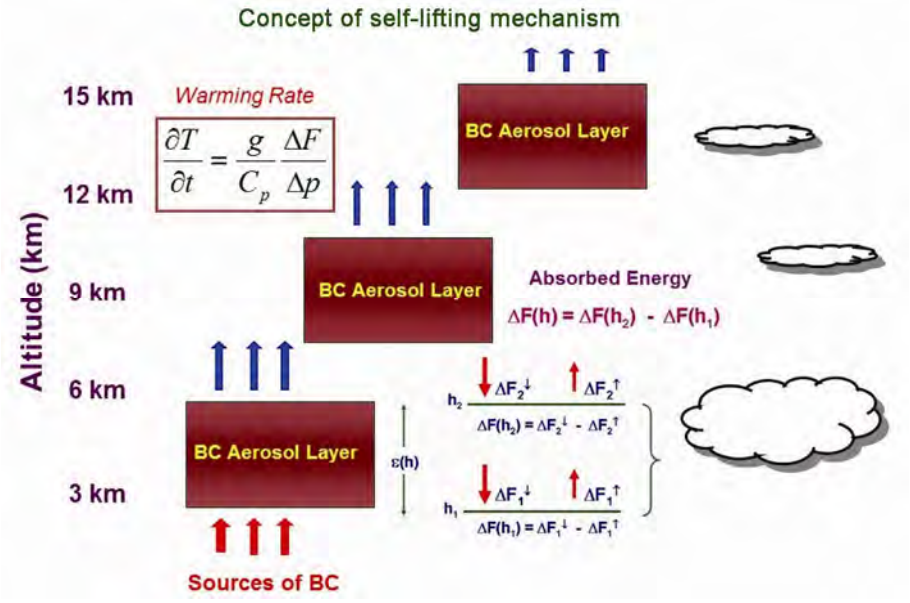

Figure 7. Schematic representation of self-lofting of carbonaceous aerosol layers.

These findings lead to the interesting concept of self-lifting, where absorption–warming–convection cycles transport black carbon aerosols from the surface to stratosphere, aided by the strong convection associated with the monsoon (Figure 7). Though self-lifting is a slow process, it can transport surface aerosols to the stratosphere throughout the year and is applicable to any part of the world as has been recently shown by de Laat *et al.*[143] by tracking a smoke plume from an Australian biomass burning episode using trajectory analysis.

While regional perturbations of aerosols, especially in the troposphere, produce strong regional signatures, their global impacts still remain largely uncertain[7]. On the other hand, significant perturbations to the stratosphere would have more global impact. Recent studies have demonstrated that air pollutants can reach stratosphere through pathways provided by the vigorous summertime circulation associated with the monsoons. During pre-monsoon and monsoon seasons (April to September), strong convection supports vertical transport of aerosols and there is a greater possibility of particles reaching higher levels. The tropical tropopause layer, being the thinnest over the Indian region during the summer, would facilitate efficient intrusion of tropospheric air along with these particles into the stratosphere, adding to the stratospheric reservoir. These aerosols are shown to interact with ozone and thus these would have implications to the stratospheric ozone layer[72].

Persistence of the layer and its spatial extent

The balloon ascents are only campaign-mode measurements, call for lot of planning and favourable weather conditions, and are rather spot measurements. With a view to examining whether the above feature occurred in isolation (despite seen on three different flights) or is a consistent phenomenon, and if so its spatial extent, the extinction profiles (derived from CALIOP (Cloud Aerosol Lidar with Orthogonal Polarization) onboard CALIPSO (Cloud Aerosol Lidar and Infra-red Pathfinder Satellite Observations) Satellite over the Indian domain were examined for seven years (2007–2013)[144]. The extinction profiles were normalized with concurrent AOD data and averaged seasonally. The results, shown in Figure 8, reveal that the above features persisted over the domain[144].

In Figure 8, the spring/pre-monsoon anomaly in the extinction profile (the averaged extinction profile for winter (DJF) season subtracted from the seasonal averaged extinction profile for pre-monsoon (MAM)) is shown as a function of altitude and latitude (averaged over the Indian longitudes). A significant enhancement in extinction coefficient is seen during pre-monsoon season, which extends almost from the equator to as far north as the Himalayan region along with a northward increase in the average height of the enhanced layer from ~2.5 km at the southern peninsula to >4 km at the Himalayan foothills as shown by the dotted ellipse in the figure. The enhancement seen over Himalayas is especially noteworthy and assumes great importance from the EHP perspective.

Aircraft measurements of aerosols across IGP

With a view to examining the seasonality in the regional vertical distribution, extensive measurements of climate-sensitive parameters of aerosols (scattering coefficients, absorption coefficients, number size distributions and single scattering albedo – SSA) have been carried out

using an aircraft instrumented with an integrating nephelometer, multi-wavelength aethalometer, aerodynamic particle sizer, photo acoustic soot spectrometer and meteorological instruments during winter 2012 and spring 2013 over Indian mainland[61]. During this, vertical profiling of aerosol properties was made from 7 base stations representing Central India (Hyderabad, Nagpur), Western India (Jaipur and Jodhpur), IGP (Lucknow, Ranchi/Patna) and Himalayan foothill region (Dehradun); typically 3 profiles being made from each base station on consecutive days. The seasonal mean of regionally averaged values of aerosol scattering coefficient at 550 nm and SSA at 520 nm are shown in Figure 9. During winter, the magnitudes of scattering coefficients within the boundary layer and close to the surface are high (200–300 Mm^{-1}) and decrease drastically with altitude to very low values (1 Mm^{-1}) by ~2 km altitude. Interestingly, in spring, though the scattering coefficients are slightly lower than the winter values within the boundary layer (<2 km), it increases to very high values (20 to 30 times the corresponding winter values) above 2 km, showing the large enhancement in aerosol concentration at higher levels. The profile remains almost steady up to ~4 km (the ceiling altitude of the aircraft for unpressurized flights). The vertical distribution of SSA shows relatively low values (higher absorption) close to the surface, increasing to higher values (0.85–0.9) as the altitude increases in both the seasons (spring and winter). However, the profile remains nearly steady above 1 km during spring, in contrast to winter, when SSA drops to the noise level above 2 km. This seasonality in the regional-mean vertical profiles is in-line with that in Figure 8, and is the crux of RAWEX–GVAX.

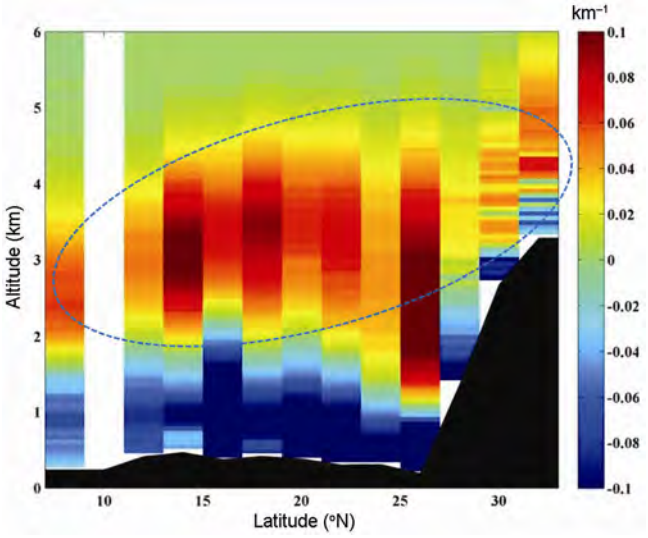

Figure 8. Spatial distribution of aerosol extinction coefficient anomaly (pre-monsoon–winter) as function of altitude and latitude, derived from long-term (7 years) CALIOP data. Details are given in the text. The dark filled structure over the X-axis schematically represents the ground elevation. Note the conspicuous pre-monsoon enhancement of extinction at higher altitudes (within the dotted ellipse).

Vertical profile of aerosols at Nainital

Measurements of aerosol vertical profile were obtained from an MPL that operated at Nainital as a part of the AMF during March, 2012. The essence of the outcome of the study is shown in Figure 10.

A significant conclusion from the measured profiles of aerosols extinction is that much of the aerosol burden in this region is due to the presence of aerosols in the lower 2 km of the atmosphere or in the planetary boundary layer. In a recent study, Feng *et al.*[18] have explored the impact of the vertical distribution of aerosol using a regional scale atmospheric chemistry and transport model (WRF-CHEM). The model has a significant bias in predicting the AOD over this region and most of the differences are due to its inability to produce the PBL maxima observed during GVAX. A series of model sensitivity tests indicate the importance of further studies that differentiate the aerosol profile between scattering and absorbing type aerosols, as the ultimate response of the regional climate depends on both the AOD profile and the

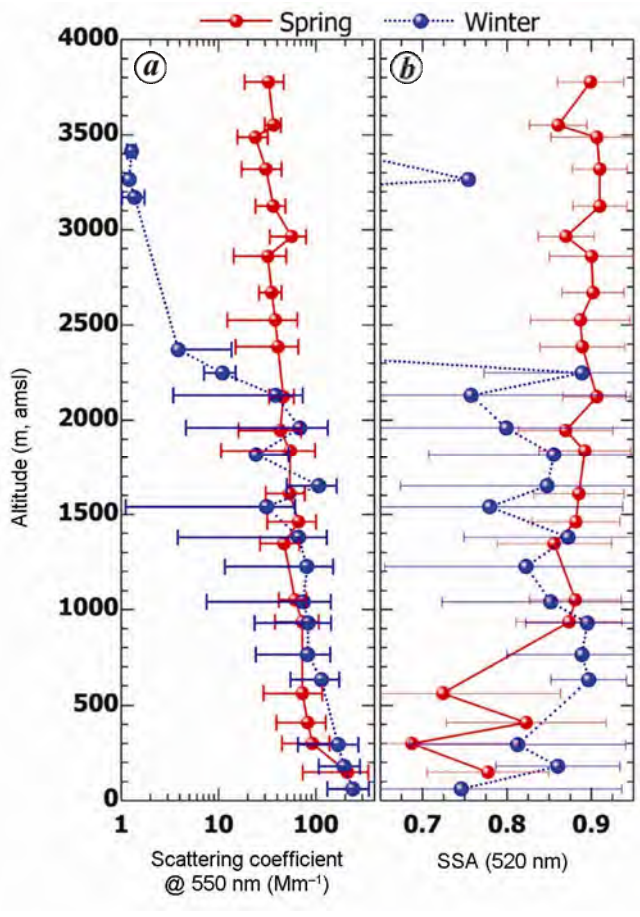

Figure 9. Regionally averaged, vertical profiles of aerosol scattering coefficient (***a***) and the single scatter Albedo (***b***) over the Indo Gangetic Plains from extensive aircraft measurements under RAWEX during winter 2011 and spring 2012, showing the conspicuous seasonality at higher altitudes, with higher (by a factor of more than 10) and nearly steady value of scattering coefficient and SSA above 2 km.

