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Effect of the summer monsoon on aerosols at two measurement stations in Northern India – Part 2: Physical and optical properties

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Abstract. Aerosol physical and optical properties were measured at two locations in northern India. The first measurement station was a background site in Mukteshwar, about 350 km northeast of New Delhi, in the foothills of the Indian Himalayas, with data from 2006 to 2009. The second measurement site was located in Gual Pahari, about 25 km south of New Delhi, with data from 2008 to 2009. At both stations, the average aerosol concentrations during the monsoon were decreased by 40–75 % compared to the pre-monsoon average concentrations. The decrease varied with the total local rainfall. In Mukteshwar, the monsoon season removed particles from all size classes, due to a combination of rain scavenging and activation to cloud and mountain fog droplets. The scavenging by rain is least effective for the size range of the accumulation mode particles. In Gual Pahari, this was the only major wet removal mechanism and, as a result, the accumulation mode particles were less effectively removed. Aerosol concentrations during the early monsoon were found to be affected by mineral dust which in Gual Pahari was observed as an increased particle volume at a diameter around 3–4 µm. The single scattering albedo varied from 0.73 to 0.93 during the monsoon season, being slightly lower in Gual Pahari than in Mukteshwar. This is due to the fact that Gual Pahari resided closer to high anthropogenic black carbon emissions. As the absorbing particles are typically in the accumulation mode, they were not effectively removed by rain scavenging. The aerosol columnar properties, which were measured in Gual Pahari, showed a somewhat different seasonal behaviour compared to the surface measurements, with the aerosol optical depth increasing to an annual maximum in the early monsoon season.

1 Introduction

The importance of the Asian Summer Monsoon is well known. In addition to being a water supply and providing agricultural irrigation, the monsoon brings a relief to the severe pollution build up during the winter and spring in the area.

In recent years it has become evident that aerosols may influence the precipitation patterns of the Summer Monsoon. This may be especially important in Southern Asia, since the area suffers from an intense and persistent particulate pollution called the "brown cloud" (e.g., Lelieveld et al., 2001; Nakajima et al., 2007; Ramanathan et al., 2007). The influence of the brown cloud on monsoon patterns has been investigated in numerous studies. Probably the most famous work on this is the so-called "Elevated Heat Pump" hypothesis (EHP) (Lau et al., 2006). According to the EHP hypothesis, absorbing aerosols accumulate against the southern slopes of the Himalayas in the pre-monsoon season, thus, modulating the tropospheric temperature gradient. The reinforced meridional temperature gradient would lead to an early onset of monsoon and intensified rainfall during June and July. Several studies have later attempted to find further evidence to

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support or criticize this hypothesis (e.g., Lau and Kim, 2006; D. Liu et al., 2008; Z. Liu et al., 2008; Meehl et al., 2008; Ramanathan and Carmichael, 2008; Randles and Ramaswamy, 2008; Bollasina et al., 2008; Collier and Zhang, 2009; Gautam et al., 2009a, b; Kuhlmann and Quaas, 2010; Nigam and Bollasina, 2010; Lau and Kim, 2010). The EHP hypothesis is certainly not the only aerosol mechanism affecting the summer monsoon precipitation. A general challenge in all these studies is that the aerosol effects are not simply confined to direct or indirect effects alone, but they can be coupled through feedback processes (Huang et al., 2007), and also semi-direct effects may become important in certain conditions (Nigam and Bollasina, 2010). The net effect of all these mechanisms is not known, and due to both spatial and temporal variability of aerosols, the error sources in these simulations are large. Therefore, observational evidence is of great importance to elucidate the effects of aerosols on monsoon. These reasons provide a comprehensive motivation to study the aerosol-monsoon interactions in the area.

In this paper, we focus on the investigation of the aerosol physical and optical properties and their development during the Summer Monsoon seasons at two stations in India; Gual Pahari in the Indo-Gangetic plains and Mukteshwar in the Himalayan foothills. The companion paper (Hyvärinen et al., 2011) focused on particulate matter and black carbon concentrations. We quantify the effect that the monsoon has on the aerosol concentrations and properties, and compare the differences between different years at the two locations. The data presented here will be useful for modelling studies of aerosol-monsoon interactions and provides information of the aerosol properties during the monsoon.

2 Measurement sites and methods

The first measurement station was a background site in Mukteshwar (29°26′ N, 79°37′ E, Fig. 1), about 350 km northeast of New Delhi in India. The site was located at 2180 m above sea level in a rural region at the Himalaya Mountains. The second measurement site was located in Gual Pahari (28.43° North, 77.15° East, 243 m a.s.l., Fig. 1), Gurgaon, about 25 km south of New Delhi. The surroundings represented a semi-urban environment. Both sites have been described in detail before, see Hyvärinen et al. (2009) and Hyvärinen et al. (2010). The presented results cover the years from 2006 to 2009 for Mukteshwar and from 2008 to 2009 for Gual Pahari.

2.1 Instruments

The parameters measured at Mukteshwar included the particle number size distribution from 10 nm to 800 nm (DMPS), $PM_{2.5}$ and PM_{10} concentrations (on-line beta-attenuation analyzers), aerosol scattering coefficient (Nephelometer at 525 nm), black carbon concentration/absorption coefficient

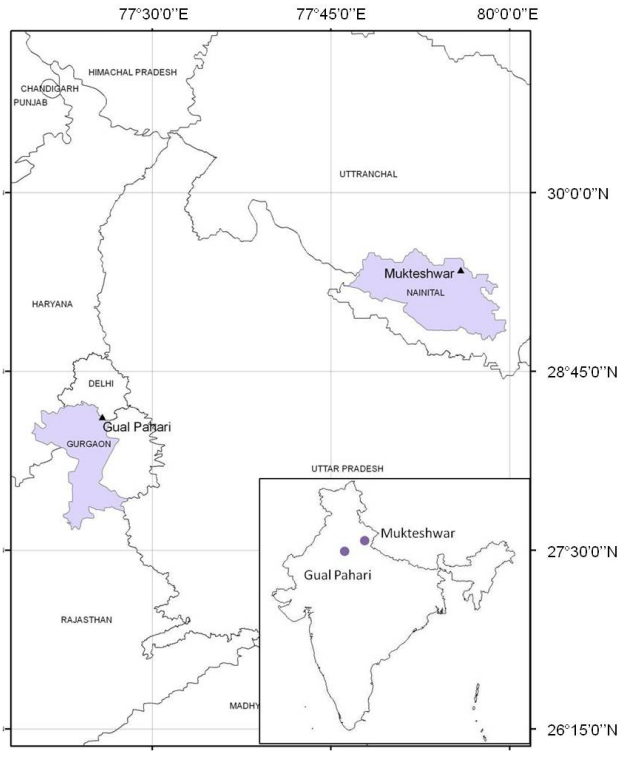

Fig. 1. Map of the measurements stations.

