Atmospheric Environment 80 (2013) 107–121

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

In situ measurements of aerosol vertical and spatial distributions over continental India during the major drought year 2009

B. Padmakumari*, R.S. Maheskumar [1], G. Harikishan [1], S.B. Morwal [1], T.V. Prabha [1], J.R. Kulkarni [1]

Indian Institute of Tropical Meteorology, Pune 411 008, India

HIGHLIGHTS

- Aerosol vertical and spatial distribution in monsoon environment over Indian sub continent.
- Dominance of fine and coarse mode aerosol and strong vertical gradient during monsoon.
- Surface level concentration and height of ABL influence the aerosol optical depth.

ARTICLE INFO

Article history:
Received 31 October 2012
Received in revised form
22 July 2013
Accepted 26 July 2013

Keywords:
CAIPEEX
Elevated pollution layers
Fine mode aerosol
Coarse mode aerosol
Drought year
Aerosol types

ABSTRACT

The variability in aerosol vertical and spatial distribution over the continental Indian region is studied using the airborne observations during the Cloud Aerosol Interactions and Precipitation Enhancement EXperiment (CAIPEEX) from May to September, 2009. The fine mode (0.1–3.0 μm) aerosol vertical profiles up to 6 km at different regions showed different vertical structures mostly influenced by the atmospheric boundary layer (ABL) depth as well as the origin of air mass trajectories and the presence of clouds. Elevated aerosol layers are observed during pre-monsoon and during monsoon at some locations but comparatively lower than the one observed in the boundary layer. During monsoon, aerosol number concentration showed strong vertical gradient and a transition is observed between the boundary layer and the free troposphere. The coarse mode (>3 μm) aerosol vertical profiles also showed elevated layers at higher altitudes due to the incursion of dry air laddened with dust. The spatial distribution shows significant variation at the elevated layers as compared to that in the boundary layer during pre-monsoon, while high variability is observed in the boundary layer during monsoon. The frequency distribution of different aerosol types from Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) showed dominating contributions from dust, polluted dust and smoke during pre-monsoon. During monsoon also traces of these pollutants were found to be high as the year 2009 is a drought year with rainfall deficiency of 22%. The surface level number concentration and the height of ABL are found to influence the aerosol optical depths significantly.

1. Introduction

Atmospheric aerosols play a major role in climate change by directly scattering and absorbing the incoming and outgoing radiation and through modifying cloud properties, such as droplet size distribution (Twomey, 1974) and cloud lifetime and extent (Albrecht, 1989; Ackerman et al., 2000). Indian sub continent has anthropogenically generated pollutants and dust from local sources as well as from long range transport (Ramanathan et al., 2001; Dey et al., 2004; Moorthy et al., 2008; Kaskaoutis et al., 2012). Fine mode aerosol dominance is observed during winter and coarse mode aerosol dominance during pre-monsoon (Gautam et al., 2007). As compared to aerosol optical depths, aerosol vertical profile retrievals give more insight on the aerosol impacts on climate change such as aerosol warming on the thermal structure and stability of the atmosphere, because of the presence of distinct aerosol layers aloft called elevated layers that are consistently observed with various observational techniques (Ramana et al., 2004; Raj et al., 2008; Babu et al., 2011; Prabha et al., 2012; Padmakumari et al.,

* Corresponding author. Tel.: +91 20 2590 4349; fax: +91 20 2589 3825.
E-mail addresses: padma@tropmet.res.in (B. Padmakumari), mahesh@tropmet.res.in (R.S. Maheskumar), harikishan@tropmet.res.in (G. Harikishan), morwal@tropmet.res.in (S.B. Morwal), thara@tropmet.res.in (T.V. Prabha), jrksup@tropmet.res.in (J.R. Kulkarni).
[1] Tel.: +91 20 2590 4349; fax : +91 20 2589 3825.

1352-2310/$ – see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.atmosenv.2013.07.064

2013). Also the vertical distribution of aerosol determines the degree with which it would interact with clouds, influence air quality and the atmospheric heating profiles. The aerosol in the ABL is often local in origin, while aerosol in elevated layers is usually transported from large distances. Hence, the physical, chemical and optical properties of aerosol in the ABL may be different from those of the free troposphere. Apart from aerosol spatial distribution, understanding of their vertical distribution is very important for the quantitative determination of their radiative effects (Heitzenberg et al., 1997). However, aerosol measurements, particularly their vertical distribution, are less and unevenly distributed in India.

Apart from ground based and satellites measurements, aircraft is proven to be an effective platform for in-situ measurements of atmospheric aerosol over a reasonably large spatial domain. Moreover, aerosol measurements during monsoon are very sparse over India to understand aerosol–cloud interactions. Especially, aerosols in the neighbourhood of clouds are seldom observed by other traditional approaches. CAIPEEX conducted over India with an instrumented aircraft led a pathway to understand the aerosol vertical distribution and their interaction with monsoon clouds. More details about CAIPEEX are presented in an overview of the experiment (Kulkarni et al., 2012). CAIPEEX phase-I was carried out for the first time over different parts of Indian sub continent during May to September 2009, which was a major drought year. In this paper, the vertical and spatial distribution of aerosol over different locations under different synoptic conditions is discussed. For the same year, earlier studies based on satellite data showed the presence of elevated aerosol layer upto 4 km (Rahul et al., 2011) and aerosol indirect effects during the long breaks in Indian summer monsoon (Manoj et al., 2012). On one hand aerosols are found to inhibit the cloud growth (Albrecht, 1989) and on the other hand they are found to enhance the deep convection (Koren et al., 2005) under suitable conditions. However, the influence of aerosol effects on monsoon under drought conditions is yet to be unravelled.

2. Data and methodology

2.1. Aircraft instrumentation and data

A twin engine Piper Cheyenne-II research aircraft was used during the intensive observation period. It was equipped with standard instrumentation for state parameters (such as temperature, pressure, relative humidity, and wind speed). More details of all the equipped instruments namely, the list of instruments, their range, accuracy, resolution, frequency and measured parameters were given elsewhere (Kulkarni et al., 2012) and at website http://www.tropmet.res.in/~caipeex/. The instrumentation used on-board for aerosol measurements was a Passive Cavity Aerosol Spectrometer Probe (PCASP-100X), with diameter ranging from 0.1 to 3 μm. This instrument is used mainly for the detection of fine and accumulation mode particles. Aerosol total number concentration is obtained by normalizing the total bin counts with the sample flow rate. While data processing the first bin data is discarded, as the data in the first bin is usually very high and treated as noise data. Cloud Droplet Probe (CDP) is used to measure the drop size distribution in the size range of 2–50 μm. In this study, the CDP number concentration outside the cloud (with a threshold of <15 μm) is used to represent the coarse mode particles. In clear sky conditions, CDP is mainly considered to detect large aerosol particles such as mineral dust and sea salt aerosols (Zhang et al., 2006; Padmakumari et al., 2013). The sizing calibration was done regularly by using glass beads of known sizes through a sampling volume. The optics of the instruments were cleaned regularly for removal of dirt and also were purged with nitrogen gas for removal of moisture content in the probes. Data is collected at an interval of 1 s (~100 m) and is quality controlled. Radiosonde (RSRW) flights were carried out at each base before the time of aircraft flight.

2.2. Area of flight operations

CAIPEEX phase-I was conducted from six different locations in India representing different environmental conditions from May to September 2009. The locations are Pune (18.52 °N; 73.85 °E) in the west, Pathankot (32.23 °N; 75.63 °E) in the north near the foot hills of the Himalayas, Hyderabad (17.45 °N; 78.38 °E) in the south central, Bareilly (28.42 °N; 79.45 °E) in the north of Indo Gangetic Plain, Bengaluru (13.14 °N; 77.62 °E) in the south and Guwahati (26.09 °N; 91.58 °E) in the north-eastern part of India. Fig. 1 shows the map of India with different bases from where the aircraft was operated and corresponding flight tracks.

The entire flight is segregated as ascent and descent flights i.e. from the surface to a maximum altitude and from maximum altitude to the surface, respectively. Thus, we have two profiles for each flight separated by a distance of ≈300 km–400 km. Data is averaged for every 200 m in vertical. The profiles are shown above the mean sea level. Standard deviations at every 200 m are also used in the study to represent the spatial variation in aerosol number concentration. ABL height above mean sea level is obtained from the vertical profiles of virtual potential temperature derived from radiosonde profiles. Surface level aerosol concentration and ABL mean concentration are obtained by averaging the lowest 200 m layer and concentrations up to the ABL height, respectively.

2.3. Other data sets

CALIPSO 5 km Aerosol Layer Product (Version 3.01) is used to categorize different aerosol sub types over the flight operational areas. The aerosols are categorized into six different types, based on their integrated attenuated backscatter and volume depolarization ratio along with ancillary information on surface type and altitude

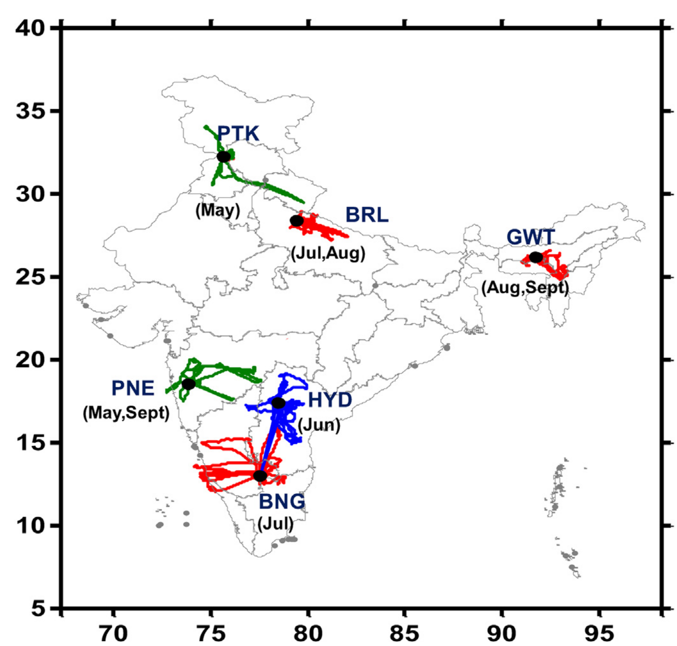

Fig. 1. Map of India showing the aircraft flight tracks from different bases.