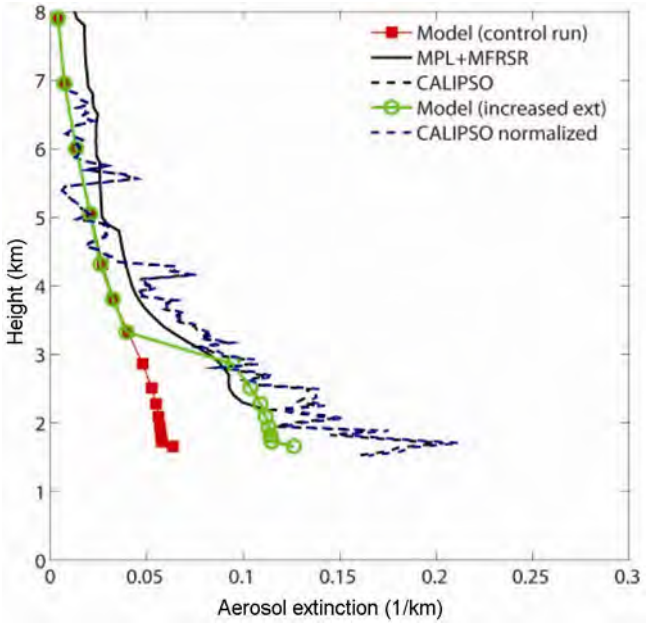

Figure 10. Aerosol extinction profiles, retrieved from the MPL and MFRSR operating at Nainital during March 2012. The concurrent profile of extinction coefficient from CALISPO is also shown for comparison. Results of model (WRF-CHEM) simulations are also shown. Details are given in text.

contribution of scattering and absorbing aerosols to the observed AOD profile in the lower 2 km of the atmosphere.

Relative humidity profiles and its effect on AOD

During the GVAX operational period, extending from June 2011 to March 2012, a microwave radiometer profiler (MRP) operated continuously, providing high temporal resolution vertical profiles of atmospheric thermodynamical parameters. In addition, RH profiles derived from 6-hourly radiosonde ascents for the entire period are also available. Similar information for the same period has also been available from Thiruvananthapuram, a coastal station at the southern tip of Indian peninsula[145]. Synthesizing data from all these, Feng *et al.*[18] have shown that there is a substantial dry bias within the boundary layer, in the model (WRF-CHEM) predicted RH under certain conditions, which could lead to an underestimation of AOD in the model. This study also emphasizes the need for future efforts to focus on a more comprehensive study of the RH measurements and model bias in the PBL over multiple years. This study has further explored the use of MRP-generated humidity and temperature profiles to develop a GVAX climatology that could be used for calculating radiative forcing for the GVAX period.

Annual cycle of aerosols during GVAX

Measurements of absorption coefficient of aerosols at Nainital were made using PSAP[146] during the GVAX. The seasonal behaviour, illustrated in Figure 11, reveals higher values for the absorption coefficient during the pre-monsoon and monsoon period, immediately followed by low values during post monsoon, increasing again into winter[146]. The absorption coefficient also exhibits a strong diurnal cycle, with low values during the daytime (post-monsoon and winter) and higher values during the evening hours. This indicated transport from below the site (from the valleys in the plains) as a primary mechanism for bringing aerosols to the AMF site, except during the monsoon, when convective mixing throughout the atmosphere is expected to dominate and thus removing the diurnal pattern from the observations. In addition, the monsoon period is also the time the region experiences air-flow impacted by biomass burning around this region[147].

Increased absorption by coarse particles

One of the significant outcomes of the GVAX is the identification of absorption by coarse particles, sized >10 μm (Figure 12), which contributed to nearly 30% of the total aerosol measured absorption, primarily during the post-monsoon period[146] and nearly 40% to the total direct radiative forcing estimated at this location.

The nature and source of these aerosols remain to be ascertained, though preliminary back trajectory calculations and meteorological conditions indicate sources that are close to the site and most likely within the valley region surrounding Nainital.

Long-term trends over Indian region

The ARFINET observatories, despite being established in a phased manner over a period of time, provide the only long-term primary data on aerosols over the Indian region; some of which span three decades. This facilitates examining long-term trends, both in near-surface mass concentrations and in the columnar spectral AOD. Though these are more or less point measurements, the high-density of the network enables a regional synthesis. Analysing these spectral AOD, Moorthy *et al.*[31] have reported a steady build-up of aerosols over the Indian region; with the regional average AOD increasing at the rate of ~2.3% year^{-1}, of the base value (0.23 at 500 nm) in 1985 (Figure 13 *a*). Studying these further, Babu *et al.*[11] have delineated the seasonality in the trends; being most conspicuous and steep during dry season and insignificant and indefinite during monsoon season. Comparing the present-day turbidity coefficients with the historical values reported[23], the authors have shown that the steady and consistent increase in the aerosol loading has resulted in a 3–5 fold increase in the turbidity coefficients over the region (Figure 13 *b*) and inferred from the spectral dependence of AOD and model simulations that

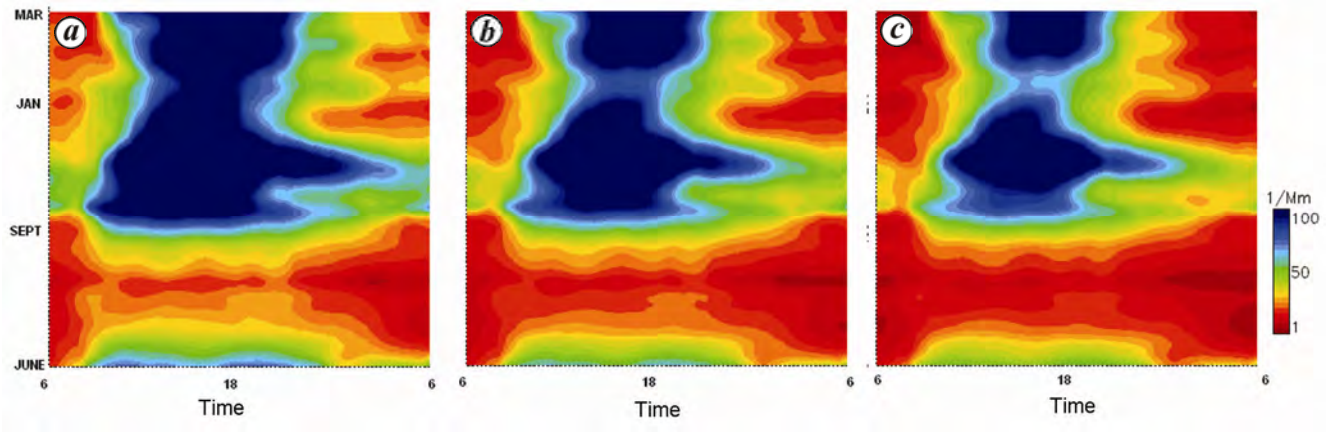

Figure 11. Absorption coefficient measured in three wavelength channels (blue, 450 nm in ***a***; green 650 nm, in ***b***, and red, 800 nm in ***c***) of the PSAP during GVAX. The *X*-axis is the diurnal cycle, starting at 06 : 00 (LT) and the *Y*-axis is the months of the experiment, extending from August, 2011 to March 2012.

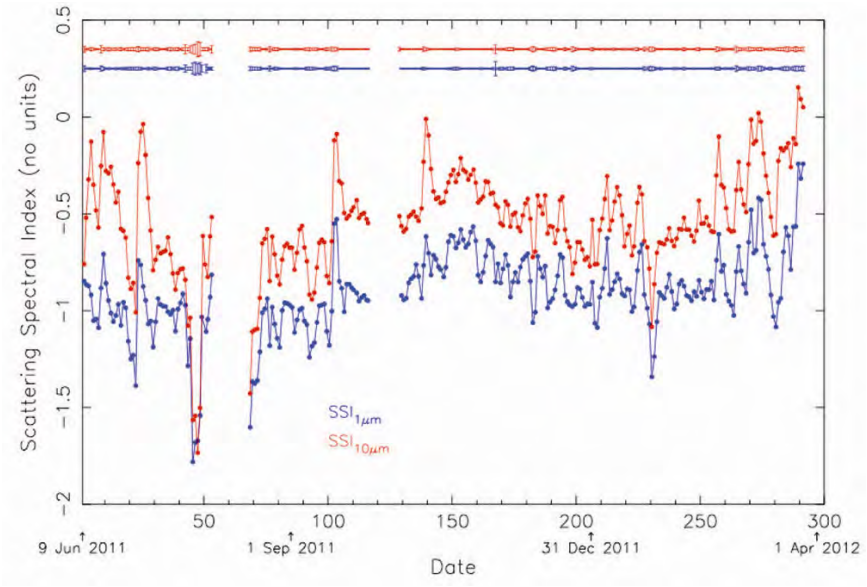

Figure 12. Time series of daily averaged values of $SSI_{10\ \mu m}$ and $SSI_{1\ \mu m}$. The blue and red bars at the top of the plot are the error bars ($\pm\sigma$) of daily averaged $SSI_{1\ \mu m}$ and $SSI_{10\ \mu m}$ respectively (ref. 146).