(7-wavelength Aethalometer) and meteorological parameters. The measurements and instruments at the site are presented in more detail by Hyvärinen et al. (2009) and Komppula et al. (2009). All instruments sampled from a single sampling line (except the particle mass monitors which each had their own inlets), with a $PM_{2.5}$ inlet located at about 5 m above ground level and about 2 m above the roof of the station building.

A similar sampling procedure was utilized in Gual Pahari. Two separate inlets with $PM_{2.5}$ and PM_{10} cut-off were used for sampling of the respective aerosol mass concentrations (on-line scattering/beta-attenuation hybrid analyzers). The measurements which were conducted from the main inlet with a PM_{10} cut-off were: particle number size distribution over the diameter range 4 nm–10 µm (twin-DMPS and APS), black carbon concentration/absorption coefficient (MAAP at 637 nm) and aerosol scattering coefficient (Nephelometer at 520 nm). Meteorological parameters were also measured. A more detailed presentation can be found in Hyvärinen et al. (2010). In addition to the in situ measurements, a Raman Lidar was operational during 2008 (see Komppula et al., 2010) and a Cimel sunphotometer during 2009. Cimel measurements were made as a part of the Aerosol Robotic Network, AERONET (e.g., Holben et al., 1998).

2.2 Data processing

The measured data was saved as five minute averages. The five minute data was checked with outliers and obvious instrument malfunctions periods removed. The data was then averaged to one hour with the condition that each hour had more than 25 min of data. All longer time averages were calculated from the hourly data, which was also converted to STP-conditions. One month averages were only calculated if the data covered more than 30 % of the time. For seasonal analyses, the year was divided into four seasons: winter (December–February), pre-monsoon (March-onset of monsoon), monsoon season and post-monsoon (withdrawal of monsoon-November). When seasonal averages were calculated, the determined onset and withdrawal dates defined the monsoon season for each year separately. Again a 30 % data coverage level was demanded, with an exception to the Gual Pahari DMPS size distribution data, which we wanted to present despite the low data coverage. The 1 h data coverage for the low size range (4–58 nm) and high size range (31–850 nm) of the twin-DMPS was 24 % and 33 %, respectively.

We calculated the ratio of Aitken-mode particles (25–75 nm) to accumulation mode (75–800 nm) particles, N_{ait}/N_{acc}. Most of the aerosol particles emitted by various combustion processes are in the size range of 15–100 nm (Chang et al., 2004). The ratio N_{ait}/N_{acc} tends to decrease with increasing distance from major source areas of combustion-derived aerosols, as they grow by condensation and cloud processing toward the accumulation mode. New particle formation increases N_{ait}/N_{acc} ratio because of the growth of nucleation mode particles into the Aitken mode.

The optical coefficients are presented for the wavelength of about 520 nm. From Mukteshwar, the absorption was obtained directly from the Aethalometer channel 520 nm. However, at Gual Pahari, the MAAP measured the absorption at 637 nm (Müller et al., 2011). We converted this absorption coefficient to match 520 nm by assuming a wavelength dependence of absorption $\sigma_{abs} = \lambda^{-\alpha}$ (Bergstrom, 1973; Bohren and Hufman, 1983), with $\alpha = 1.2$ as measured at the Mukteshwar station (Hyvärinen et al., 2009). This value represents a mixture of absorbing materials with mostly black carbon (which has $\alpha = 1.0$, Bergstrom et al., 2007), and some organic carbon and/or mineral dust. We tested the sensitivity of α to the SSA by testing values between 1 and 2. The effect on SSA was less than 5 %.

3 Results

3.1 General features

The monsoon characteristics are presented in Fig. 2. During the four measurement years, there were contrasting monsoon seasons; with both excessive rain and drought. Rainfall was more intensive in Mukteshwar than in Gual Pahari, probably due to the mountain location, as air masses flowing uphill are more likely to form clouds and rain. Year 2008 exhibited the most rainfall, and an early monsoon onset date of 16 June, which is one of the earliest onset dates recorded in the area with rainfall data available since 1901 (Tyagi et al., 2009). The weakest monsoon occurred in 2006. A thorough trajectory analysis of incoming air masses was reported by Raatikainen et al. (2011). In summary, during the monsoon season both sites have air masses from both Bay of Bengal (SE) and Arabian Sea (SW) directions. At other times, westerly air masses dominate.

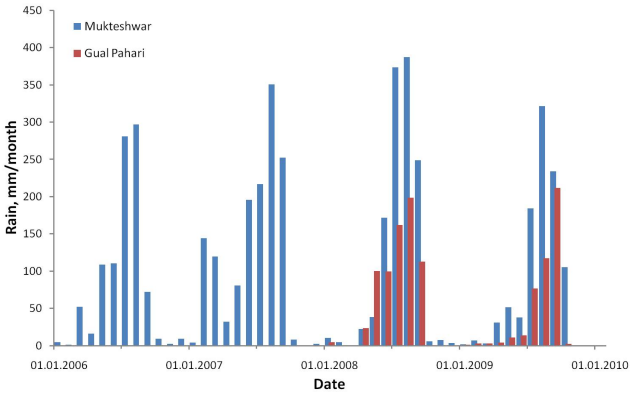

Fig. 2. Monthly rain accumulation during the running of the measurement stations.