Table 1
Flight information at the bases Pune, Pathankot, Hyderabad and Bengaluru.

Base	Date	Flight information	Remarks
Pune	17.5.09	1210–1315 h; SW of Pune.	Haze Layer up to 15,000 ft
	19.5.09	1510–1615 h; NE of Pune.	Haze Layer up to 15,000 ft
Pathankot	23.5.09	1320–1515 h; over Pathankot and NW covering Srinagar valley.	Dark haze layer over Srinagar up to 17,000 ft
	24.5.09	1400–1600 h; local and towards NE and SE of Pathankot.	Forest fires over the hills
	26.5.09	1435–1800 h; towards SW and then to north of Pathankot.	Brown Haze layer up to 15000 ft. Haze rising along the foothills.
	27.5.09	1340–1650 h; towards NE along the foot hills.	Clouds grow through the brown haze layer.
	28.5.09	1300–1550 h; vertical profile in local area.	Haze layer up to 14,000 ft.
Hyderabad	11.6.09	1310–1450 h; towards SE of Hyderabad.	No clouds in this sector. Sky is hazy.
	12.6.09	1215–1515 h; towards SE, S, W and then NE of Hyderabad.	Partially cloudy sky with strong NW wind.
	14.6.09	1335–1620 h; towards N and NE of Hyderabad.	Partially cloudy sky with moderate winds.
	15.6.09	1330–1600 h; towards east of Hyderabad.	Cu clouds emerging from the lower haze layer.
	16.6.09	1340–1630 h; towards SSW of Hyderabad.	Cu and Ac in haze layer
	17.6.09	1330–1630 h; towards N of Hyderabad.	Hazy weather with warm cumuli
	18.6.09	1300–1615 h; towards south and SE of Hyderabad.	Hazy weather
	19.6.09	1550–1650 h;	Hazy atmosphere with few cumuli
	20.6.09	1310–1635 h; towards east of Hyderabad.	Cumuli in hazy atmosphere
Bengaluru	1.7.09	1330–1535 h; towards western ghats.	Maritime airmass.
	2.7.09	1520–1650 h; towards Anantapur, east of Bengaluru.	Westerly monsoon flow prevails with convergence over the area.
	3.7.09	1155–1435 h; towards western ghats, west coast and 20 miles into the Arabian sea to measure in the off shore trough.	Westerly monsoon flow prevails and caused heavy rain at the west coast.
	5.7.09	1155–1510 h; Same as above	Partly cloudy with low clouds and overcast with high clouds. Hazy up to 12,000 ft
	6.7.09	1310–1620 h; Towards Anantapur, east of Bengaluru.	Westerly monsoon flow prevails, hazy with visibility about 25 km.
	7.7.09	1140–1520 h; towards west coast.	Heavy haze up to 12,000 ft.
	8.7.09	1140–1450 h; towards western ghats-NW-W-E.	Hazy atmosphere with marine airmass
	13.7.09	1215–1600 h; towards western ghats.	Haze up to 16,000 ft over the Arabian Sea and western ghats.

(Omar, 2009). The six aerosol types analysed are clean continental, polluted continental, dust, polluted dust, clean marine, and smoke. Clean continental represents the background aerosol, polluted continental represents the pollution emanating from the continent and polluted dust represents the mixture of dust and biomass burning smoke. For more details on these aerosol models see Omar, 2009.

To represent the source of the observed aerosol concentrations, Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model is used to obtain 8 days back trajectories based on the Global Data Assimilation System (GDAS) data products. Aerosol optical depths (AOD) derived from MODIS-Aqua satellite (Level 3) were also used in the study.

3. Results and discussion

3.1. Synoptic conditions

The year 2009 witnessed 22% deficit south west monsoon rainfall of its long period average. This large deficiency in south west monsoon rainfall due to the prevalence of two longer duration dry spells, one in June and the other in July–August, categorizes the year 2009 under all India severe drought year, preceded by 2002 and 2004 (Weather in India, 2010). The prolonged hiatus in June is due to weak equatorial flow and lack of formation of low pressure systems in the Bay of Bengal. During this period severe heat wave conditions prevailed over many parts of northwest, central and adjoining

Table 2
Flight information at the bases Bareilly, Guwahati and Pune.

Base	Date	Flight information	Remarks
Bareilly	16.7.09	1425–1605 h; Towards E-SE of Bareilly over Indo Gangetic Plain (IGP).	Clear atmosphere without haze
	17.7.09	1335–1640 h; same as above.	Hazy atmosphere
	18.7.09	1330–1550 h; same as above.	Good visibility
	22.7.09	1500–1730 h; same as above.	Clear atmosphere without haze
Bareilly	19.8.09	1500–1615 h; Towards E-SE of Bareilly over IGP.	Deep Hazy atmosphere
	20.8.09	1555–1835 h; Same as above	Clouds formed in hazy air
	23.8.09	1415–1700 h; Same as above	Hazy atmosphere
	24.8.09	1410–1620 h; vertical profile in the local airspace.	Very Hazy, moist and polluted atmosphere.
	25.8.09	1415–1615 h; Same as above	Hazy and moist polluted atmosphere
Guwahati	29.8.09	1530–1655 h; profiling in Assam valley.	Polluted atmosphere
	30.8.09	1445–1700 h; in the SE sector of Guwahati.	Hazy and polluted atmosphere
	4.9.09	1245–1605 h; vertical profile towards south of Silchar, south of Meghalaya over a paper mill.	Polluted atmosphere
	6.9.09	1145–1415 h; Same as above.	Polluted atmosphere
Pune	16.9.09	1435–1722 h; Flight towards west (Mumbai).	Hazy and heavily polluted atmosphere towards Mumbai.
	22.9.09	1450–1707 h; Flight towards north of Pune (Sholapur).	Hazy sky
	23.9.09	1519–1823 h; Flight towards east of Pune (Nanded).	Hazy sky
	24.9.09	1451–1810 h; Same as above.	Thick haze up to 15,000 ft

eastern parts of India. According to the study of Krishnamurti et al. (2010), the first break is associated with a blocking high around the Arabian Peninsula, which is responsible for enhanced intrusion of dry continental air in association with northwest winds from mid-latitude region and subsequent inhibition of vertical growth of deep convection. The second break is attributed to internal dynamics associated with the west-ward propagation of planetary-scale equatorial Rossby waves (Neena et al., 2011).

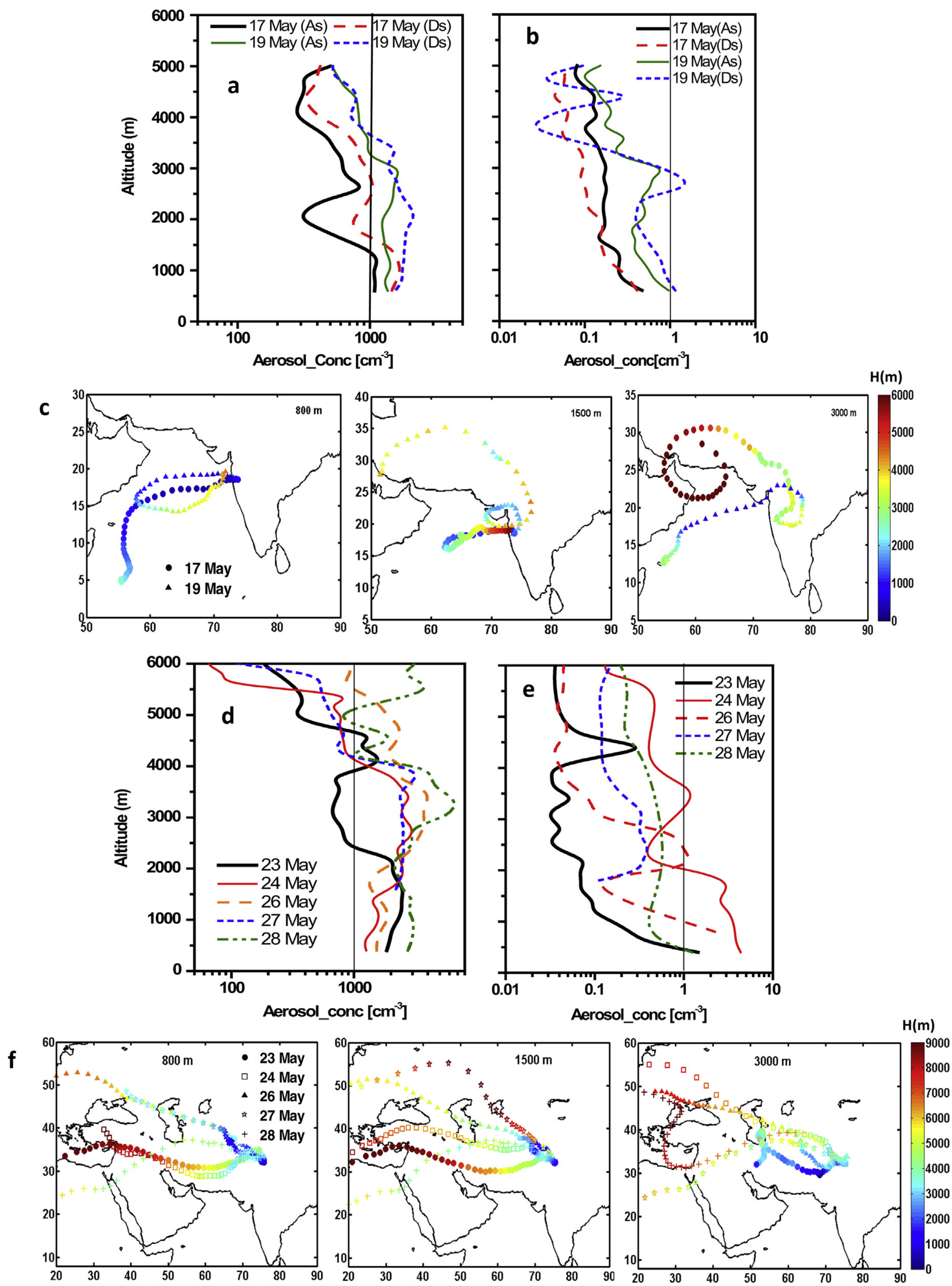

Fig. 2. Vertical profiles of aerosol (a) fine and (b) coarse mode number concentrations and (c) the respective back trajectories with vertical information at three different levels during May at location Pune. Similarly (d), (e) and (f) represent the same but for location Pathankot.