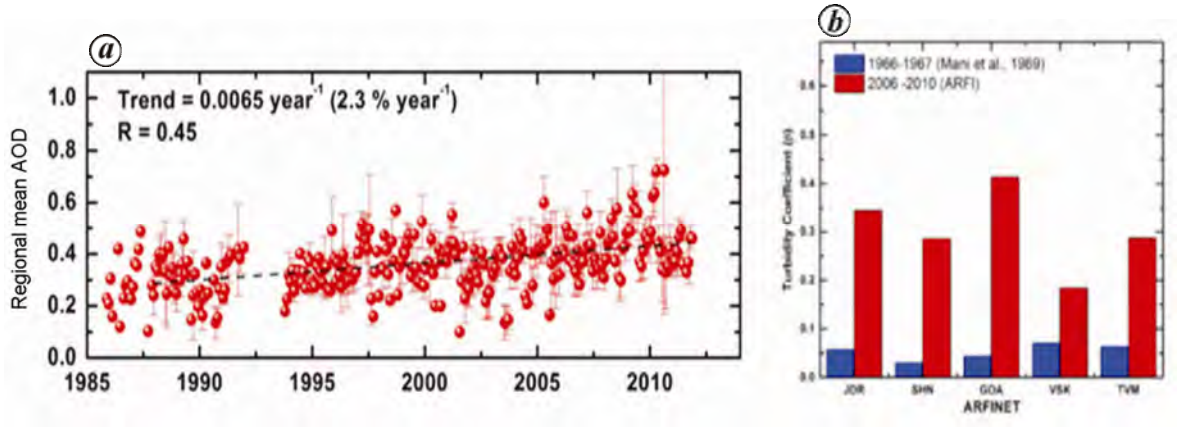

Figure 13. Long-term regional mean AOD (at 500 nm) depicting a steady increasing trend (***a***) and comparison of the present-day turbidity coefficients with historical values (more than 45 years back) over a few selected locations (***b***). The points in ***a*** are the monthly mean AOD, the vertical bars through them stand for the standard deviation. The gap in the figure for the period 1991–93 arises due to not using the data for that period, because of the strong perturbation to the AOD by the aerosols from Mount Pinatubo volcanic eruption of June 1991.

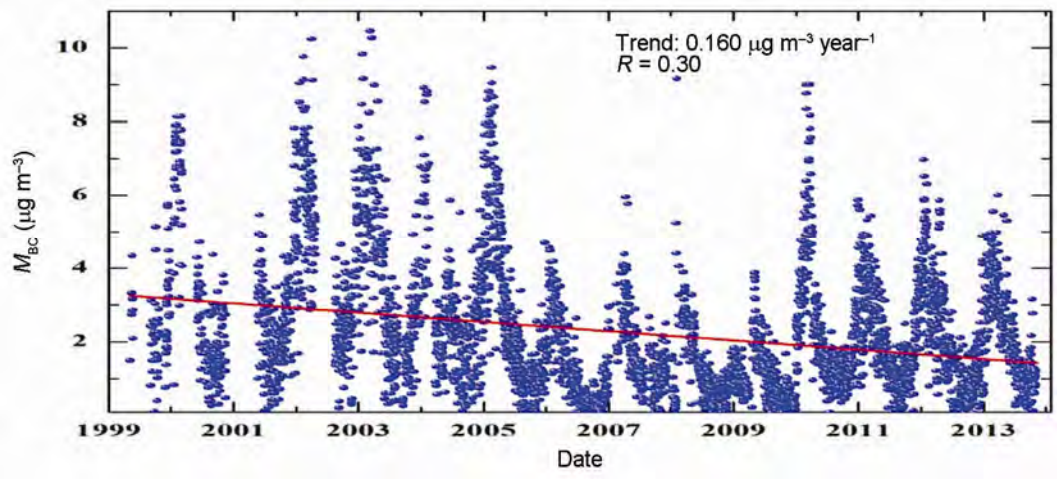

Figure 14. Long term trend in the near-surface BC mass concentration depicting a decreasing trend @160 ng m^{-3} $year^{-1}$ at the coastal station, Thiruvananthapuram, where the database is longest. The blue points are daily mean values of BC and the red line shows the trend. The oscillations are associated with annual variations; with a winter peak and monsoon minimum.

this increase is mostly due to the increase in the abundance of aerosols of anthropogenic origin. A few possible impact indicators, such as a decreasing trend in the number of rainy days during spring season, are also reported. Radiative transfer simulations reveal that by 2050 aerosol-induced surface solar dimming will increase from its current value of 14% to 32% and lower atmospheric warming from 0.5 to 1.4 K d^{-1}.

While the above results are of concern, both from environmental and climate perspectives, the analysis of long-term data of near-surface BC mass concentration – a tracer of anthropogenic emissions and a species strongly contributing to atmospheric warming – presents a very interesting contrast. Analysis of regular BC data from 12 of the ARFINET stations, where the data length has been longer than 5 years and included urban, semi-urban, remote, coastal, continental, island and high-altitude stations, has revealed a steady and consistent decreasing trend (at an average rate of ~100 ng m^{-3} $year^{-1}$) in all of them, except at Pune, where a very weak increasing trend was seen. A typical example is shown in Figure 14, for the station (TVM) having the longest data, where the concentration has been decreasing at an average rate of 160 ng m^{-3} superposed by the strong annual variations driven by synoptic meteorology. Analysing long-term satellite products, Girach and Nair[148] have reported a decreasing trend (of 1–2% per year) in the near-surface and columnar concentration of CO over the Indian region during 2000–2014. This further corroborates the above finding, considering that both BC and CO are normally cogents.

This result becomes all the more important when examined in the light of rapidly increasing energy production (from about 771.1 TWh in 2010–11 to 1048.7 TWh in 2014–15, with an average annual growth rate of 6.3%; a major part of it (~69% as on 2014–15) coming from fossil fuel fired thermal power plants) and rapidly increasing fossil fuel burning resulting from the exponential growth in the automobile sector (the total production soaring up from ~6.1 million vehicles of all types in 2004–05 to over 23 million in 2014–15 (the number of cars manufactured in India increasing from ~6500 per month in 1991 to >230000 per month in 2014; the number of passenger vehicles going up from 1.2 million in 2004–05 to 3.07 milliion in 2013–14 (ref. 149) with an average growth annual rate of 17% over the 2004–05 numbers) with two wheelers accounting for nearly 80% (source www.tradingeconomics.com). The consistency of the observed decrease in surface BC, despite the above increases in fuel use, is important and raises the following interesting possibilities, among others: (i) The fact that AOD is increasing over the years is a clear indicator of the increasing trend in the abundance of aerosols in the vertical column of the atmosphere. (ii) The decrease in trend in BC near-the surface, thus, would indicate combined effects of stringent emission control measures being adopted in the recent years and the increasing use of cleaner fuels. (iii) Increasing abundances of aerosols aloft (above the planetary boundary layer) which would not be reflected in the near surface concentrations. (iv) A slow, but steady change in the aerosol composition, with increased abundance of non-BC species (as also supported by the increase in aerosol SSA above the boundary layer (Figure 9)).

There have been sensational reports on the buildup of aerosols over Indian region and its global consequences; and also that India is one of the world's largest emitters of BC. The recent results, shown above, however, have shown a statistically significant increasing trend in total aerosol abundance and a simultaneous significant

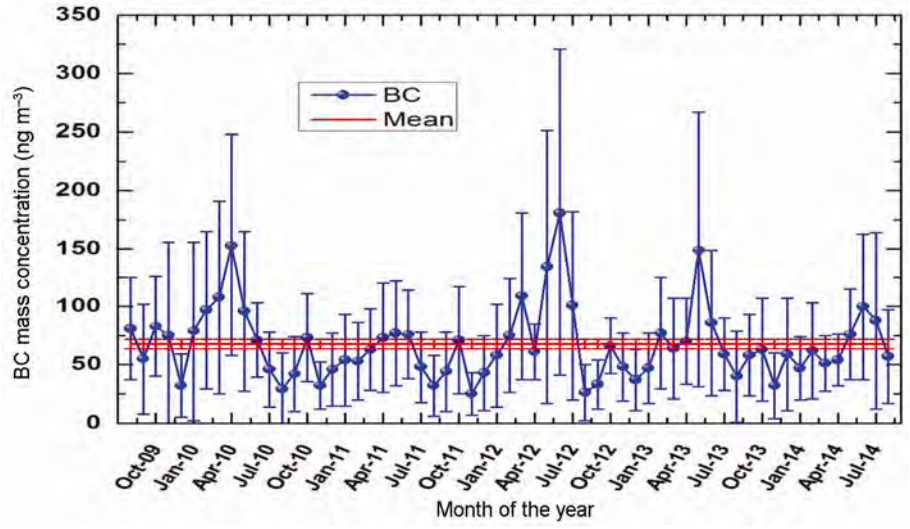

Figure 15. Long-term time series of BC concentration at Hanle (4500 m amsl) in the Trans-Himalayas; the points are monthly mean values and the vertical bars through them are the ensemble standard deviations showing the variations. The red lines are the long-term mean (middle line) and the standard errors (upper and lower lines).

decreasing trend in BC aerosols. The atmospheric warming effect of aerosols strongly depends on the relative contribution of absorbing aerosols. An aerosol system with high BC fraction would have net warming effect and complement the greenhouse warming. However, an increasing trend in composite (total) aerosols simultaneous with a decreasing trend in BC aerosols suggests a decreasing warming potential of aerosols. It is interesting that most of the impact studies concentrate only on the increasing trend in total aerosols, and tend to overlook the decrease in BC and hence the conclusions are likely to be unrealistic. For future projections of aerosol–climate impacts would be realistic only if all the above aspects are considered *in toto* and accounts for the gradually changing aerosol SSA due to change in the composition.