The aerosol concentrations and the subsequent optical coefficients in the pre-monsoon season were highest in 2008 and 2009 (Table 1), with an exception to absorption coefficient which was highest in 2007. Lowest pre-monsoon concentrations occurred in 2006. The average aerosol concentrations and optical coefficients during monsoon were decreased by about 40–75 % compared to the pre-monsoon average concentrations at both stations (Fig. 3), having a linear relationship with the total rainfall of the yearly monsoon season. The most effective decrease of the aerosol concentrations were observed during 2008 in Mukteshwar. During 2008, the rain amounts were the highest of the study period. The relative decrease seems nearly independent of the measurement location and can be estimated from the rain accumulation alone by an accuracy of ±10 %. During the post-monsoon season, concentrations rose again. In Gual Pahari, the post monsoon averages were higher than the pre-monsoon averages. In Mukteshwar, however, the post monsoon averages were lower than the pre-monsoon averages. This behaviour can be explained by the lower boundary layer height in the post-monsoon season (Raatikainen et al., 2011). In the following sections, we will detail the behaviour of different aerosol properties during and around monsoon.

Table 1. Pre-monsoon, monsoon and post-monsoon average aerosol properties in Mukteshwar and Gual Pahari. 30 % hourly data coverage is demanded. ± denotes standard deviation calculated from the hourly data.

Mukteshwar Season	Year	Abs coeff.[a] Mm^{-1}	Scat coeff.[b] Mm^{-1}	N_{tot} $\#/cm^3$	$N < 25$ nm $\#/cm^3$	$N25 < d < 75$ nm $\#/cm^3$	$N > 75$ nm $\#/cm^3$	
Pre	2006	21.6±14.6	88.6±54.0	4613±2531	274±581	1715±1230	2976±1534	
Monsoon		7.4±5.2	42.3±40.6	2023±876	54±56	730±323	1228±639	
Post		18.2±12.7	96.3±93.0	3108±1570	69±88	844±573	2125±1124	
Pre	2007	25.8±16.7	96.7±57.5	5761±3447	298±736	1886±1453	3545±2111	
Monsoon		8.9±5.1	56.8±40.6	2653±1011	68±75	976±473	1605±699	
Post		19.6±14.0	97.4±82.5	4010±1965	118±142	1101±636	2446±1236	
Pre	2008	25.1±18.9	133.8±95.0	6901±4054	393±795	2133±1513	4365±2907	
Monsoon		6.9±4.9	34.3±37.5	1909±900	66±70	726±318	1115±640	
Post		18.3±11.4	104.9±87.5	3195±2683	125±208	999±1225	2065±1397	
Pre	2009	25.7±16.6	128.7±119.8	6395±4841	368±685	2176±1831	3824±3448	
Monsoon			74.1±60.3	3244±1819	119±169	1266±781	1929±1222	
Post			31.5±26.5	2124±1234	156±248	777±464	1258±721	
Gual Pahari Season	**Year**	**Abs coeff.[c] Mm^{-1}**	**Scat coeff.[d] Mm^{-1}**	**N_{tot} $\#/cm^3$**	**$N < 25$ nm $\#/cm^3$**	**$N25 < d < 75$ nm $\#/cm^3$**	**$N > 75$ nm $\#/cm^3$**	**V_{tot} $.4 < d < 10$ µm $\mu m^3\,cm^{-3}$**
Pre	2008	62.8±51.3						77.2±57.5
Monsoon							6342±4104	
Post			843.9±626.6					
Pre	2009	60.8±49.7	312.7±253.2				6953±4835	50.6±45.4
Monsoon		28.6±26.9					5607±4059	29.2±20.6
Post		109.0±59.9	1235.5±1291.2	25860±11707	1950±4046	6904±4299	1696±9486	66.0±43.8

Measured at [a] 520 nm [b] 525 nm [c] 637 nm [d] 520 nm.

3.2 Aerosol size distribution

The number size distributions during the pre-monsoon, monsoon and post-monsoon seasons are presented for both stations in Fig. 4a and c, together with the monthly average modal concentrations (Fig. 4b and d).

In Mukteshwar, the average number size distribution was unimodal at all times. The total particle concentrations decreased by about 50–75 % from the pre-monsoon to monsoon season, depending on the amount of rainfall. The nucleation mode decreased by an average of 77 %, the Aitken mode by 53 % and the accumulation mode by 60 %. In addition to the concentration decrease, the seasonal average mode diameter decreased from the pre-monsoon value of ∼101 nm to the monsoon average of ∼81 nm. The average ratio of Aitken-mode particles (25–75 nm) to accumulation mode (75–800 nm) particles, N_{ait}/N_{acc} increased from the pre-monsoon time value of 0.58 to monsoon time value of 0.65. An increased ratio points towards fresher emissions or increased removal of the accumulation mode particles. New particle formation was not observed in Mukteshwar during the monsoon season (Neitola et al., 2011). No major inter-annual variations were found in the monsoon season modal concentration ratios, indicating that the total accumulated rainfall had little influence on this. During post-monsoon the average value for the mode diameter was ∼108 nm. The ratio of Aitken-mode to accumulation mode particles, N_{ait}/N_{acc} decreased to 0.50, illustrating that aerosol aging processes started to take place.

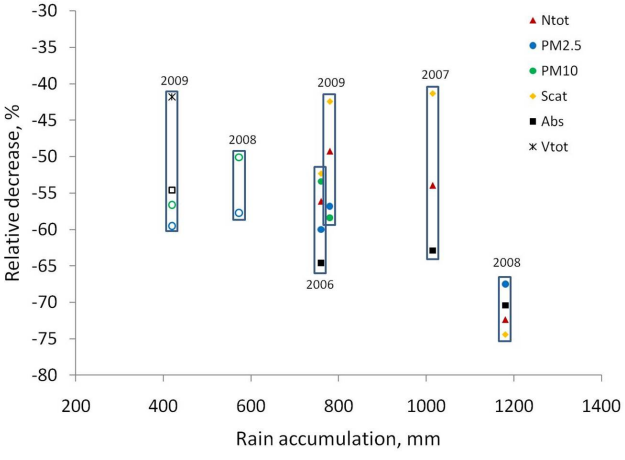

Fig. 3. The relative change of the aerosol concentrations from the pre-monsoon to monsoon season as a function of the June–September rain accumulation. Solid symbols: data from Mukteshwar; open symbols: data from Gual Pahari. Each year is surrounded by a box.