3.2. Flight information

Aircraft measurements were made in the month of May representing pre-monsoon conditions over the locations Pune and Pathankot. During this period sky was very hazy. There was lot of biomass burning activity over Pathankot. With the progress of monsoon, the flight moved to different bases from June to September. During this period the sky was mostly hazy with moist airmass and polluted atmosphere. Day wise flight information is given in Tables 1 and 2. Salient characteristics of aerosol distribution at various base locations are illustrated below.

3.3. Aerosol vertical distribution

3.3.1. Measurements during pre-monsoon

Pune & Pathankot

Over Pune, the measurements were made on 17th and 19th May, while on 23rd, 24th, 26th, 27th and 28th May over Pathankot. Over both the locations elevated aerosol layers were observed between 2 and 4 km as shown in Fig. 2(a, d). Over Pune, the fine mode surface concentrations varied from 1100 to 1500 cm^{-3} and ≈800 to 1600 cm^{-3} at the elevated layers. The fine mode surface concentrations over Pathankot varied from 1300 to 2800 cm^{-3} while the concentrations in the elevated layers are higher, varied from 2500 to 6800 cm^{-3}, as compared to Pune. Mean surface level number concentration and AOD from MODIS are high over Pathankot as compared to Pune (Fig. 3).

Over Pune, the mean ABL height is 1.4 km. On the 17th, the fine mode aerosol concentration dropped above the boundary layer and again increased from 2 km. While on the 19th May, consistent increase in aerosol concentration is observed (Fig. 2a). The coarse mode aerosol profiles also showed elevated layer at 2–3 km on the 19th May (Fig. 2b). The back trajectories at different altitudes (800 m, 1500 m, 3000 m) (Fig. 2c) show that at the elevated layer, the long range transport is from the land and also there exists large scale subsidence into this layer, whereas at lower levels the transport is from the sea. Over this region, there was no biomass burning activity during this period. The elevated layers are due to the influx of dust aerosol from the long range transport from the Thar desert in the north western India contributing to the increase in the coarse mode aerosol. During this period, WRF-Chem (Weather Research and Forecasting – Chemistry version) model simulations also confirmed that Thar desert is acting as source of dust aerosol over this region (Dipu et al., 2013). WRF-Chem constructed airmass trajectories for 17th, 19th and 20th May 2009 are shown at http://www.tropmet.res.in/~majfiles/thara-dipu.html (Note: to open with internet explorer). 1–5 km layers is considered and averaged for every 1 km depth (colour of trajectories indicate the height level, with blue indicating surface to 1000 m above ground and red colour indicating 5000 m above ground).

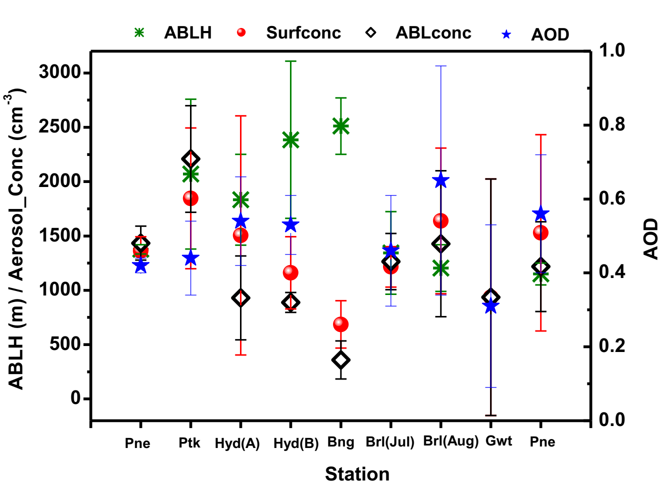

Fig. 3. Variability of mean (of all flight days) atmospheric boundary layer height (ABLH), surface aerosol number concentration, ABL mean concentration and AOD at different bases with standard deviations.

Over Pathankot, the mean ABL height for the observational period is about 2.2 Km. Above the ABL, enhanced aerosol layer is observed on all the days, while on 23rd enhanced layer is observed at higher altitude between 3.5 and 4.5 km (Fig. 2d). The coarse mode aerosol profiles showed high surface concentrations as well as elevated aerosol layers at different altitudes (Fig. 2e). The back trajectories over Pathankot (Fig. 2f) indicate that on all the flight days and at different altitudes, the long range transport of dust and other pollutants are from the north and north-western parts of the continent and there exists large scale subsidence over the observational region. The biomass burning activity over this region was also more, as noted in the MODIS fire maps, and more details on the characteristics of aerosols in the elevated pollution layers and their sources over the same observational region with in situ measurements of size distribution are given elsewhere (Padmakumari et al., 2013). Their study also showed that the elevated layers are composed of both dust from long range transport as well as local and biomass burning particles. There are also several other studies with satellite data which showed elevated pollution layers in the IG plains (Gautam et al., 2011; Vadrevu et al., 2012).

During the above observational periods, the surface temperatures reached about 40 °C which might have induced deep convection. The elevated layers of aerosols would occur as a consequence of strong thermal convection, which lifts aerosol near the ground to higher altitudes (Stull, 1999; Satheesh et al., 2006). Near the Himalayan region, the evidence for ejection of pollution to higher levels in association with dynamically forced updrafts is illustrated in Prbaha et al. (2012). It is also illustrated in that study that the Valley pollution could be brought to the heights above the Haze layer in pockets, which can also be aided through the buoyancy generated due to the biomass burning plumes, which are found to reach to 3–4 km near the slopes. Long-range transport of aerosols from other places could also lead to elevated layers at higher altitudes (Niranjan et al., 2007) as inferred from trajectory analysis. During pre-monsoon, the aerosol vertical distribution is influenced by both the ABL and the air-mass trajectories and a sharp transition is observed between the ABL and the free troposphere, in the presence of elevated aerosol layers. ABL mean concentration, which is higher than the surface level concentration, increased with increase in ABL height (Fig. 3).

3.3.2. Measurements during transition and monsoon

(1) Hyderabad

Over Hyderabad, the experiment was conducted from 11 to 22 June. Based on back trajectories, the days from 11 to 15 June (A) and 16 to 20 June (B) were plotted separately (Fig. 4) to observe the changes in the aerosol vertical distribution. For the period A, the fine mode aerosol vertical profiles showed high concentrations and remained nearly constant up to 4 km (Fig. 4a). The vertical gradient of the aerosol number concentration is small and no sharp transition is observed between the ABL and the free troposphere. The origin of the winds, at all levels, is from the northwest (Fig. 4c) and there exists large-scale subsidence over the observational region.

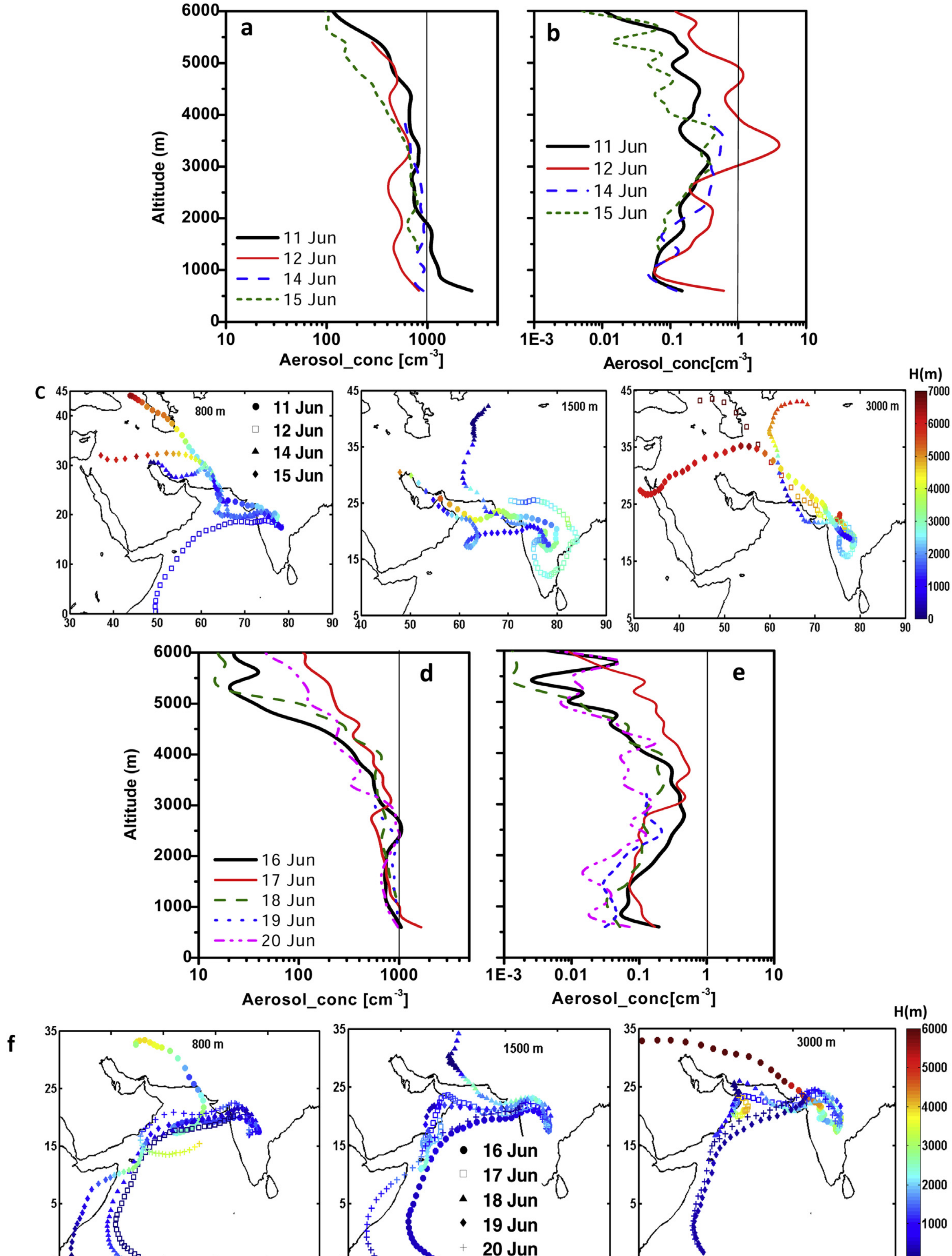

Fig. 4. Vertical profiles of aerosol (a) fine and (b) coarse mode number concentrations and (c) the respective back trajectories with vertical information at 3 different levels during June (11–15) at location Hyderabad. Similarly (d), (e) and (f) represent the same but for dates 16–20 June at Hyderabad.