Trends at background sites (free troposphere)

With a view to monitoring long-term changes in the background aerosol abundance, a high-altitude aerosol observatory has been established at Hanle, in the western trans-Himalayas. Located at a very remote mountain peak at an altitude of >4.5 km amsl, this station provides continuous data on BC mass concentration and aerosol spectral optical depth since 2009 (ref. 150). In Figure 15, we have shown the time series of monthly mean BC mass concentration, measured at this station since its inception. It shows a few interesting facts: (i) The mean (long-term) BC concentration is low (~66 ng m^{-3}); still not so meagre as could be expected for such a remote free-tropospheric location. (ii) Strong seasonal variations, with a very conspicuous spring peak – a point that emerges consistently over the Indian region. This could be the result of complex processes involving the synoptic meteorology, boundary layer dynamics and long-range transport, besides the local sources. (iii) The amplitude of this spring peak, varies from year to year. (iv) No conspicuous trend is observed over the last 5-year period.

However, longer term measurements from such observatories would be providing the much needed vital information on possible aerosol-climate linkages over this region.

Aerosol–cloud processes

Cloud properties such as cloud-base height, liquid water path and thermodynamic profiles through the atmospheric column were measured at Nainital during the GVAX period; some of the results of this appear in this issue[151,152]. The summary of the results are shown in Figure 16. While the monsoon period is dominated by low-clouds, with the cloud-base very close to the surface and approximately corresponding to the PBL depth, the winter season has cloud-bases in the mid and upper troposphere and spring and pre-monsoon season is characterized by high cloud-bases, primarily in the 3–4 km range above the site ground level (Figure 16 *a*). Figure 16 *b* shows the liquid water path estimated from the MRP operated at Nainital.

During the same period, measurements of CCN at two super saturation ratios and total particles counts were also made using the CPC at the surface. The analyses of these data are still underway; though two significant summaries have been generated. Gogoi *et al.*[153] address the question 'What is the seasonality of aerosol optical properties and cloud condensation nuclei (CCN) activity and their mutual dependence under varying meteorological conditions

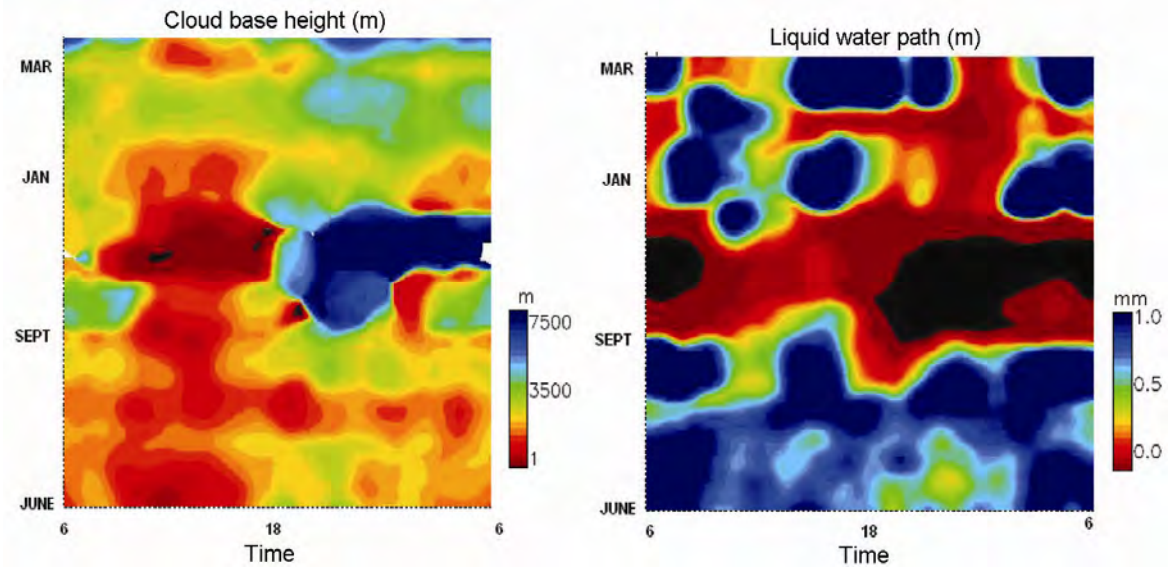

Figure 16. Cloud base heights measured at Nainital as a function of the time of the day (diurnal profile) and month of the year.

in the Indo-Gangetic plains'. They used the following data: (a) angular scattering and hemispheric backscattering coefficients at 450, 550 and 700 nm for particle sizes <1 μm and <10 μm as a function of ambient humidity conditions, measured with an integrating nephelometer, (b) Aerosol absorption coefficients at 467, 530 and 660 nm for particle sizes <1 μm and <10 μm size particles, measured by the particle soot absorption photometer, (c) Aerosol number density (#CN) for particle diameters of 10 nm to 3 μm, measured with a CN counter and (d) Cloud condensation nuclei number density (#CCN) at seven SS levels (0.2–1.0), measured with a CCN counter. Collocated measurements indicated enhanced aerosol scattering and absorption coefficients, as well as CCN and total condensation nuclei (CN) concentrations, during the dry seasons. In addition, during periods of high aerosol absorption (single-scattering albedo or SSA <0.80), the CCN concentration was higher than under relatively strongly scattering aerosol conditions (SSA >0.85), indicating seasonal changes in the aerosol composition and significant influence on CCN activity (Figure 17).

Furthermore, although fine-mode aerosols contributed to the activation of CN to CCN mostly during winter, coarse-mode aerosols assumed importance in CCN formation during monsoon and autumn. The monthly mean CCN activation ratio (irrespective of supersaturation) was highest (>0.6) in November and lowest (<0.3) in June. Comparison of these results with those from other parts of the globe indicates that CCN activity is more efficient at this high-altitude site, Nainital, than at several urban or polluted locations, but is comparable to or less efficient than the activity at a few other high-altitude or pristine locations.

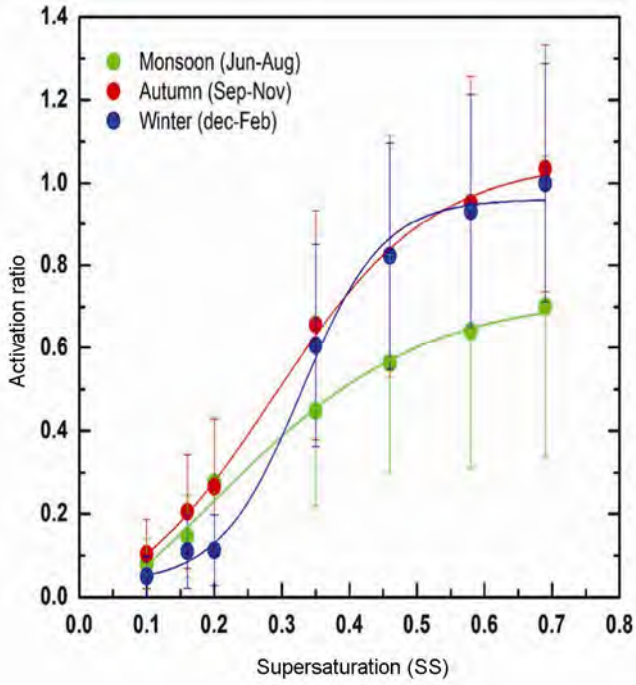

Figure 17. Variation of mean activation ratio at two distinct periods (JJA and DJFM) with supersaturation. The activation as a function of supersaturation is lower during the monsoon (JJA) season compared to the winter. This indicates that the aerosols observed during the summer are primarily dust or similar inactive aerosols and the winter aerosols are more chemically active and inclined to produce CCN.

Another study on these lines[154] reported strong diurnal variability in both the CCN and CN number densities and a diurnal dependence of AR. It also reported that the dependence of CCN on supersaturation is lowest during monsoon, in line with those reported by Gogoi *et al.*[153].

Way ahead

Having gone thus far, the most important question, the aerosol–climate community needs to address in near future is the role of aerosols in influencing cloud microphysical and radiative properties. For this purpose, a number of surface climate observatories, a series of field campaigns utilizing instrumented aircrafts and ships need to work in tandem; some of which are planned in the near future. Integrated experiments will be useful both in understanding the processes at work and providing answers regarding the effects of aerosols on climate in a region where the perturbation is probably the highest. Monsoons cover approximately 30% of the planet and are significant mechanisms for transferring energy and mass through the atmosphere from oceans to continental landmass. The Indian summer monsoon (ISM) rainfall has been remarkably stable over much of India over the past 100 years. Recently, there has been growing awareness of changes in precipitation since ISM rainfall patterns showed a trend of increasing extreme rainfall events over the Indian subcontinent. Modelling and simulation studies suggest intensification of monsoon rainfall and an accompanying increase in variability of the rainfall[69,141]. However, some of the recent analyses[155] indicate contradictory effects of aerosols and GHGs on monsoon-related rainfall over India. However, increasing temperatures due to GHGs might increase land–ocean temperature contrasts and hence strengthen the ISM circulation and precipitation. Thus, there is some evidence that anthropogenic aerosols impact monsoon and precipitation in South Asia. In summary, models have significant difficulty in simulating monsoons in the current climate and leads to considerable uncertainty in quantifying the role of aerosols in modifying the ISM. Such efforts also would throw more light on the 'self-lifting' of absorbing aerosols and stratospheric ozone as well as the question of whether absorbing aerosols enhance their residence time in the atmosphere by 'building their own home'[35]. With more integrated experiments, refining of models and a better synergy of measurements and models the current understanding of regional and global climate implications of aerosols would improve.