From Gual Pahari, the average size distributions look rather different (Fig. 4c). During pre-monsoon, the nucleation- and Aitken-modes had a strong contribution,

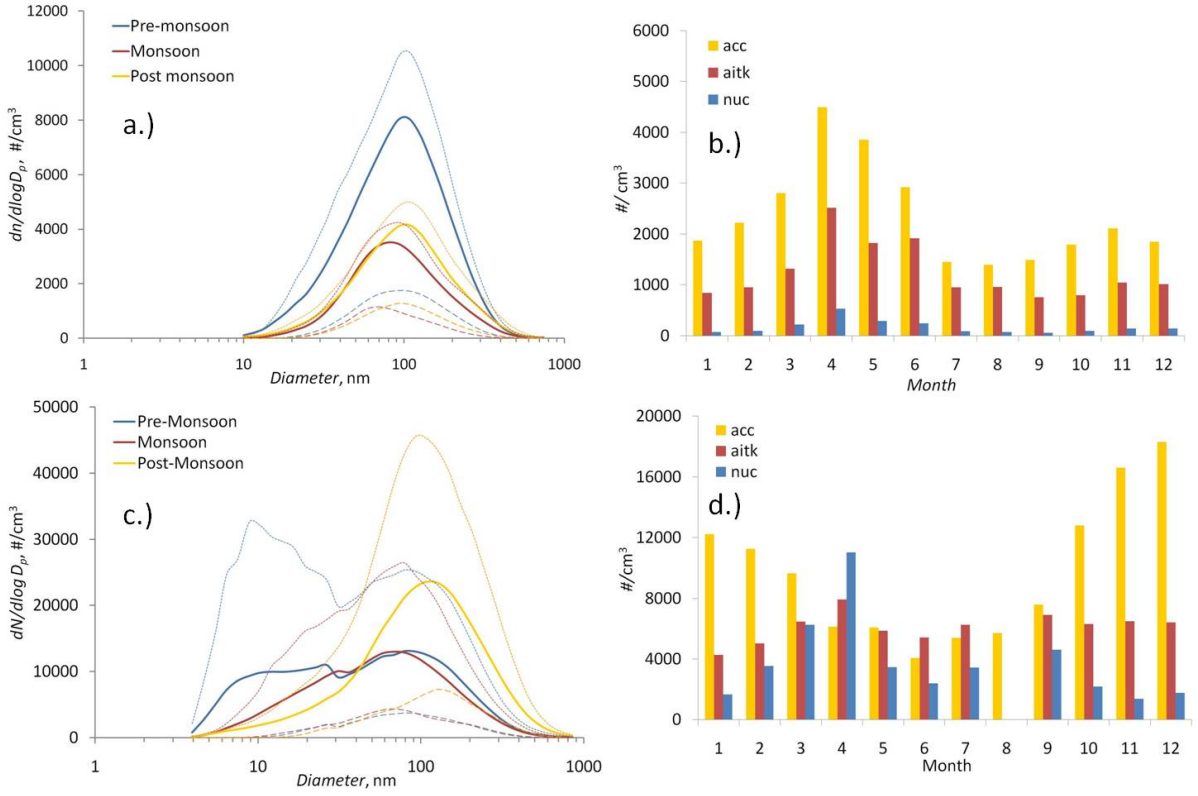

Fig. 4. Average size distributions for different seasons in **(a)** Mukteshwar, and **(c)** Gual Pahari. Solid lines are averages and dashed and dotted lines represent the 10th and 90th percentiles, respectively. Annual variation of the modal particle number concentrations in **(b)** Mukteshwar, and **(d)** Gual Pahari. Nucleation mode $d_p < 25$ nm, Aitken mode 25 nm $< d_p < 75$ nm, accumulation mode $d_p > 75$ nm.

equal to the accumulation mode. This was due to strong new particle formation. The concentration decrease during the monsoon season was less obvious than in Mukteshwar, with the accumulation and Aitken-modes showing very similar values during both seasons (Fig. 4d). Also the nucleation mode concentration decreased only about 20 %, and new particle formation events were still observed. Despite the wet deposition, nucleation precursors apparently existed in Gual Pahari during the monsoon season. During monsoon, the accumulation mode diameter decreased from the pre-monsoon value of ∼80 nm to ∼70 nm. The ratio N_{ait}/N_{acc} was smaller than unity during both the pre-monsoon and the monsoon season, probably due to new particle formation and fresh emissions in the area. During post-monsoon season, similar aging of the aerosols took place as in Mukteshwar, resulting in an elevated accumulation mode, with $N_{ait}/N_{acc} < 1$. It has to be emphasized that the data coverage from the Gual Pahari DMPS was very low, (lowest coverage during the monsoon season: 11 % for the nucleation-, 34 % for the Aitken- and 32 % for the accumulation-mode), and the average size distributions do not necessarily represent the full seasons very well.

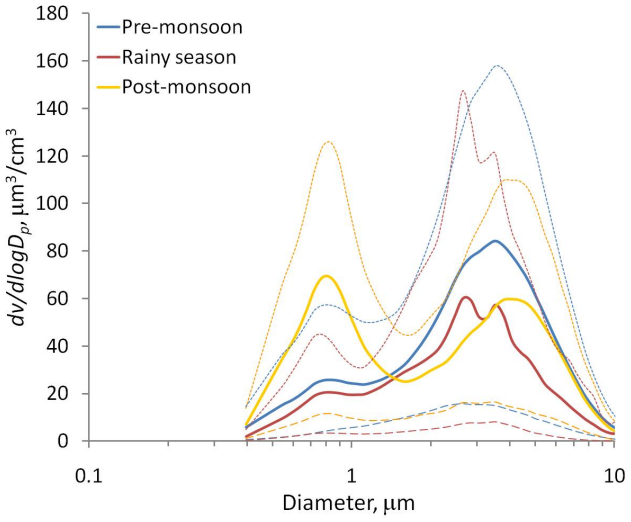

Fig. 5. The volume size distribution in Gual Pahari during different seasons, $0.4 < d_p < 10$ µm. Solid lines are averages and dashed and dotted lines represent the 10th and 90th percentiles, respectively.