High surface concentrations are due to the local pollution as well as from long range transport. The coarse mode aerosol profiles show very prominent elevated layer from 2 to 5 km (Fig. 4b), which is consistent with the large scale subsidence of dry air with dust component. MODIS AOD is also found to be high (as seen in Fig. 3) for this period.

While for the period B, the aerosol profiles show high concentrations from the surface to 3 km and there after a decreasing trend is observed (Fig. 4d). During this period winds at all the levels are mostly from the sea (Fig. 4f) and sweeping through the central India on all the days. As compared to case A, higher ABL height and lower mean surface level concentration is observed (Fig. 3). There exists vertical gradient of number concentration and transition between the ABL and the free troposphere is also noted. In both the cases, the number concentration remained almost constant in the ABL, but varied above ABL depending upon the direction of the air mass trajectories. The coarse mode aerosol profiles continued to show elevated aerosol layers from 2 to 5 km (Fig. 4e) and MODIS AOD is also high.

Fine mode aerosol concentrations in the ABL is due to local anthropogenic pollutants and the persistence of dust in the free troposphere for the entire observational period over Hyderabad is due to the incursion of west Asian dry desert air toward central India. The spatial distribution of humidity during this period showed dry condition extending toward central India from north-west (Krishnamurti et al., 2010).

(2) Bengaluru

The experiment was conducted from 1 to 13 July in the southern peninsula from the base Bengaluru towards the west coast, east and north of the base. During this period the westerly flow prevailed and caused heavy rains in the west coast. The aerosol vertical profiles and the respective back trajectories are shown in Fig. 5 (a, b, c). The back trajectories at lower levels showed the airmass originating from the sea. The mean ABL height is about 2.6 Km and surface level concentration is 700 cm^{-3}. All the days (except on 3rd and 6th July), showed elevated layers above the ABL at different altitudes but with less number concentrations. The flight on 3rd July was towards the west coast, while it was towards east of the base on 6th July. On all other days haze was persistent up to higher altitudes and largely influenced by the south westerly flow. The fine mode aerosol number concentration decreases with altitude forming vertical gradient and transition between the ABL and the free troposphere is observed. The coarse mode aerosol profiles showed elevated layers in the ABL as well as above 4 km (Fig. 5b). This might be due to the dust component coming from the Arabian Peninsula at higher altitudes as seen from the trajectories. Haze at lower levels is due to the local pollution and the marine westerly flow.

(3) Bareilly

Over Bareilly, the experiment took place towards east of its location from 16 to 22 July and from 19 to 25 August. Bareilly is located in the IG plains close to the foot hills of the Himalayas and also falls in the monsoon trough region. The vertical profiles and the respective back trajectories for July and August are shown in Fig. 6(a, b, c) and 6(d, e, f), respectively. The mean ABL height is very low due to the developed monsoon conditions (Fig. 3). In July, the fine mode aerosol profiles showed high and nearly constant concentrations in the ABL and thereafter rapidly decreased with altitude forming a strong vertical gradient. While, the coarse mode aerosol profiles showed less number concentrations in the ABL and elevated layers above ABL (Fig. 6b). The back trajectories at all levels show south-westerly flow but sweeping through the polluted east and north eastern regions before reaching the observational location.

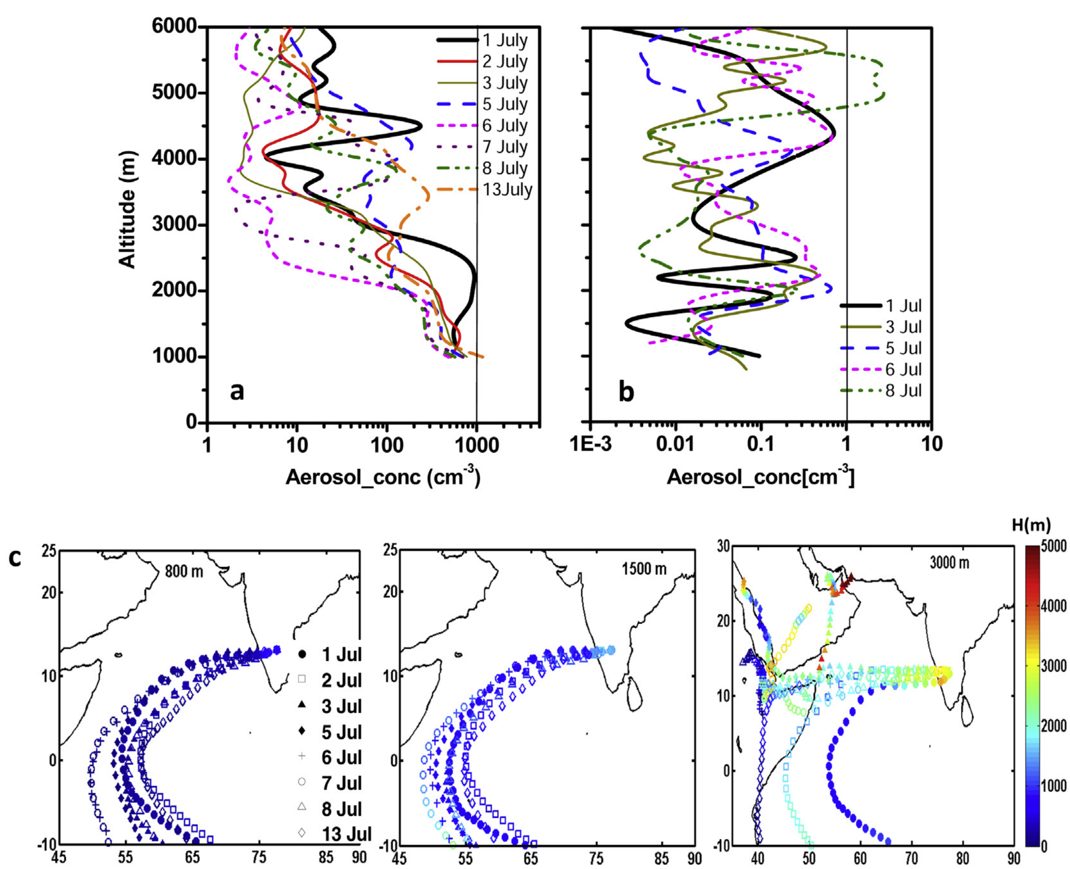

Fig. 5. Vertical profiles of aerosol (a) fine and (b) coarse mode number concentrations and (c) the respective back trajectories with vertical information at 3 different levels during July at Bengaluru.