1. Andreae, M. O., Jones, C. D. and Cox, P. M., Strong present-day aerosol cooling implies a hot future. *Nature*, 2005, **435**, 1187–1190; doi: 10.1038/nature03671.
2. Satheesh, S. K. and Moorthy, K. K., Radiative effects of natural aerosols: a review. *Atm. Enviorn.*, 2005, **39**, 2089–2110.
3. Kaufman, Y. J. and Koren, I., Smoke and pollution aerosol effect on cloud cover. *Science*, 2006, **313**, 655–658; doi: 10.1126/science.1126232.
4. Bréon, F. M., Climate. How do aerosols affect cloudiness and climate. *Science*, 2006, **313**, 623–624; 10.1126/science.1131668.
5. Ramanathan, V. and Carmichael, G., Global and regional climate changes due to black carbon. *Nature Geosci.*, 2008, **1**, 221–227; doi: 10.1038/ngeo1562008.
6. Bollasina, M. A., Ming, Y. and Ramaswamy, V., Anthropogenic aerosols and weakening of the south Asian summer monsoon. *Science*, 2011, **334**, 502–505; 10.1126/science.1204994.
7. IPCC, Summary for policymakers. In *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change* (eds Stocker, T. F. *et al.*), Cambridge University Press, Cambridge, UK and New York, USA, 2013, p. 28.
8. Smith, S. J. and Bond, T. C., Two hundred fifty years of aerosols and climate: the end of the age of aerosols. *Atmos. Chem. Phys.*, 2014, **14**, 537–549; doi: 10.5194/acp-14-537-2014.
9. Di Girolamo, L. *et al.*, Analysis of multi-angle imaging spectroradiometer (MISR) aerosol optical depths over greater India during winter 2001–2004. *Geophys. Res. Lett*., 2004, **31**, doi: 10.1029/2004GL021273.
10. Dey, S. and Di Girolamo, L., A climatology of aerosol optical and microphysical properties over the Indian subcontinent from 9 years (2000–2008) of Multiangle Imaging Spectroradiometer (MISR) data. *J. Geophys. Res.*, 2010, **115**, D15204; doi: 10.1029/2009JD013395.
11. Babu, S. S. *et al.*, Trends in aerosol optical depth over Indian region: potential causes and impact indicators. *J. Geophys. Res.*, 2013, **118**, 1–13; doi: 10.1002/2013JD020507.
12. Badarinath, K. V. S., Sharma, A. R., Kaskaoutis, D. G., Kharol, S. K. and Kambezidis, H. D., Solar dimming over the tropical urban region of Hyderabad, India: effect of increased cloudiness and increased anthropogenic aerosols. *J. Geophys. Res.*, 2010, **115**, D21208; doi: 10.1029/2009JD013694.
13. Menon, S., James Hansen, J., Nazarenko, L. and Luo, Y., Climate effects of black carbon aerosols in China and India. *Science*, 2002, **297**, 2250–2253; doi: 10.1126/science.1075159.
14. Satheesh, S. K., Moorthy, K. K., Babu, S. S., Vinoj, V. and Dutt, C. B. S., Climate implications of large warming by elevated aerosol over India. *Geophys. Res. Lett.*, 2008, **35**, doi: 10.1029/2008GL034944.
15. Chinnam, N., Dey, S., Tripathi, S. N. and Sharma, M., Dust events in Kanpur, Northern India: chemical evidence for source and implications to radiative forcing. *Geophys. Res. Lett.*, 2006, **33**, L08803; doi: 10.1029/2005GL025278.
16. Meehl, G. A., Arblaster, J. M. and Tebaldi, C., Contributions of natural and anthropogenic forcing to changes in temperature extremes over the United States. *Geophys. Res. Lett.*, 2007, **34**, L19709; doi: 10.1029/2007GL030948.
17. Gautam, R., Hsu, N. C., Lau, K. M., Tsay, S. C. and Kafatos, M., Enhanced pre-monsoon warming over the Himalayan-Gangetic Region from 1979 to 2007. *Geophys. Res. Lett*., 2009, **36**, L07704; doi: 10.1029/2009GL037641.
18. Feng, Y., Kotamarthi, V. R., Coulter, R., Zhao, C. and Cadeddu, M., Radiative and thermodynamic responses to aerosol extinction profiles during the pre-monsoon month over South Asia. *Atm. Chem. Phys.*, 2016, **16**, 247–264; doi: 10.5194/acp-16-247-2016.
19. Mishra, S. K., Dey, S. and Tripathi, S. N., Implications of particle composition and shape to dust radiative effect: a case study from the Great Indian Desert. *Geophys. Res. Lett.*, 2008, **35**, doi: 10.1029/2008GL036058.
20. Sahai, S. *et al.*, A study for development of emission factors for trace gases and carbonaceous particulate species from *in situ* burning of wheat straw in agricultural fields in India. *Atm. Environ.*, 2007, **39**, 9173–9186; doi: 10.1016/j.atmosenv.2007.07.054.
21. Feng, Y., Ramanathan, V. and Kotamarthi, V. R., Brown carbon: a significant atmospheric absorber of solar radiation? *Atm. Chem. Phys*., 2013, **13**, 8607–8621.
22. Junge, C. E., *Air Chemistry and Radioactivity*, Academic Press, London, 1963, p. 382.
23. Mani, A., Chacko, O. and Hariharan, S., A study of Angstrom's turbidity parameters from solar radiation measurements in India. *Tellus*, 1969, **21**, 829–843.

24. Mani, A. and Huddar, B. B., Studies of surface aerosols and their effects on atmospheric electric parameters. *Pure Appl. Geophys.*, 1972, **100**, 154–166.
25. Mani, A. and Chacko, O., Attenuation of solar radiation in the atmosphere. *Sol. Ener.*, 1980, **24**, 347–349.
26. Moorthy, K. K., Nair, P. B. and Murthy, B. V. K., A study of aerosol optical depth at a coastal station, Trivandrum. *Indian J. Radio Space Phys.*, 1988, **17**, 16–22.
27. Moorthy, K. K. *et al.*, Results from the MWR network of IMAP. *Indian J. Radio Space Phys.*, 1993, **22**, 243–258.
28. Moorthy, K. K., Babu, S. S. and Satheesh, S. K., Temporal heterogeneity in aerosol characteristics and the resulting radiative impact at a tropical coastal station – Part 1: microphysical and optical properties. *Ann. Geophys.*, 2007, **25**, 2293–2308.
29. Murthy, B. V. K., Aerosols and radiation budget in the middle atmosphere. *Indian J. Radio Space Phys.*, 1988, **17**, 203–219.
30. Subbaraya, B. H., Jayaraman, A., Moorthy, K. K. and Mohan, M., Atmospheric aerosol studies under ISRO's Geosphere Biosphere Programme. *J. Indian Geophys. Un.*, 2000, **4**, 77–90.
31. Moorthy, K. K., Babu, S. S., Manoj, M. R. and Satheesh, S. K., Buildup of aerosols over the Indian Region. *Geophys. Res. Lett.*, 2013, **40**, 1011–1014; doi: 10.1002/grl.50165.
32. Moorthy, K. K., Satheesh, S. K., Babu, S. S. and Dutt, C. B. S., Integrated campaign for aerosols, gases and radiation budget (ICARB): an overview. *J. Earth Syst. Sci.*, 2008, **117(**S1), 243–262.
33. Moorthy, K. K. *et al.*, Optical and physical characteristics of Bay of Bengal aerosols during W-ICARB: spatial and vertical heterogeneities in the marine atmospheric boundary layer and in the vertical column. *J. Geophys. Res.*, 2010, **115**, D24213; doi: 10.1029/2010JD014094.
34. Jayaraman, A. and Subbaraya, B. H., *In situ* measurements of aerosol extinction profiles and their spectral dependencies at tropospheric levels. *Tellus B*, 1993, **45**, 473–478.
35. Babu, S. S. *et al.*, Free tropospheric black carbon aerosol measurements using high altitude balloon: Do BC layers build 'their own homes' up in the atmosphere? *Geophys. Res. Lett.*, 2011, **38**, L08803; doi: 10.1029/2011GL046654.
36. Parameswaran, K., Rose, K. O. and Murthy, B. V. K., Aerosol characteristics from bistatic lidar observations. *J. Geophys. Res.*, 1984, **89**, 2541–2552; doi: 10.1029/JD089iD02p02541.
37. Parameswaran, K., Rose, K. O. and Murthy, B. V. K., Relationship between backscattering and extinction coefficient of aerosols with application to turbid atmosphere. *Appl. Opt.*, 1991, **30**, 3059–3071; doi: 10.1364/AO.30.003059.
38. Devara, P. C. S. and Raj, E., Remote sensing of aerosols in the lower atmosphere using a bi-static helium-neon lidar. *J. Aerosol Sci.*, 1989, **20**, 37–44.
39. Devara, P. C. S., Raj, E., Sharma, S. and Pandithurai, G., Lidar-observed long-term variations in urban aerosol characteristics and their connection with meteorological parameters. *Int. J. Remote-sensing*, 1994, **14**, 581–591.
40. Satheesh, S. K., Vinoj, V. and Moorthy, K. K., Vertical distribution of aerosols over an urban continental site in India inferred using a micro pulse lidar. *Geophys. Res. Lett.*, 2006, **33**, L20816; doi: 10.1029/2006GL027729.
41. Rajeev, K., Ramanathan, V. and Meywerk, J., Regional aerosol distribution and its long-range transport over the Indian Ocean. *J. Geophys. Res.*, 2000, **105**, 2029–2043.
42. Jethva, H., Satheeh, S. K. and Srinivasan, J., Seasonal variability of aerosols over the Indo-Gangetic basin. *J. Geophys. Res.*, 2005, **110**; doi: 10.1029/2005JD005938.
43. Jethva, H., Satheeh, S. K. and Srinivasan, J., Assessment of second-generation MODIS aerosol retrieval (Collection 005) at Kanpur, India. *Geophys. Res. Lett.*, 2007, **34**; doi: 10.1029/2007GL029647.
44. Ramachandran, S. and Cherian, R., Regional and seasonal variations in aerosol optical characteristics and their frequency distributions over India during 2001–2005. *J. Geophys. Res.*, 2008, **113**; doi: 10.1029/2007JD008560.
45. Rengarajan, R., Sarin, M. M. and Sudheer, A. K., Carbonaceous and inorganic species in atmospheric aerosols during wintertime over urban and high-altitude sites in North India. *J. Geophys. Res.*, 2007, **112**, D21307; doi: 10.1029/2006JD008150.
46. Ram, K., Sarin, M. M. and Hegde, P., Atmospheric abundances of primary and secondary carbonaceous species at two high-altitude sites in India: sources and temporal variability. *Atm. Environ.*, 2008, **43**, 6785–6796; doi: 10.1016/j.atmosenv.2008.05.031.
47. Rastogi, N. and Sarin, M. M., Quantitative chemical composition and characteristics of aerosols over western India: one-year record of temporal variability. *Atm. Environ.*, 2009, **43**, 3481–3488; doi: 10.1016/j.atmosenv.2009.04.030.
48. Ganguly, D., Gadhavi, H., Jayaraman, A., Rajesh, T. A. and Misra, A., Single scattering albedo of aerosols over the central India: implications for the regional aerosol radiative forcing. *Geophys. Res. Lett.*, 2005, **32**, L18803; doi: 10.1029/2005GL023903.
49. Nair, V. S., *et al.*, Wintertime aerosol characteristics over the Indo-Gangetic Plain (IGP): impacts of local boundary layer processes and long-range transport. *J. Geophys. Res.*, 2007, **112**, D13205; doi: 10.1029/2006JD008099.
50. Sreekanth, V., Niranjan, K. and Madhavan, B. L., Radiative forcing of black carbon over eastern India. *Geophys. Res. Lett.*, 2007, **34**, L17818; doi: 10.1029/2007GL030377.
51. Niranjan, K., Sreekanth, V., Madhavan, B. L. and Moorthy, K. K., Aerosol physical properties and radiative forcing at the outflow region from the Indo-Gangetic plains during typical clear and hazy periods of wintertime. *Geophys. Res. Lett.*, 2007, **34**, L19805, doi: 10.1029/2007GL031224.
52. Kumar, A., Sarin, M. M. and Srinivas, B., Aerosol iron solubility over Bay of Bengal: Role of anthropogenic sources and chemical processing. *Mar. Chem.*, 2010, **121**, 167–175; doi: 10.1016/j.marchem.2010.04.005.
53. Moorthy, K. K., Nair, P. R. and Murthy, B. V. K., Size distribution of coastal aerosols: effects of local sources and sinks. *J. Appl. Meteorol.*, 1991, **30**, 844–852.
54. Moorthy, K. K., Murthy, B. V. K. and Nair, P. R., Sea-breeze front effects on boundary layer aerosols at a tropical coastal station. *J. Appl. Meteorol.*, 1993, **32**, 1196–1205.
55. Babu, S. S., Moorthy, K. K. and Satheesh, S. K., Temporal heterogeneity in aerosol characteristics and the resulting radiative impacts at a tropical coastal station – Part 2: direct short wave radiative forcing. *Ann. Geophys.*, 2007, **25**, 2309–2320.
56. Moorthy, K. K., Saha, A., Prasad, B. S. N., Niranjan, K., Jhurry, D. and Pillai, P. S., Aerosol optical depths over peninsular India and adjoining oceans during the INDOEX campaigns: spatial, temporal and spectral characteristics. *J. Geophys. Res.*, 2001, **106**, 28539–28554.
57. Moorthy, K. K., Babu, S. S. and Satheesh, S. K., Aerosol spectral optical depths over the Bay of Bengal: role of transport. *Geophys. Res. Lett.*, 2003, **30(**5), 1249, 53-1–53-4; doi: 10.1029/2002GL016520.
58. Parameswaran, K., Rose, K. O., Murthy, B. V. K., Osborn, M. T. and McMaster, L. R., Comparison of aerosol extinction profiles from lidar and SAGE II data at a tropical station. *J. Geophys. Res.*, 1991, **96**(D6), 10861–10866; doi: 10.1029/91JD01044.
59. Babu, S. S., Satheesh, S. K., Moorthy, K. K., Dutt, C. B. S., Nair, V. S., Alappattu, D. P. and Kunhikrishnan, P. K., Aircraft measurements of aerosol black carbon from a coastal location in the north-east part of peninsular India during ICARB. *J. Earth Syst. Sci.*, 2008, **117(**S1), 263–271.
60. Babu, S. S., Moorthy, K. K. and Satheesh, S. K., Vertical and horizontal gradients in aerosol black carbon and its mass fraction