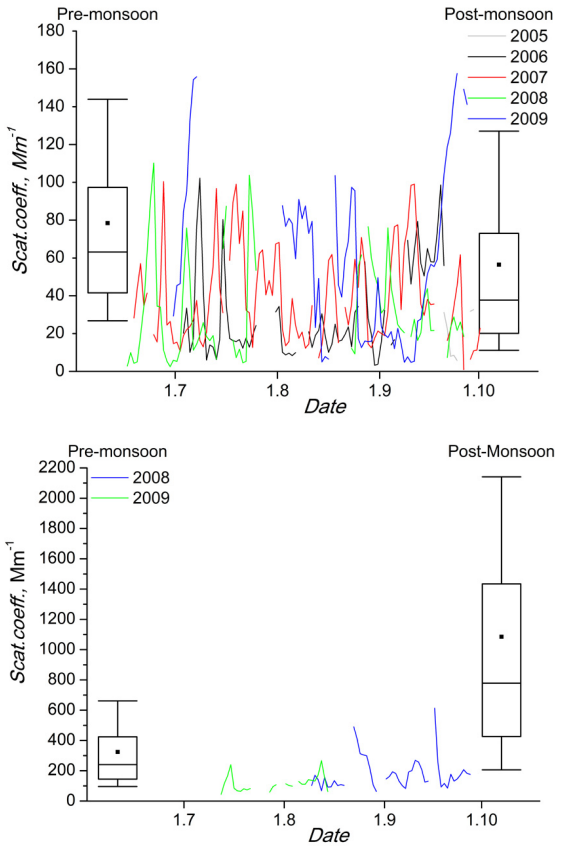

Fig. 6. Scattering coefficient **(a)** at 525 nm in Mukteshwar **(b)** at 520 nm in Gual Pahari. Lines are 24 h running averages from the monsoon season, box plots denote to pre- and post-monsoon seasonal values of 10 %-ile, 25 %-ile, median (line), average (dot), 75 %-ile and 90 %-ile.

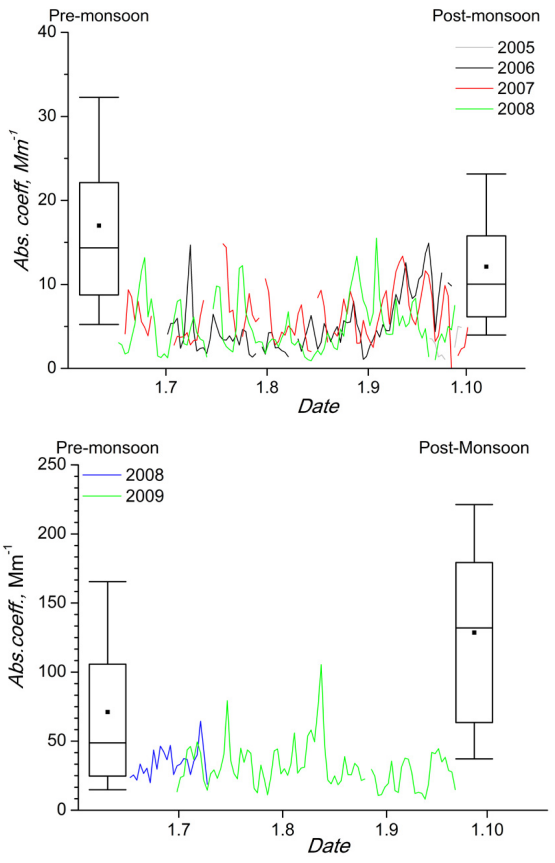

Fig. 7. Absorption coefficient **(a)** at 520 nm in Mukteshwar **(b)** at 637 nm in Gual Pahari. Lines are 24 h running averages from the monsoon season, box plots denote to pre- and post-monsoon seasonal values of 10 %-ile, 25 %-ile, median (line), average (dot), 75 %-ile and 90 %-ile.

When interpreting the size distribution data from the stations, two important aspects related to wet removal processes have to be kept in mind. First, the scavenging by rain is size dependent. The scavenging coefficient has a minimum for particles with a diameter around 150 nm. With a near parabolic shape as a function of particle size, the scavenging coefficient is two times higher for 20 nm and for ∼2 µm particles (e.g., Laakso et al., 2003). This helps to explain why accumulation particles in Gual Pahari were not lost very effectively. On the other hand, the mountain location of Mukteshwar promotes the occurrence of clouds and fogs, which enhance the removal of particles by the activation mechanism to cloud droplets. This mechanism is very effective for particles bigger than 100 nm, and explains the accumulation mode decrease in Mukteshwar.

Volume size distributions were deducted from Gual Pahari APS-measurements in the aerodynamic size range of 0.4–10 µm (Fig. 5) allowing for the inspection of larger particles. The average size distributions during pre-monsoon and monsoon seasons were strikingly similar (albeit lower in concentration during monsoon), with a coarse mode at ∼4 µm during pre-monsoon and ∼3 µm during monsoon. This mode is strongly related to primary particles, indicating that these were a prominent source during the pre-monsoon and monsoon season. The occurrence of the coarse mode also illustrates that sources are an important factor affecting the variability of aerosols during monsoon, not only the wet deposition loss processes. The contribution of mineral dust to the coarse mode was discussed in context of high concentration episodes during monsoon in the companion paper (Hyvärinen et al., 2011). A trajectory analysis conducted in that paper indicated that the origin of mineral dust was mainly from the Thar Desert. A smaller mode, which appears as a shoulder at ∼0.8 µm was significantly higher during the post-monsoon season, exceeding the concentration of the coarse mode. This relates to the elevated accumulation mode during post-monsoon.