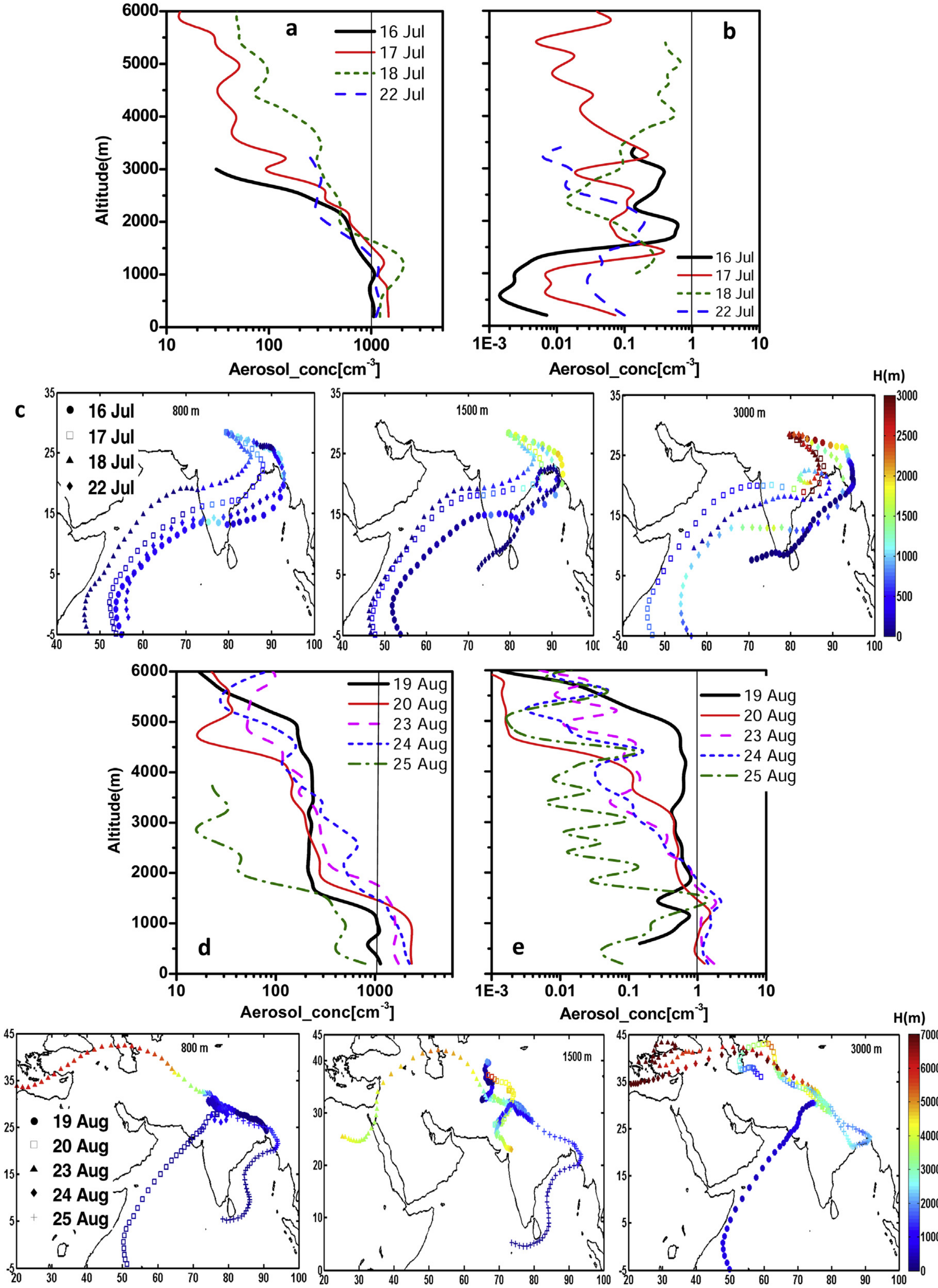

Fig. 6. Vertical profiles of aerosol (a) fine and (b) coarse mode number concentrations and (c) the respective back trajectories with vertical information at 3 different levels during July at location Bareilly. Similarly (d), (e) and (f) represent the same but for August.

During August also the ABL concentrations are high but showed large day to day variability. A very sharp transition in aerosols is observed between the ABL and the free troposphere as compared to July. As compared to July, lower ABL height and higher mean surface level concentration is observed in Aug (Fig. 3). MODIS AOD is also more in August as compared to July. The coarse mode profiles peak at 1.5 km and then decrease with height (Fig. 6e). The wind flow is mostly from northwest at all the altitudes. High fine mode

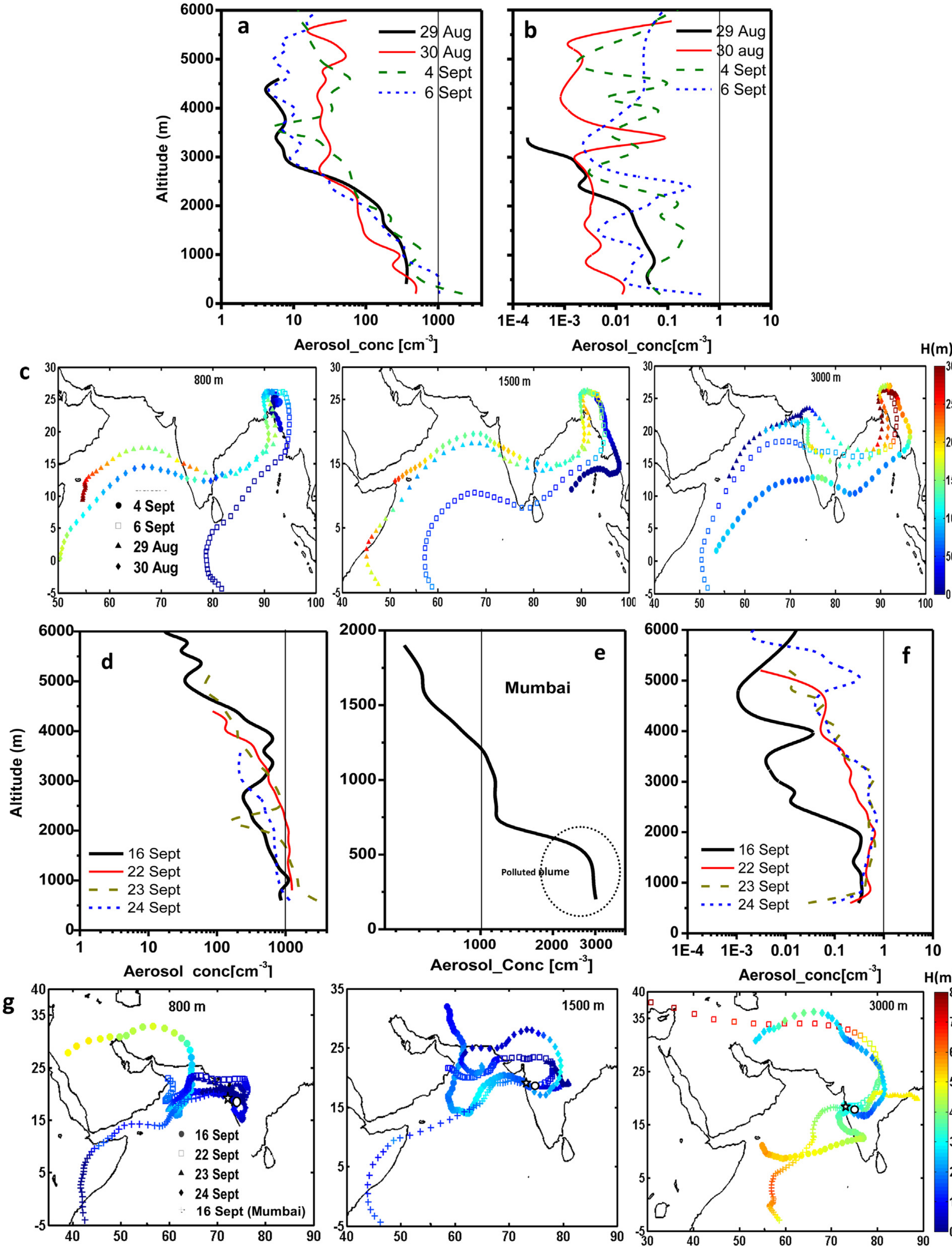

Fig. 7. Vertical profiles of aerosol (a) fine and (b) coarse mode number concentrations and (c) the respective back trajectories with vertical information at three different levels at location Guwahati. Similarly (d), (f) and (g) represent the same but for location Pune and (e) represents the vertical profile of fine mode aerosol in the south of Mumbai. In Fig g, star and circle represent Mumbai and Pune, respectively.

concentration in the ABL could be attributed to the local emissions even after wash out, while at higher levels it is due to the long range transport. On 24th August the winds are entirely from the northwest, whereas on 25th August the winds are entirely from the southwest. Due to this difference in air mass directions in two consecutive days a large difference in the aerosol number concentration is observed. The concentrations on 25th August are much less at all altitudes as compared to other days in August.

(4) Guwahati

Research flights were planned for the first time in Assam valley in North East India with Guwahati as base. Guwahati is the largest city in the valley and near the foot hills of the eastern Himalayas. The valley extends from west to east. Relatively stable and light wind conditions prevailed during the experiment. Pollution levels are relatively high and concentrated mostly in the valleys and near the surface. The mean surface level fine mode concentration is about 1000 cm^{-3} and decreased up to 3 km and remained nearly constant above that (Fig. 7a). On 4th September the observations were taken inside a smoke plume over a Nagaon paper mill, where high number concentration is observed at 1000 m. During the observational period all the trajectories are south westerly (Fig. 7c) corresponding to cleaner air associated with the monsoon circulation. The observed fine mode concentration is due to the local sources in the terrain induced ABL. The observed coarse mode concentrations are relatively less but isolated enhanced layers are observed at different altitudes on different days (Fig. 7b). This may be due to the moisture transport from the south (oceanic Bay) and the local pollution led to the formation of haze that filled the entire valley. There are also blocking effects in the valley which can lead to the trapping of pollution in these valleys. The stable conditions do not allow vertical mixing and may lead to extended lifetime of pollution in the valley.

(5) Pune

During 16 to 24 September, the experiment was again conducted from Pune base. The observations were taken towards the west, south of Mumbai and along the west coast over the Arabian sea and then towards the east of the base. Weak monsoon conditions prevailed over Pune during this period. The mean surface level concentration is about 1530 cm^{-3} and decreased slowly with height (Fig. 7d). On 16 September, the flight towards south of Mumbai flew as low as 200 m into the valley like terrain and passed through the pollution plume over Mumbai and measured high fine mode concentrations of 3000 cm^{-3} up to 600 m (Fig. 7e). The coarse mode profiles showed more concentrations in the ABL and decreased with height (Fig. 7f). The winds, almost at all levels, originated from the northwest (except on 16 September at higher level) (Fig. 7g). MODIS AOD is also high (0.56) for this period.

From Fig. 3 it is noticed that ABL height showed a negative correlation with ABL mean number concentration and also with surface level number concentration. During pre monsoon ABL mean concentrations are more than surface concentrations, while during monsoon it is vice versa. During monsoon, good correlation is observed between AOD and surface level number concentration as compared to that with ABL mean number concentration. Dominance of fine/coarse mode aerosols at different altitudes might control the AOD. The correlations are valid for most of the individual days also. However, a few deviations are observed during the monsoon, as monsoon boundary layer is generally influenced by the prevailing large scale synoptic and dynamical conditions. Thus, the surface level number concentration and the height of ABL could significantly influence AOD during monsoon. Similar study carried out over Beijing also observed the same (Liu et al., 2009).

3.4. Aerosol spatial distribution

The aerial extent of the entire flight is about 300–500 km. The ascent and the descent aerosol profiles with a time gap of 3–4 h showed different number concentrations indicating the spatial and temporal distribution of aerosol. In some cases, the aircraft passed through plumes of pollution indicating large spatial distribution of aerosol. Fig. 8a shows a case study of the aerosol spatial distribution from the base Pune towards the south of Mumbai. As the aircraft passed through the pollution plume over Mumbai high number concentrations are observed. The west coast is also affected by this pollution as seen in the figure. Fig. 8b shows another case study where the aircraft flew through a small white plum emitted by a Nagaon paper mill which is 80 nm to the east of Guwahati.