to composite aerosols over the east coast of peninsular India from aircraft measurements. *Adv. Meteorol.*, 2010, Article ID 812075; doi: 10.1155/2010/812075.

61. Babu, S. S., Nair, V. S., Gogoi, M. M. and Moorthy, K. K., Seasonal variation of vertical distribution of aerosol single scattering albedo over Indian sub-continent: RAWEX aircraft observations. *Atmos. Environ.*, 2015; doi: 10.1016/j.atmosenv.2015.09.041.
62. Prijith, S. S., Babu, S. S., Lakshmi, N. B., Satheesh, S. K. and Moorthy, K. K., Meridional gradients in aerosol vertical distribution over Indian Mainland: observations and model simulations. *Atmos. Environ.*, 2016, **125B**, 37–345; doi: 10.1016/j.atmosenv.2015.10.066.
63. Moorthy, K. K. *et al.*, Fine and ultrafine particles at a near-free tropospheric environment over the high-altitude station Hanle in the Trans-Himalaya: new particle formation and size distribution. *J. Geophys. Res.*, 2011, **116**, D20212; doi: 10.1029/2011JD016343.
64. Satheesh, S. K., Ramanathan, V., Li-Jones, X., Prospero, J. M., Holben, B. N. and Loeb, N. G., A model for the natural and anthropogenic aerosols over the tropical Indian Ocean derived from Indian Ocean Experiment data. *J. Geophys. Res.*, 1999, **104**, 27,421–27,440.
65. Babu, S. S., Satheesh, S. K. and Moorthy, K. K., Aerosol radiative forcing due to enhanced black carbon at an urban site in India. *Geophys. Res. Lett.*, 2002, **29**, doi: 10.1029/2002GL015179.
66. Moorthy, K. K., Nair, V. S., Babu, S. S. and Satheesh, S. K., Spatial and vertical heterogeneities in aerosol properties over oceanic regions around India: implications for radiative forcing. *Q. J. R. Meteorol. Soc.*, 2009, **135**, 2131–2145; doi: 10.1002/qj.525.
67. Satheesh, S. K. and Ramanathan, V., Large differences in tropical aerosol forcing at the top of the atmosphere and Earth's surface. *Nature*, 2000, **405**, 60–63; doi: 10.1038/35011039.
68. Ramanathan, V. *et al.*, Atmospheric brown clouds: impacts on south Asian climate and hydrological cycle, *PNAS*, 2005, **102**(15), 5326–5333; doi: 10.1073/pnas.0500656102.
69. Lau, K. M., Kim, M. K. and Kim, K. M., Asian summer monsoon anomalies induced by aerosol direct forcing: the role of the Tibetan Plateau. *Climate Dyn.*, 2006, **26**, 855–864.
70. Nair, V. S., Babu, S. S., Moorthy, K. K., Sharma, A. K., Marinoni, A. and Ajai, Black carbon aerosols over the Himalayas: direct and surface albedo forcing. *Tellus B*, 2013, **65**; doi: 10.3402/tellusb.v65i0.19738.
71. Nair, V. S., Babu, S. S., Moorthy, K. K. and Prijith, S. S., Spatial gradients in aerosol-induced atmospheric heating and surface dimming over the oceanic regions around India: anthropogenic or natural? *J. Climate*, 2013, **26**, 7611–7621; doi: 10.1175/jcli-d-12-00616.
72. Satheesh, S. K., Moorthy, K. K. and Srinivasan, J., New directions: elevated layers of anthropogenic aerosols aggravate stratospheric ozone loss? *Atm. Environ.*, 2013, **79**, 879–882; doi: 10.1016/j.atmosenv.2013.07.052.
73. Deepshikha, S., Satheesh, S. K. and Srinivasan, J., Regional distribution of absorbing efficiency of dust aerosols over India and adjacent continents inferred using satellite remote sensing. *Geophys. Res. Lett.*, 2005, **32**; doi: 10.1029/2004GL022091.
74. Deepshikha, S., Satheesh, S. K. and Srinivasan, J., Dust aerosols over India and adjacent continents retrieved using METEOSAT infrared radiance Part I: sources and regional distribution. *Ann. Geophys.*, 2006, **24**, 37–61.
75. Deepshikha, S., Satheesh, S. K. and Srinivasan, J., Dust aerosols over India and adjacent continents retrieved using METEOSAT infrared radiance Part II: quantification of wind dependence and estimation of radiative forcing. *Ann. Geophys.*, 2006, **24**, 63–79.
76. Moorthy, K. K., Babu, S. S., Satheesh, S. K., Srinivasan, J. and Dutt, C. B. S., Dust absorption over the 'Great Indian Desert' inferred using ground-based and satellite remote sensing. *J. Geophys. Res.*, 2007, **112**, D09206, doi: 10.1029/2006JD007690.
77. Srinivas, B., Sarin, M. M. and Rengarajan, R., Atmospheric transport of mineral dust from the Indo-Gangetic Plain: temporal variability, acid processing, and iron solubility. *Geochem. Geophys. Geosyst.*, 2014, **15**; doi: 10.1002/2014GC005395.
78. Chandra, S., Satheesh, S. K. and Srinivasan, J., Can the state of mixing of black carbon aerosols explain the mystery of 'excess' atmospheric absorption? *Geophys. Res. Lett.*, 2004, **31**, L19109, doi: 10.1029/2004GL020662.
79. Dumka, U. C., Satheesh, S. K., Pant, P., Hegde, P. and Moorthy, K. K., Surface changes in solar irradiance due to aerosols over central Himalayas. *Geophys. Res. Lett.*, 2006, **33**, L20809; doi: 10.1029/2006GL027814.
80. Singh, R. P., Dey, S., Tripathi, S. N., Tare, V. and Holben, B., Variability of aerosol parameters over Kanpur, northern India. *J. Geophys. Res.*, 2004, **109**, D23206; doi: 10.1029/2004JD004966.
81. Tripathi, S. N., Dey, S., Tare, V. and Satheesh, S. K., Aerosol black carbon radiative forcing at an industrial city in northern India. *Geophys. Res. Lett.*, 2005, **32**, L08802; doi: 10.1029/2005GL022515.
82. Srinivas, B. and Sarin, M. M., Light absorbing organic aerosols (brown carbon) over the tropical Indian Ocean: impact of biomass burning emissions. *Environ. Res. Lett.*, 2013, **8**, 044042; doi: 10.1088/1748-9326/8/4/044042.
83. Moorthy, K. K., Satheesh, S. K. and Murthy, B. V. K., Investigations of marine aerosols over the tropical Indian Ocean. *J. Geophys. Res.*, 1997, **102**, 18,827–18,842.
84. Babu, S. S., Moorthy, K. K. and Satheesh, S. K., Aerosol black carbon over Arabian Sea during inter monsoon and summer monsoon seasons. *Geophys. Res. Lett.*, 2004, **31**, L06104; 10.1029/2003GL018716.
85. Moorthy, K. K. *et al.*, Wintertime spatial characteristics of boundary layer aerosols over Peninsular India. *J. Geophys. Res.*, 2005, **110**, D08207; doi: 10.1029/2004JD005520.
86. Lawrence, M. G. and Lelieveld, J., Atmospheric pollutant outflow from southern Asia: a review. *Atm. Chem. Phys.*, 2010, 11017–11096; doi: 10.5194/acp-10-11017-2010.
87. Nair, V. S., Satheesh, S. K., Moorthy, K. K., Babu, S. S., Nair, P. R. and George, S. K., Surprising observation of large anthropogenic aerosol fraction over the 'near pristine' southern Bay of Bengal: climate implications. *J. Geophys. Res.*, 2010, **115**, doi: 10.1029/2010JD013954.
88. Babu, S. S., Sreekanth, V., Nair, V. S., Satheesh, S. K. and Moorthy, K. K., Vertical profile of aerosol single scattering albedo over west coast of India during W_ICARB. *J. Atmos. Sol. Terr. Phys.*, 2010, **72**, 876–882; doi: 10.1016/j.jastp. 2010.04.013.
89. Sinha, P. R., Manchanda, R. K., Subbarao, J. V., Dumka, U. C., Sreenivasan, S., Babu, S. S. and Moorthy, K. K., Spatial distribution and vertical structure of the MABL aerosols over Bay of Bengal during winter: results from W-ICARB experiment. *J. Atmos. Solar Terr. Phys.*, 2011, **73**, 430–438.
90. Nair, V. S., Moorthy, K. K. and Babu, S. S., Influence of continental outflow and ocean bio-geochemistry on the distribution of fine and ultra-fine particles in the marine atmospheric boundary layer over Arabian Sea and Bay of Bengal. *J. Geophys. Res.*, 2013, **118**, 7321–7331; doi: 10.1002/jgrd.50541.
91. Khemani, L. T. and Murty, Bh. V. R., Chemical composition of rain water and rain characteristics at Delhi. *Tellus*, 1968, **20**, 284–292.
92. Khemani, L. T., Momin, G. A., Naik, M. S., Vijayakumar, R. and Murty, Bh. V. R., Chemical composition and size distribution of atmospheric aerosols over the Deccan Plateau, India. *Tellus*, 1982, **34**, 151–158.
93. Khemani, L. T., Momin, G. A., Naik, M. S., Rao, P. S. P., Kumar, R. and Murty, Bh. V. R., Trace elements and sea salt aerosols over the sea areas around the Indian sub-continent. *Atmos. Environ*, 1985, **19**, 277–284; doi: 10.1016/0004-6981(85)90095-2.
94. Ball, W. P., Dickerson, R. R., Doddridge, B. G., Stehr, J. W., Miller, T. L., Savoie, D. L. and Carsey, T. P., Bulk and size