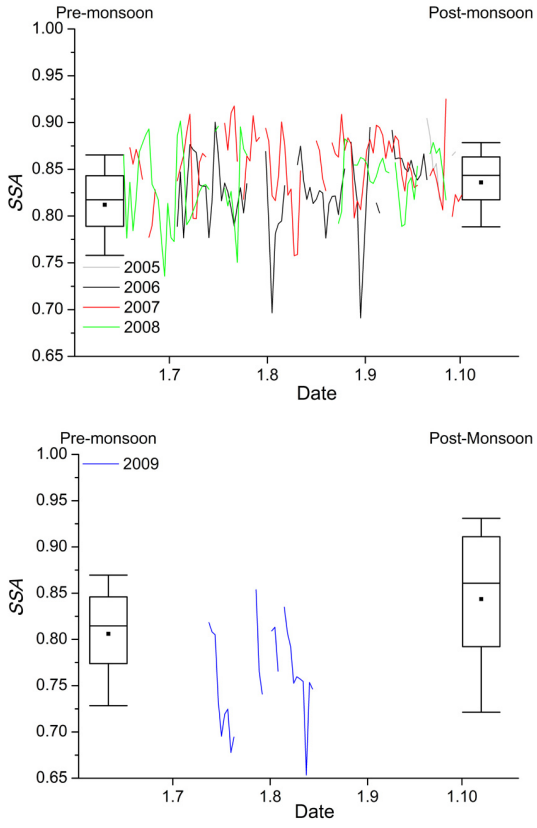

Fig. 8. Single scattering albedo, SSA at 525 nm. **(a)** Mukteshwar **(b)** Gual Pahari. Lines are 24 h running averages from the monsoon season, box plots denote to pre- and post-monsoon seasonal values of 10 %-ile, 25 %-ile, median (line), average (dot), 75 %-ile and 90 %-ile.

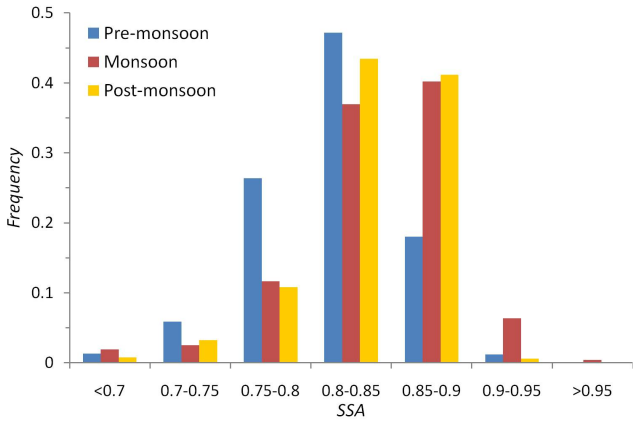

Fig. 9. Histogram of SSA values in Mukteshwar during pre-monsoon, monsoon and post-monsoon.

3.3 Aerosol scattering and absorption coefficient

The optical properties (Figs. 6 and 7) generally followed the seasonal behaviour of concentration levels decreasing between 40–75 % from the pre-monsoon average during monsoon (Table 1 and Fig. 3). In Mukteshwar, the scattering coefficient decreased, on average, less than the absorption coefficient (Fig. 3), mostly due to episodic peak concentrations (Fig. 6) during the monsoon season. Light scattering is more effective, the bigger the particles are. So, again the increased light scattering can be related to mineral dust which was occasionally present during the break spells of the monsoon. The more effective decrease of the absorption coefficient indicates an efficient removal of absorbing aerosols in Mukteshwar. As discussed in the previous section, cloud and fog occurrence provided an additional activation mechanism for aerosol removal in Mukteshwar. The effective decrease of the absorption coefficient implies that the absorbing material was in a hygroscopic form, thus, favouring the activation process.

In Gual Pahari, less data is available for the monsoon time scattering and absorption coefficients. It is notable that, similarly to Mukteshwar, scattering coefficient showed occasional high concentrations, especially during 2009 (Fig. 6). In contrast to Mukteshwar, absorption coefficient in Gual Pahari did not decrease very efficiently (Fig. 3) in the monsoon season. This can be explained by two reasons: first, the accumulation mode with the related absorbing particles was less effectively decreased during the monsoon in Gual Pahari, because of the missing activation removal mechanism. Second, Gual Pahari is located closer to pronounced anthropogenic combustion sources. This can be observed by the occasionally high absorption coefficients (Fig. 7), and in general the high N_{ait}/N_{acc} ratio above unity supports this.

During post-monsoon, a clear difference can be seen at the two stations – at Mukteshwar, both the absorption and scattering coefficient decreased compared to pre-monsoon, while in Gual Pahari a substantial increase was observed. As mentioned before, this is related to the boundary layer evolution, as most of the pollution was confined below the altitude of Mukteshwar. This is studied in more detail by Raatikainen et al. (2010).

3.4 Single scattering albedo

We calculated the single scattering albedo, SSA from the scattering and absorption coefficients measured at both stations. Single scattering albedo is the ratio of the scattering coefficient to the extinction coefficient (scattering + absorption). In Mukteshwar, the single scattering albedo had values of 0.75–0.90 (Fig. 8), being slightly higher during the monsoon season than during the other seasons. The wide range of SSA values illustrates that the aerosol was highly variable in terms of optical properties. Looking at the Mukteshwar data in more detail (Fig. 9), we notice that a higher frequency of low SSA values was observed during the pre-monsoon season. This indicates the abundance of absorbing aerosols, which may originate from anthropogenic combustion sources or from the seasonal forest fires in the area. During monsoon

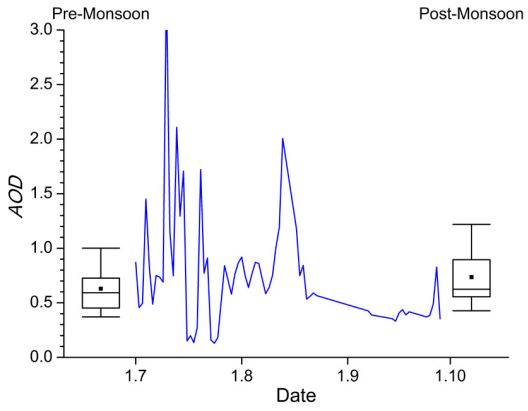

Fig. 10. Aerosol optical depth, AOD at 500 nm, measured with the Cimel sunphotometer during 2009 in Gual Pahari. Line is daily averages, box plots denote pre- and post-monsoon seasonal values of 10 %-ile, 25 %-ile, median (line), average (dot), 75 %-ile and 90 %-ile.

season, a small fraction (∼5 %) of SSA values higher than 0.9 were also observed. These high values corresponded mostly with the higher scattering coefficients and were probably related to mineral dust. The fact that high SSA values were not observed during the pre-monsoon even though this is also the season for dust events suggests that during pre-monsoon the mineral dust was mixed with other, more absorbing aerosol. Some episodes during the monsoon season showed decreased SSA of about 0.70, indicating highly absorbing aerosol. This occurred only with low absorption and scattering coefficients, and was probably from local sources, such as cooking (Hyvärinen et al., 2009). We also compared the SSA against the total particle count N_{tot} from Mukteshwar. As a general finding, the SSA values converged towards 0.9 as N_{tot} increased.