The standard deviations (SD) of the averaged profiles of the ascent and the descent flights at every 200 m are shown in Fig. 9 to represent the spatial distribution of fine mode aerosol. It is noticed that during May, over Pune and Pathankot, the SD is less in the ABL and more in the elevated polluted layers. Over Pathankot the SD is very high near the polluted layers as compared to Pune. High

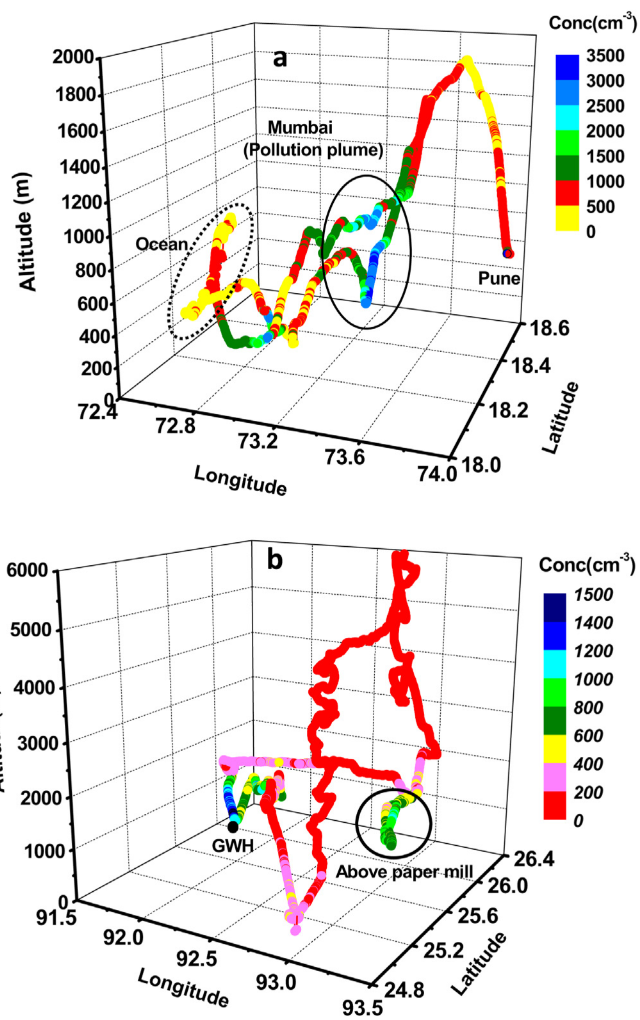

Fig. 8. Aerosol spatial distribution along the flight track from the base (a) Pune towards the south of Mumbai and (b) Guwahati (GWH) towards Nagaon paper mill.

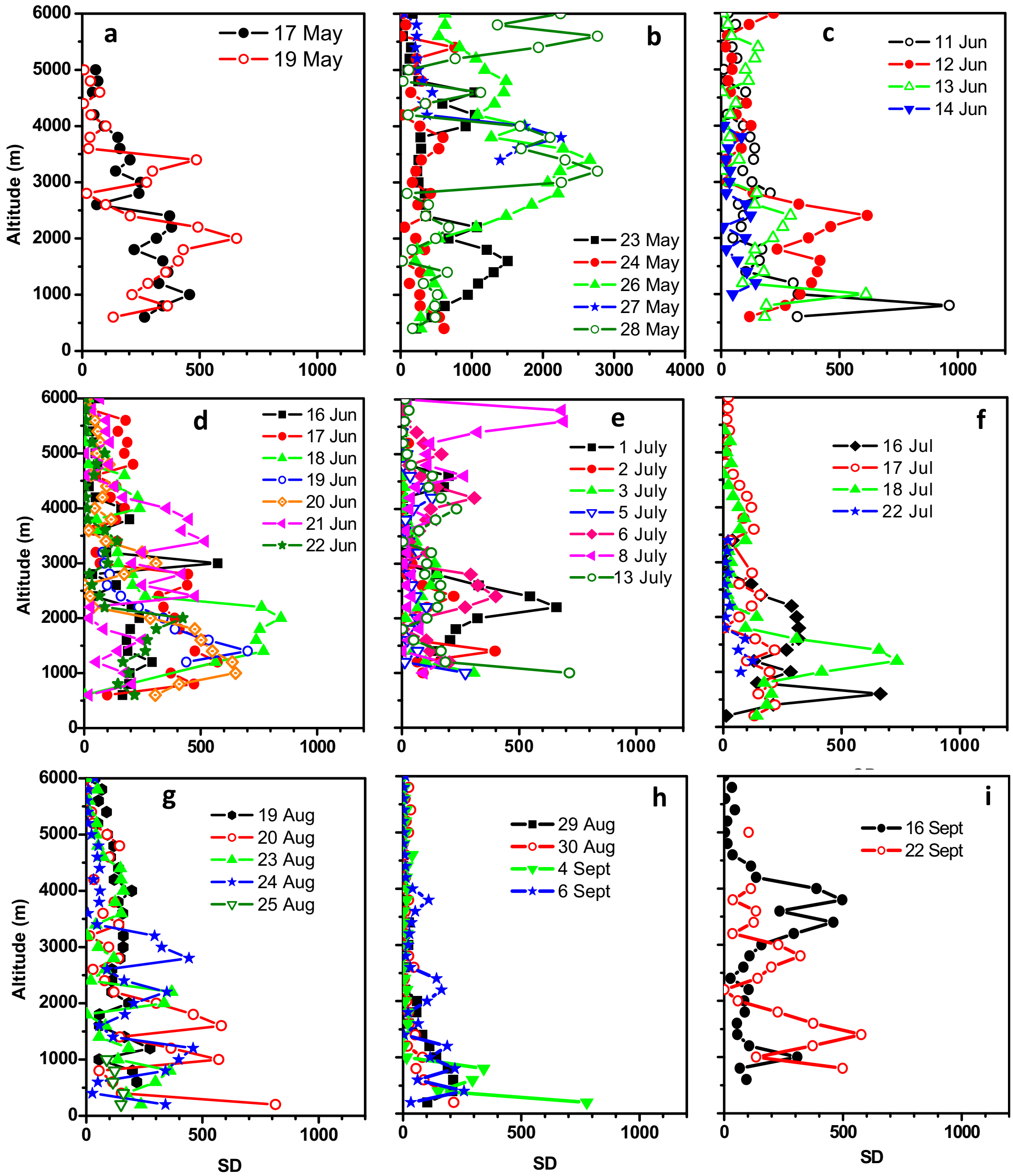

Fig. 9. The standard deviations (SD) of the averaged profiles of the ascent and the descent flights to represent the spatial distribution of fine mode aerosol number concentration at (a) Pune (May) (b) Pathankot (May) (c) Hyderabad-A (d) Hyderabad-B (e) Bengaluru (Jul) (f) Bareilly (July) (g) Bareilly (August) (h) Guwahati (Aug–Sept) (i) Pune (Sept).

spatial variability is observed over Pathankot. The variability in the number concentrations above the ABL is due to the long range transport from other places especially from north-western countries.

During monsoon, SDs are less as compared to May. But their variability is found to be more in the ABL as compared to higher altitudes except for few days. The higher variability in the ABL is due to the contribution from various local sources influenced by the terrain characteristics. The spatial variability is found to be more over Hyderabad as compared to Bareilly and Bengaluru. At higher altitudes, less variability during monsoon is mostly due to the marine component in the south westerly flow, while high variability is due to the intrusion of dry air from higher levels. The spatial distribution of coarse mode aerosol is found to be mostly uniform with less variability. Few days (such as 19 May over Pune, 12, 17 and 22 June over Hyderabad) showed more SD indicating more spatial variability (Figure not shown).

3.5. Aerosol characterization

The frequency distribution of AOD for different aerosol types derived from CALIPSO, at and around the observational area averaged for the flight days at each location is shown in Fig. 10. It is noticed that during pre-monsoon, at Pune and Pathankot, dust, polluted dust and smoke are the dominant aerosol types and the aerosol profiles also showed high fine and coarse mode number concentrations. During monsoon, over Hyderabad, polluted dust is