segregated aerosol composition observed during INDOEX 1999: overview of meteorology and continental impacts. *J. Geophys. Res.*, 2003, **108**, 8001; doi: 10.1029/2002JD002467.
95. Mayol-Bracero, O. L. *et al.*, Carbonaceous aerosols over the Indian Ocean during the Indian Ocean experiment (INDOEX): chemical characterization, optical properties, and probable sources. *J. Geophys. Res.*, 2002, **107**, 8030; doi: 10.1029/2000JD000039.
96. Podgorny, I. A., Conant, W., Ramanathan, V. and Satheesh, S. K., Aerosol modulation of atmospheric and surface solar heating over the tropical Indian Ocean. *Tellus B*, 2000, **52**, 947–958; doi: 10.1034/j.1600-0889.2000.
97. Moorthy, K. K., Niranjan, K., Narasimhamurthy, B., Agashe, V. V. and Murthy, B. V. K., Spatial and temporal characteristics of aerosol spectral optical depths over tropical Indian regions–1: The network and database, Scientific report, ISRO:GBP:SR: 03:99, 1999.
98. Babu, S. S. and Moorthy, K. K., Aerosol black carbon over a tropical coastal station in India. *Geophys. Res. Lett.*, 2002, **29**(23), 2098; doi: 10.1029/2002GL015662.
99. Moorthy, K. K. *et al.*, Performance evaluation of chemistry transport models over India. *Atmos. Environ.*, 2013, **71**, 210–225; dx.doi.org/10.1016/j.atmosenv.2013.01.056.
100. George, S. K., Nair, P. R., Parameswaran, K. and Jacob, S., Wintertime chemical composition of aerosols at a rural location in the Indo-Gangetic Plains. *J. Atmos. Sol. Terr. Phys.*, 2011, **73**; doi: 10.1016/j.jastp.2011.04.005.
101. Srinivas, B. and Sarin, M. M., Atmospheric pathways of phosphorous to the Bay of Bengal: contribution from anthropogenic sources and mineral dust. *Tellus B*, 2012, **64**, 17174; http://dx.doi.org/10.3402/tellusb.v64i0.17174.
102. Srinivas, B. and Sarin, M. M., Atmospheric dry-deposition of mineral dust and anthropogenic trace metals to the Bay of Bengal. *J. Mar. Syst.*, 2013, **126**, 56–68.
103. Rajput, P., Sarin, M. M., Sharma, D. and Singh, D., Organic aerosols and inorganic species from postharvest agricultural-waste burning emissions over northern India: impact on mass absorption efficiency of elemental carbon. *Environ. Sci.: Processes Impacts*, 2014, **16**, 2371–2379.
104. Srinivas, B. and Sarin, M. M., Brown carbon in atmospheric outflow from the Indo-Gangetic Plain: mass absorption efficiency and temporal variability. *Atmos. Environ.*, 2014, **89**, 835–843.
105. Srivastava, A. K., Ram, K., Pant, P., Hegde, P. and Joshi, H., Black carbon aerosols over Manora Peak in the Indian Himalayan foothills: implications for climate forcing. *Environ. Res. Lett.*, 2012, **7**, 014002 (8pp); doi: 10.1088/1748-9326/7/1/014002.
106. Aryasree, S., Nair, P. R., Girach, I. A. and Jacob, S., Winter time chemical characteristics of aerosols over the Bay of Bengal: continental influence. *Environ. Sci. Poll. Res.*, 2015, **22**, 14901–14918; doi: 10.1007/s11356-015-4700-7.
107. Pant, P., Hegde, P., Dumka, U. C., Sagar, R., Satheesh, S. K. and Moorthy, K. K., Aerosol characteristics at a high altitude location in Central Himalayas: optical properties and radiative forcing. *J. Geophys. Res.*, 2006, **111**, D17206; 10.1029/2005JD006768.
108. Niranjan, K., Sreekanth, V., Madhavan, B. L. and Moorthy, K. K., Wintertime aerosol characteristics at a north Indian site Kharagpur in the Indo-Gangetic plains located at the outflow region into Bay of Bengal. *J. Geophys. Res.*, 2006, **111**, D24209; doi: 10.1029/2006JD007635.
109. Kulshrestha, U. C., Saxena, A., Kumar, N., Kumari, K. M. and Srivastava, S. S., Chemical composition and association of size-differentiated aerosols at a Suburban Site in a semi-arid tract of India. *J. Atmos. Chem.*, 1998, **29**, 109–118.
110. Kulshrestha, A., Satsangi, P. G., Masih, J. and Taneja, A., Metal concentration of PM2.5 and PM10 particles and seasonal variations in urban and rural environment of Agra, India. *Sci. Tot. Environ.*, 2009, **407**, 6196–6204; doi: 10.1016/j.scitotenv.2009.08.050.
111. Ram, K. and Sarin, M. M., Day-night variability of EC, OC, WSOC and inorganic ions in urban environment of Indo-Gangetic Plain: implications to secondary aerosol formation. *Atmos. Environ.*, 2011, **45**, 460–468; doi: 10.1016/j.atmosenv.2010.09.055.
112. Behera, S. N. and Sharma, M., Reconstructing primary and secondary components of PM2.5 composition for an urban atmosphere. *Aerosol. Sci. Technol.*, 2010, **44**, 983–992; doi: 10.1080/02786826.2010.504245.
113. Ram, K., Sarin, M. M. and Tripathi, S. N., A 1-year record of carbonaceous aerosols from an urban site in the Indo-Gangetic Plain: characterization, sources, and temporal variability. *J. Geophys. Res.*, 2010, **115**, D24313; doi: 10.1029/2010JD014188.
114. Gogoi, M. M., Moorthy, K. K., Babu, S. S. and Bhuyan, P. K., Climatology of columnar aerosol properties and the influence of synoptic conditions: first-time results from the northeastern region of India. *J. Geophys. Res.*, 2009, **114**, D08202, doi: 10.1029/2008JD010765.
115. Kumar, K. R. *et al.*, Temporal and spectral characteristics of aerosol optical depths in a semi-arid region of southern India. *Sci. Tot. Environ.*, 2009, **407**, 2673–2688.
116. Gopal, K. R. *et al.*, *In situ* measurements of atmospheric aerosols by using Integrating Nephelometer over a semi-arid station, southern India. *Atmos. Environ.*, 2014, **86**, 228–240; doi: 10.1016/j.atmosenv.2013.12.009.
117. Ramachandran, S. and Kedia, S., Radiative effects of aerosols over Indo-Gangetic plain: environmental (urban vs rural) and seasonal variations. *Environ. Sci. Poll. Res.*, 2012, **19**, 2159–2217; doi: 10.1007/s11356-011-0715-x.
118. Ramanathan, V. *et al.*, Indian Ocean experiment: an integrated analysis of the climate forcing and effects of the great Indo-Asian haze. *J. Geophys. Res.*, 2001, **106**, 28,371–28,398.
119. Beegum, S. N. *et al.*, Characteristics of spectral aerosol optical depths over India during ICARB. *J. Earth Sys. Sci.*, 2008, **117(**S1), 303–313.
120. Beegum, S. N. *et al.*, Spatial distribution of aerosol black carbon over India during pre-monsoon season. *Atmos. Environ.*, 2009, **43**, 1071–1078.
121. Jethva, H., Satheesh, S. K., Srinivasan, J. and Moorthy, K. K., How good is the assumption of visible surface reflectance in MODIS aerosol retrieval over land? A comparison with aircraft measurements over an urban site in India. *IEEE Trans. Geosci. Rem. Sensing*, 2009, **47**, 1990–1998.
122. Moorthy, K. K., Babu, S. S. and Satheesh, S. K., Aerosol characteristics and radiative impacts over the Arabian Sea during the Intermonsoon season: results from the ARMEX field campaign. *J. Atmos. Sci.*, 2005, **62**(1), 192–206.
123. Prasad, A. K., Singh, S., Chauhan, S. S., Srivastava, M. K., Singh, R. P. and Singh, R., Aerosol radiative forcing over the Indo-Gangetic plains during major dust storms. *Atmos. Environ.*, 2007, **4**, 6289–6301; doi: 10.1016/j.atmosenv.2007.03.060.
124. Habib, G., Venkataraman, C., Chiapello, I., Ramachandran, S., Boucher, O. and Reddy, M. S., Seasonal and interannual variability in absorbing aerosols over India derived from TOMS: relationship to regional meteorology and emissions. *Atm. Environ.*, 2006, **40**, 1909–192; doi: 10.1016/j.atmosenv.2005.07.077.
125. Pathak, B. *et al.*, Aerosol characteristics in north–east India using ARFINET spectral optical depth measurements. *Atmos. Environ. B*, 2016, **125**, 461–473; doi: 10.1016/j.atmosenv.2015.07.038.
126. Nair, V. S., Solmon, F., Giorgi, F., Mariotti, L., Babu, S. S. and Moorthy, K. K., Simulation of South Asian aerosols for regional climate studies. *J. Geophys. Res.*, 2012, **117**, D04209; doi: 10.1029/2011JD016711.
127. Govardhan, G., Nanjundiah, R. S., Satheesh, S. K., Moorthy, K. K. and Kotamarthi, V. R., Performance of WRF-Chem over