The SSA in Gual Pahari showed slightly lower values than in Mukteshwar, especially during the pre-monsoon, varying from 0.73 to 0.93, with a majority of SSA values (37 %) falling between 0.8 and 0.85. This is hardly surprising, as many anthropogenic sources produce BC in the area. There is very little information about the SSA in Gual Pahari during the monsoon, but the values observed were typically below 0.8, indicating the dominance of absorbing particles. Reasons for the dominance of absorbing particles were discussed in the previous section. Firm conclusions cannot be made from the Gual Pahari monsoon time SSA due to the poor data coverage. For the post-monsoon, the average SSA increased compared to pre-monsoon at both locations. No significant inter-annual variation was observed in the SSA data.

3.5 Aerosol columnar properties

Aerosol columnar properties were observed at Gual Pahari. During 2008, a Raman lidar was installed (Komppula et al., 2010) and during 2009 a Cimel Sunphotometer. The sunphotometer data we present here are level 1.5 data, which means that the data is cloud screened, but some quality checks are missing. The average AOD measured with the sunphotometer at 500 nm during the pre-monsoon was about 0.61 in Gual Pahari (Fig. 10). During the monsoon season the average AOD increased to about 0.70. It is noted that these are daytime values.

The aerosol columnar properties show a different seasonal behaviour compared to the surface measurements, with the AOD having a maximum in the early monsoon season (Fig. 10). This observation is supported by the Lidar height-resolved profiles which show that the aerosol backscatter and extinction were at elevated levels during the monsoon season (Fig. 11). These profiles show that during the pre-monsoon and monsoon seasons the aerosol backscatter was highest in the lower altitudes, below 2 km, and decreased monotonically with altitude. During post-monsoon season, a low-level, but thick aerosol layer was observed. This corresponds well with the boundary layer height (Raatikainen et al., 2011), which implies that during the pre-monsoon the aerosol would be efficiently vertically mixed, opposite to post-monsoon. However, the boundary layer height does not explain the aerosol vertical distribution during the monsoon season.

This illustrates that the columnar measurements during the monsoon season, their interpretation and comparison to ground-level measurements is not a straightforward task. The conflict between the average boundary layer heights may be explained by the fact that during the break spells of the monsoon (and, thus, times when the columnar measurements were actually conducted), the boundary layer heights are elevated compared to the average values (Kusuma et al., 1991). This still does not solve the discrepancy between surface in situ and columnar measurements. As noted in Hyvärinen et al. (2011), dust episodes were a probable reason for the early monsoon high concentrations. We studied this prospect by analysing the refractive index data from the sunphotometer measurements (Fig. 12). The real part of the refractive index obtained values mostly above 1.47. This corresponds well with mineral dust (mixed with other aerosol), which has a refractive index of about 1.5 (e.g., d'Almeida et al., 1991). The AOD's during the early monsoon showed much higher values than during pre-monsoon, when mineral dust was also present in surface measurements. Relative humidity may provide an explanation in the observed discrepancies, as it affects the particle size and, thus, its optical properties (Zieger et al., 2011). While the columnar measurements provide properties of ambient aerosol, the in situ measurements are from a dried aerosol. Drying may reduce the particle sizes by a factor of 2–4, depending on aerosol composition and ambient conditions.

We tested this hypothesis by comparing the Lidar extinction profiles with RH soundings available for the Lidar measurement periods from the near-by New Delhi airport (Fig. 13) (University of Wyoming, department