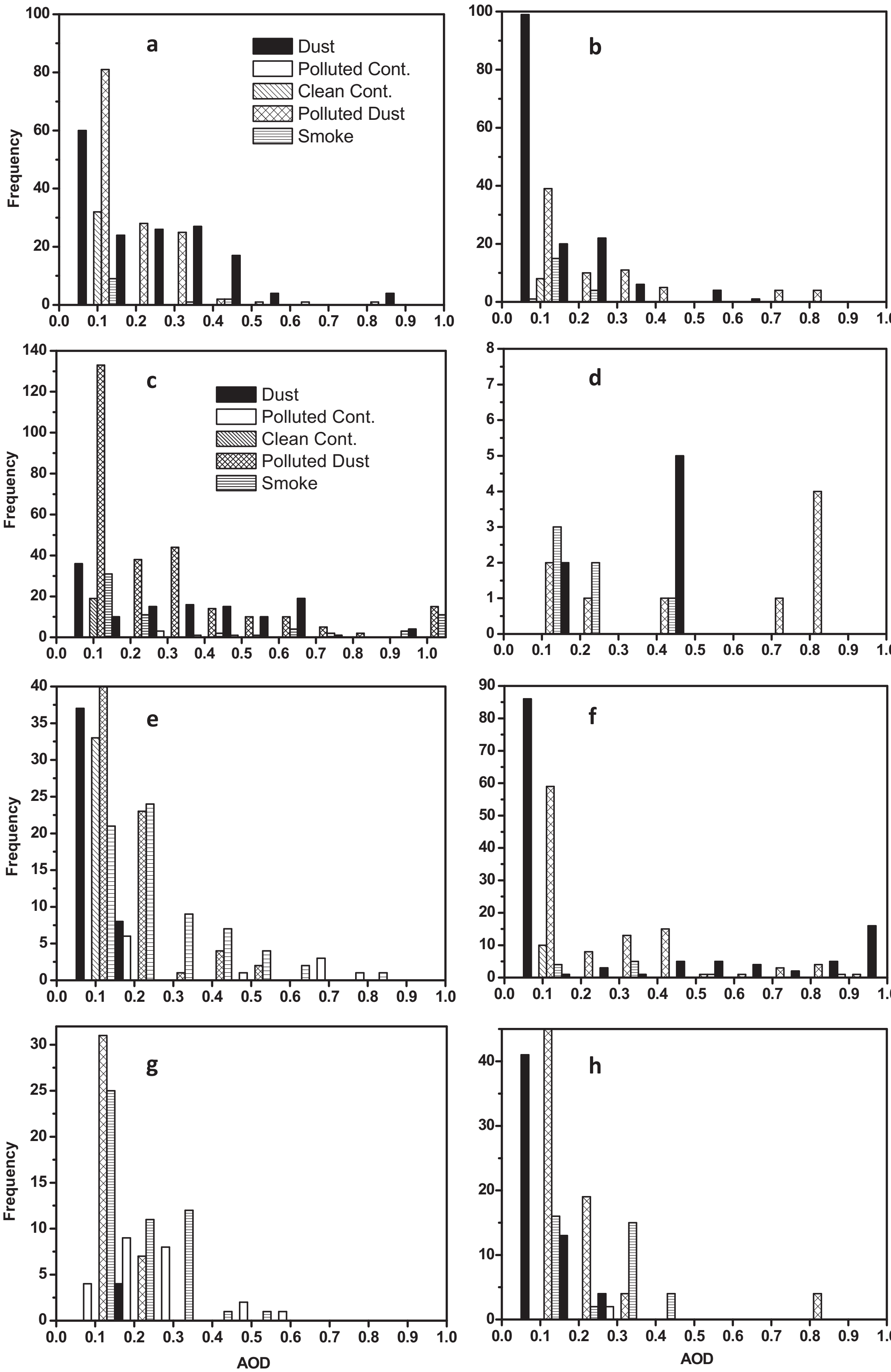

Fig. 10. CALIPSO derived frequency distribution of AOD of different pollutants at and around the observational area averaged for the available flight days for (a) Pune (May) (b) Pathankot (c) Hyderabad (d) Bengaluru (e) Bareilly (July) (f) Bareilly (Aug) (g) Guwahati and (h) Pune (Sept).

the dominating aerosol type followed by dust and smoke. The mixture of both dust and smoke, from local anthropogenic activities, led to high levels of polluted dust. In concurrence with these observations, the aerosol profiles also showed high fine and coarse mode number concentrations. Over Bengaluru, as the monsoon is active during July less number of data samples are available due to cloud contamination. However, traces of pollutants are still observed leading to the formation of haze in the presence of moisture from the southerly flow. At Bareilly, in July the dominance of smoke is more followed by polluted dust and dust. As the winds are south westerly, traces of polluted clean continental aerosol is also noticed. Over this region thick haze is observed even after washout indicating highly polluted ABL due to more anthropogenic activities and moisture supply from the south westerly flow. Hence, high fine mode number concentration is observed in the ABL and high coarse mode above the ABL. In August, the frequency of dust and polluted dust are more with traces of smoke as the higher level winds are from the northwest and there is large scale subsidence over this region, also reflected in the observed aerosol profiles. At Guwahati, smoke is dominant as discussed in the above section, along with polluted dust and polluted continental type. During September, again dust, polluted dust and smoke are dominant over Pune. On the whole, the frequency of occurrence of dust, polluted dust and smoke are found to be high during 2009 due to weak south westerly flow and less wash out effect.

The vertical variability of aerosol effective radius (R_e) (mean at each level) derived from PCASP (Fig. 11) also depicts that during

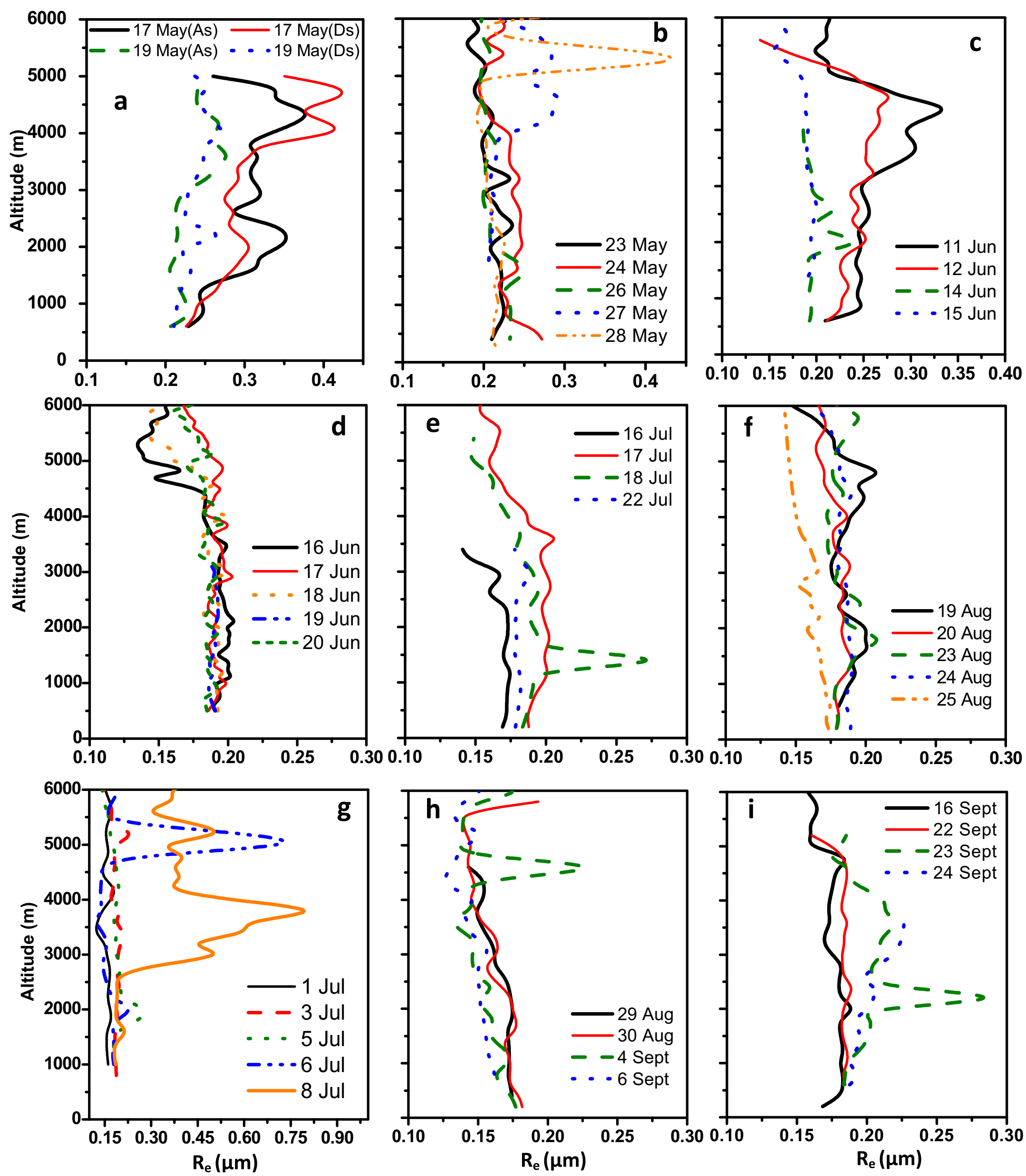

Fig. 11. Vertical distribution of aerosol effective radius (R_e) at (a) Pune (May) (b) Pathankot (May) (c) Hyderabad-A (d) Hyderabad-B (e) Bengaluru (Jul) (f) Bareilly (July) (g) Bareilly (August) (h) Guwahati (Aug–Sept) and (i) Pune (Sept).

pre-monsoon, $R_e > 0.2$ μm, while during monsoon $R_e \leq 0.2$ μm with occasional increase at elevated layers.

4. Conclusions

In situ measurements of aerosol especially during summer monsoon are very important in understanding the role of aerosol in modulating the Indian monsoon. The salient features observed in the vertical and spatial distribution of aerosol, their sources and types over the continental India, covering pre-monsoon and monsoon months with an instrumented aircraft during CAIPEEX in 2009 are as follows.

The aerosol vertical profiles at different regions showed different vertical structures mostly influenced by ABL height as well as the origin of air mass trajectories. During pre-monsoon, surface level as well as mean ABL concentrations increased with increase in ABL height. While, during monsoon, surface level concentration increased with decrease in ABL height. As compared to mean ABL concentration, surface level concentration showed good correlation with AOD during monsoon.

Elevated pollution layers are observed during pre-monsoon as well as during monsoon at some locations but comparatively with less number concentration. In IG plains, a large difference in the aerosol number concentration is observed in two consecutive days due to the difference in the direction of air mass. The spatial distribution showed high variability at the elevated layers as compared to that in the boundary layer during pre-monsoon. While during monsoon high variability is observed in the boundary layer and occasional high variability at higher altitudes due to the intrusion of dry air at upper levels.

The frequency distribution of different aerosol types from CALIPSO showed the dominance of dust, polluted dust and smoke during pre-monsoon. During monsoon also traces of these pollutants were found to be high as the year 2009 is a drought year. The observations of aerosol types from CALIPSO are in concurrence with the aircraft measurements of aerosol number concentrations. The concentrations might be less during a good monsoon year due to continuous wash out and the dominance of marine airmass.

The in-situ data reported in this study is the first of its kind in the monsoon environment over India. The information on the vertical and spatial distribution of aerosols will help in understanding the aerosol behaviour in the monsoon environment and also incorporating them in regional models could improve the parameterization schemes.

Acknowledgements

The authors wish to thank Prof. B. N. Goswami, Director, IITM for his support and encouragement and CAIPEEX team members for making the mission successful. Also thank science data team of CALIPSO and MODIS. This project is funded by Ministry of Earth Sciences, India. We also thank anonymous reviewers for their suggestions in improving the manuscript.