Indian region: comparison with measurements. *J. Earth Syst. Sci.*, 2015, **124**, 875–896.
128. Pan, X. *et al.*, A multi-model evaluation of aerosols over South Asia: common problems and possible causes. *Atmos. Chem. Phys.*, 2015, **15**, 5903–5928; doi: 10.5194/acp-15-5903-2015.
129. Chin, M., Rood, R. B., Lin, S. J., Muller, J. F. and Thompson, A. M., Atmospheric sulfur cycle simulated in the global model GOCART: model description and global properties. *J. Geophys. Res.*, 2000, **105**, 24671–24687.
130. Menut, L. *et al.*, CHIMERE 2013: a model for regional atmospheric composition modeling. *Geosci. Model Dev.*, 2013, **6**, 981–1028; doi: 10.5194/gmd-6-981-2013.
131. Skamarock, W. *et al.*, A description of the advanced research WRF version 3. NCAR technical note NCAR/TN-475+STR, 2008; doi: 10.5065/D68S4MVH.
132. Takemura, T., Nakagima, T., Higurashi, A., Ohta, S. and Sugimoto, N., Aerosol distributions and radiative forcing over the Asian Pacific region simulated by Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS). *J. Geophys. Res.*, 2003, **108**, 8659; doi: 10.1029/2002JD003210.
133. Takemura, T., Egashira, M., Matsuzawa, K., Ichijo, H., O'ishi, R. and Abe-Ouchi, A., A simulation of the global distribution and radiative forcing of soil dust aerosols at the Last Glacial Maximum. *Atmos. Chem. Phys.*, 2009, **9**, 3061–3073; doi: 10.5194/acp-9-3061-2009.
134. Emmons, L. K. *et al.*, Description and evaluation of the model for ozone and related chemical tracers, version 4 (MOZART-4). *Geosci. Model Dev.*, 2010, **3**, 43–67.
135. Robertson, L., Langner, J. and Engardt, M., An Eulerian limited-area atmospheric transport model. *J. Appl. Meteorol.*, 1999, **38**, 190–210.
136. He, C. *et al.*, A global 3-D CTM evaluation of black carbon in the Tibetan Plateau. *Atmos. Chem. Phys.*, 2014, **14**, 7091–7112; doi: 10.5194/acp-14-7091-2014.
137. Kumar, R. *et al.*, Sources of black carbon aerosols in South Asia and surrounding regions during the integrated campaign for aerosols, gases and radiation budget (ICARB). *Atmos. Chem. Phys.*, 2015; doi: 10.5194/acp-15-5415-2015.
138. Hu, Z. Z., Latif, M., Roeckner, E. and Bengtsso, L., Intensified Asian summer monsoon and its variability in a coupled model forced by increasing greenhouse gas concentrations. *Geophys. Res. Lett.*, 2000, **27**, 2681–2684; 0094-8276/00/2000GL-011550505.
139. Ganguly, D., Rasch, P. J., Wang, H. and Yoon, J., Fast and slow responses of the South Asian monsoon system to anthropogenic aerosols. *Geophys. Res. Lett.*, 2012; **39**, doi: 10.1029/2012-GL053043.
140. Rosenfeld, D. *et al.*, Flood or drought: how do aerosols affect precipitation? *Science*, **321**, 1309–1313; doi: 10.1126/science. 1160606.
141. Goswami, B. N., Venugopal, V., Sengupta, D., Madusoodanan, M. S. and Xavier, P. K., Increasing trend of extreme rain events over India in a warming environment. *Science*, 2006, **314**, 1442–1445.
142. Yadav, S. and Rajamani, V., Geochemistry of aerosols of the northwestern part of India adjoining the Thar Desert. *Geochim. Cosmochim. Acta*, 2004, **68**, 1975–1988.
143. de Laat, A. T. J., Zweers, S. D. C., Boers, R. and Tuinder, O. N. E., A solar escalator: observational evidence of the self-lifting of smoke and aerosols by absorption of solar radiation in the February 2009 Australian Black Saturday plume. *J. Geophys. Res.*, 2012, **117**, D04204.
144. Prijith, S. S., Babu, S. S., Lakshmi, N. B., Satheesh, S. K. and Moorthy, K. K, Meridional gradients in aerosol vertical distribution over Indian Mainland: observations and model simulations. *Atmos. Environ. B*, 2016, **125**, 337–345; doi: 10.1016/j.atmosenv. 2015.10.066.
145. Renju, R., Raju, C. S., Mathew, N., Antony, T. and Moorthy, K. K., Microwave radiometer observations of interannual water vapor variability and vertical structure over a tropical station. *J. Geophys. Res. Atmos.*, 2015, **120**, 4585–4599; doi: 10.1002/ 2014JD022838.
146. Manoharan, V. S. *et al.*, Increased absorption by coarse aerosol particles over the Gangetic–Himalayan region. *Atmos. Chem. Phys.*, 2014, **14**, 1159–1165; doi: 10.5194/acp-14-1159-2014.
147. Dumka, U. C. and Kaskaoutis, D. G., *In situ* measurements of aerosol properties and estimates of radiative forcing efficiency over Gangetic-Himalayan region during the GVAX field campaign. *Atm. Environ.*, 2014, **94**, 96–105; doi: 10.1016/j.atmosenv. 2014.05.021.
148. Girach, I. A. and Nair, P. R., Carbon monoxide over India as observed by MOPPIT. *Atmos. Environ.*, 2015, **99**, 599–609; doi: 10.1016/j.atmosenv.2014.10.019.
149. Krishnaveni, M. and Vaidya, R., Growth of Indian automobile industry. *Int. J. Curr. Res. Aca. Rev.*, 2015, **3**(2), 110–118.
150. Gogoi, M. M. *et al.*, Physical and optical properties of aerosols in a free tropospheric environment: results from long-term observations over western trans-Himalayas. *Atmos. Environ.*, 2014, **84**, 262–274; http://dx.doi.org/10.1016/j.atmosenv.2013.11.029.
151. Singh, N. *et al.*, Variations in the cloud base height over the Central Himalayas during the GVAX: association with the monsoon rainfall. *Curr. Sci.*, 2016, **111**(1), 109–116.
152. Naja, M. *et al.*, High frequency vertical profiling of meteorological parameters using AMF1 facility during RAWEX–GVAX at ARIES, Nainital, *Curr. Sci.*, 2016, **111**(1), 132–140.
153. Gogoi, M. M., Babu, S. S., Jayachandran, V., Moorthy, K. K., Satheesh, S. K., Naja, M. and Kotamarthi, V. R., Optical properties and CCN activity of aerosols in a high-altitude Himalayan environment: results from RAWEX–GVAX. *J Geophys. Res.*, 2015, **120**, doi: 10.1002/2014JD022966.
154. Dumka, U. C., Kaskaoutis, D. G., Srivastava, M. K. and Devara, P. C. S., Scattering and absorption properties of near-surface aerosol over Gangetic–Himalayan region: the role of boundary-layer dynamics and long-range transport. *Atmos. Chem. Phys.*, 2015; **15**, 1555–1572; doi: 10.5194/acp-15-1555-2015.
155. Chung, C. E. and Ramanathan, V., Weakening of North Indian SST gradients and the monsoon rainfall in India and the Sahel. *J. Clim.*, 2006, **19**, 2306–2405; doi: 10.1175/JCLI3820.1.

doi: 10.18520/cs/v111/i1/53-75