References

Ackerman, A.S., Toon, O.B., Stevens, D.E., Heymsfield, A., Ramanathan, V., Welton, J., 2000. Reduction of tropical cloudiness by soot. Science 288, 1042–1047. http://dx.doi.org/10.1126/science.288.5468.1042.

Albrecht, B.A., 1989. Aerosol, cloud microphysics and fractional cloudiness. Science 245, 1227–1230.

Babu, S.S., Moorthy, K.K., Manchanda, R.K., Sinha, Puna Ram, Satheesh, S.K., Vajja, Dinkar Prasad, Srinivasan, S., Arun Kumar, V.H., 2011. Free tropospheric black carbon aerosol measurements using high altitude balloon: do BC layers build "their own homes" up in the atmosphere? Geophysical Research Letters 38, L08803 http://dx.doi.org/10.1029/2011GL046654.

Dey, S., Tripathi, S.N., Singh, R.P., Holben, B.N., 2004. Influence of dust storms on aerosol optical properties over the Indo-Gangetic basin. Journal of Geophysical Research 109, D20211. http://dx.doi.org/10.1029/2004JD004924.

Dipu, S., Prabha, T.V., Pandithurai, G., Dudhia, J., Pfister, G., Rajesh, K., Goswami, B.N., 2013. Impact of elevated aerosol layer on the cloud macrophysical properties prior to monsoon onset. Atmospheric Environment 70, 454–467. http://dx.doi.org/10.1016/j.atmosenv.2012.12.036.

Gautam, R., Christina Hsu, N., Kafatos, M., Tsay, S.-C., 2007. Influences of winter haze on fog/low cloud over the Indo-Gangetic plains. Journal of Geophysical Research 112, D05207. http://dx.doi.org/10.1029/2005JD007036.

Gautam, R., Hsu, N.C., Tsay, S.C., Lau, K.M., Holben, B.N., Bell, S., Smirnov, A., Li, C., Hansell, R., Ji, Q., Payra, S., Aryal, D., Kayastha, R., Kim, K.M., 2011. Accumulation of aerosol over the Indo-Gangetic plains and southern slopes of the Himalayas: distribution, properties and radiative effects during the 2009 pre-monsoon season. Atmospheric Chemistry and Physics 11, 12,841–12,863. http://dx.doi.org/10.5194/acp-11-12841-2011.

Heitzenberg, J., Charlson, R.J., Clarke, A.D., Liousse, C., Ramanathan, V., Shine, K.P., Wendisch, M., Helas, G., 1997. Measurements and modeling of aerosol single scattering albedo: progress, problems and prospects. Beitraege zur Physik der Atmosphaere 70, 249–263.

Kaskaoutis, D.G., Hsu, C., Holben, B.N., Takemura, Toshihiko, Gautam, R., Singh, R.P., Houssos, E.E., Goto, Daisuke, Singh, Sachchidanand, Bartzokas, A., Kosmopoulos, P.G., Sharma, Manish, 2012. Influence of anomalous dry conditions on aerosols over India: transport, distribution and properties. Journal of Geophysical Research 117, D09106. http://dx.doi.org/10.1029/2011JD017314.

Koren, I., Kaufman, Y.J., Rosenfeld, D., Remer, L.A., 2005. Aerosol invigoration and restructuring of Atlantic convective clouds. Geophysical Research Letters 32, L14282. http://dx.doi.org/10.1029/2005GL023187.

Krishnamurti, T.N., Thomas, A., Simon, A., Kumar, Vinay, 2010. Desert air incursions, an overlooked aspect, for the dry spells of the Indian Summer Monsoon. Journal of Atmospheric Science 67, 3423–3441. http://dx.doi.org/10.1175/2010JAS3440.1.

Kulkarni, J.R., Maheskumar, R.S., Morwal, S.B., Padma Kumari, B., Konwar, M., Deshpande, C.G., Joshi, R.R., Bhalwankar, R.V., Pandithurai, G., Safai, P.D., Narkhedkar, S.G., Dani, K.K., Nath, A., Nair, Sathy, Sapre, V.V., Puranik, P.V., Kandalgaonkar, S.S., Mujumdar, V.R., Khaladkar, R.M., Vijayakumar, R., Prabha, T.V., Goswami, B.N., 2012. The cloud aerosol interactions and Precipitation Enhancement experiment (CAIPEEX): overview and preliminary results. Current Science 102, 413–425.

Liu, P., Zhao, Chunsheng, Zhang, Qiang, Deng, Zhaoze, Huang, Mengyu, Ma, Xincheng, Tie, Xuexi, 2009. Aircraft study of aerosol vertical distributions over Beijing and their optical properties. Tellus 61B, 756–767. http://dx.doi.org/10.1111/j.1600-0889.2009.00440.

Manoj, M.G., Devara, P.C.S., Joseph, S., Sahai, A.K., 2012. Aerosol indirect effect during the aberrant Indian summer monsoon breaks of 2009. Atmospheric Environment 60, 153–163.

Moorthy, K.K., Satheesh, S.K., Suresh Babu, S., Dutt, C.B.S., 2008. Integrated campaign for aerosols, gases and radiation budget (ICARB): an overview. Journal of Earth System Science 117, 243–262.

Neena, J.M., Suhas, E., Goswami, B.N., 2011. Leading role of internal dynamics in the 2009 Indian summer monsoon drought. Journal of Geophysical Research 116, D13103. http://dx.doi.org/10.1029/2010JD015328.

Niranjan, K., Madhavan, B.L., Sreekanth, V., 2007. Micro pulse lidar observation of high altitude aerosol layers at Visakhapatnam located on the east coast of India. Geophysical Research Letters 34 (3), L03815. http://dx.doi.org/10.1029/2006GL028199.

Omar, A., 2009. The CALIPSO automated aerosol classification and lidar ratio selection algorithm. Journal of Atmospheric and Oceanic Technology 26, 1994–2014. http://dx.doi.org/10.1175/2009-JTECHA1231.1.

Padmakumari, B., Maheskumar, R.S., Morwal, S.B., Harikishan, G., Konwar, M., Kulkarni, J.R., Goswami, B.N., 2013. Aircraft observations of elevated pollution layers near the foothills of the Himalayas during CAIPEEX-2009. Quarterly Journal of the Royal Meteorological Society 139, 625–638. http://dx.doi.org/10.1002/qj.1989.

Prabha, T.V., Karipot, A., Axisa, D., Padma Kumari, B., Maheskumar, R.S., Konwar, M., Kulkarni, J.R., Goswami, B.N., 2012. Scale interactions near the foothills of Himalaya during CAIPEEX. Journal of Geophysical Research 117, D10203. http://dx.doi.org/10.1029/2011JD0167.

Rahul, P.R.C., Bhawar, R.L., Salvekar, P.S., Devara, P.C.S., Jiang, J.H., 2011. Evidence of atmospheric brown clouds over India during the 2009 drought year. IEEE 99, 1–6. http://dx.doi.org/10.1109/JSTARS.2011.2170554.

Raj, P.E., Saha, S.K., Sinbawne, S.B., Deshpande, S.M., Devara, P.C.S., Jaya Rao, Y., Dani, K.K., Pandithurai, G., 2008. Lidar observation of aerosol stratification in the lower troposphere over Pune during pre-monsoon season of 2006. Journal of Earth System Science 117 (S1), 293–302.

Ramana, M.V., Ramanathan, V., Podgorny, I.A., Pradhan, B.B., Shrestha, B., 2004. The direct observations of large aerosol radiative forcing in the Himalayan region. Geophysical Research Letters 31, L05111. http://dx.doi.org/10.1029/2003GL018824.

Ramanathan, V., Crutzen, P.J., Lelieveld, J., Mitra, A.P., Althausen, D., Anderson, J., Andreae, M.O., Cantrell, W., Cass, G.R., Chung, C.E., Clarke, A.D., Coakley, J.A., Collins, W.D., Conant, W.C., Dulac, F., Heintzenberg, J., Heymsfield, A.J., Holben, B., Howell, S., Hudson, J., Jayaraman, A., Kiehl, J.T., Krishnamurti, T.N., Lubib, D., McFarquhar, G., Novakov, T., Ogren, J.A., Podgorny, I.A., Prather, K., Priestley, K., Prospero, J.M., Quinn, P.K., Rajeev, K., Rasch, P., Rupert, S., Sadourney, R., Satheesh, S.K., Shaw, G.E., Sheridan, P., Valero, F.P.J., 2001. Indian Ocean Experiment: an integrated analysis of the climate forcing and effects of the great Indo Asian haze. Journal of Geophysical Research 106 (D22), 28371–28398. http://dx.doi.org/10.1029/2001JD900133.

Satheesh, S.K., Vinoj, V., Moorthy, K.K., 2006. Vertical distribution of aerosols over an urban continental site in India inferred using a micro pulse lidar. Geophysical Research Letters 33 (20), L20816. http://dx.doi.org/10.1029/2006GL027729.

Stull, R.B., 1999. An Introduction into Boundary Layer Meteorology. Kluwer Academic Publishers, Dordrecht, Netherlands.

Twomey, S., 1974. Pollution and the planetary albedo. Atmospheric Environment 8, 1251–1256.

Vadrevu, K.P., Ellicott, E., Giglio, L., Badarinath, K.V.S., Vermote, E., Justice, C., Lau, W.K.M., 2012. Vegetation fires in the Himalayan region: aerosol load, black carbon emissions and smoke plume heights. Atmospheric Environment 47, 241–251. http://dx.doi.org/10.1016/j.atmosenv.2011.11.009.

Weather in India, 2010. Monsoon season 2009 (June – September), 2010. Compiled by Meteorological office, Pune, India. Mausam 61 (3), 411–454.

Zhang, Q., Zhao, C., Tie, X., Wei, Q., Huang, M., Li, G., Ying, Z., Li, C., 2006. Characterizations of aerosols over the Beijing region: a case study of aircraft measurements. Atmospheric Environment 40, 4513–4527.