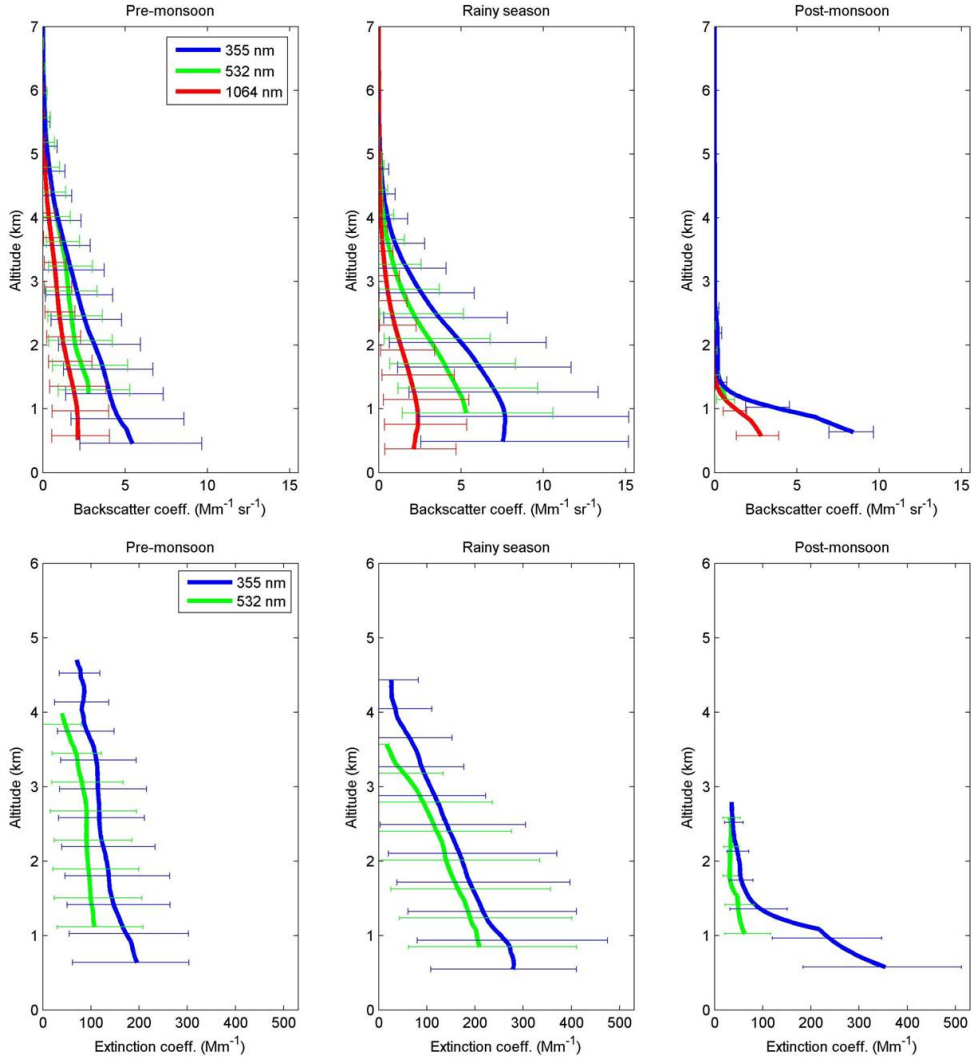

Fig. 11. Seasonal averaged backscatter and extinction profiles at two wavelengths for the Gual Pahari site during 2008. The 10 and 90 percentiles are also shown.

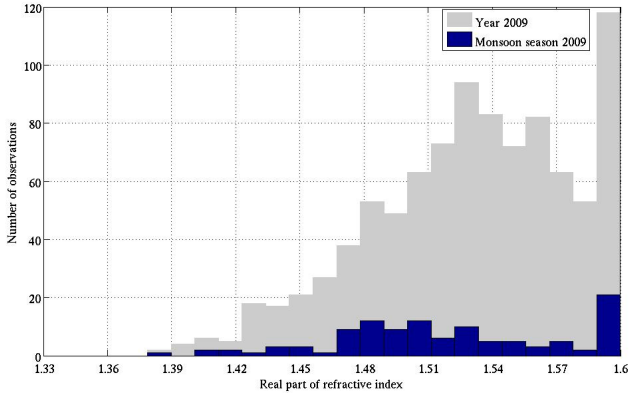

Fig. 12. Histogram of (real part) refractive index from Gual Pahari sunphotometer measurements during 2009.

of atmospheric research, http://weather.uwyo.edu/upperair/sounding.html). It can be seen that the profiles are very similar. This could well be a reason for the discrepancy between surface in situ and columnar measurements. However, to quantify the effect will be difficult without state-of-the-art instruments such as the hygroscopic tandem DMA or a humidified nephelometer. Another explanation for the differences could be that high concentration aerosols, such as mineral dust, occur at higher altitudes rather than near the surface. Indeed, the maximum extinction observed in the Lidar profiles occurred at around 1 km altitude. Another reason for the observed differences could be that the columnar measurements were only conducted during cloud free periods. However, comparing the in situ concentrations only when AOD was measured did not solve the difference.

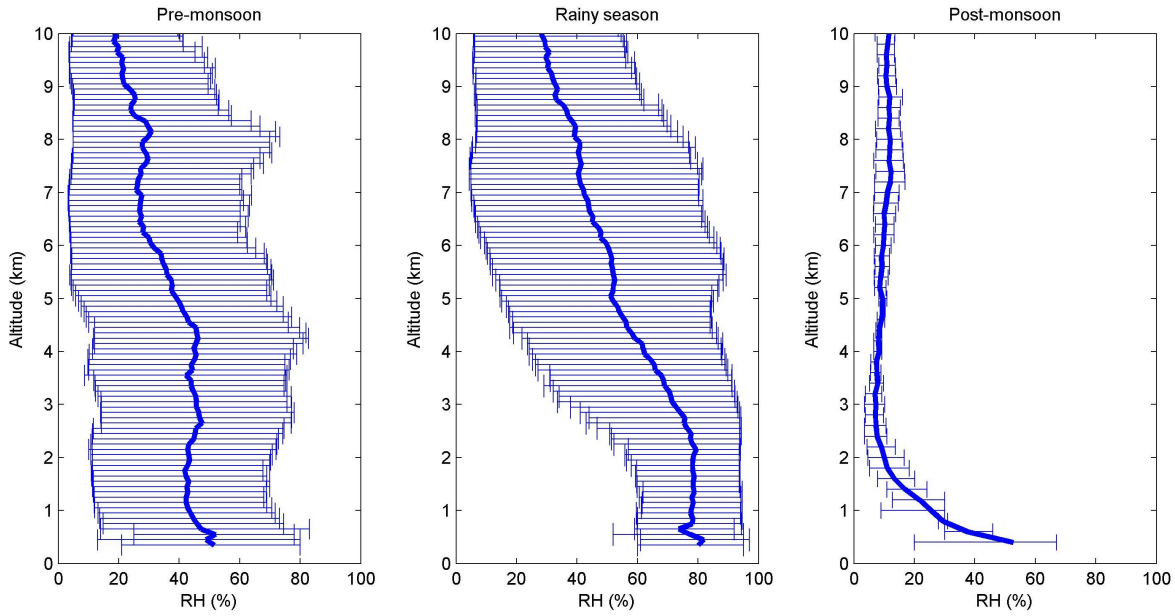

Fig. 13. Seasonal averaged RH-profiles from soundings at New Delhi airport during 2008 for the same time periods the lidar data was available. The 10th and 90th percentiles are also shown.

4 Conclusions

A 4-yr data set from Mukteshwar, Indian Himalayas, and a 2-yr data set from Gual Pahari, Indo-Gangetic plains, was utilized to study the variation of aerosol physical and optical properties before, during and after the monsoon. We observed that at both stations, the average monsoon aerosol concentrations were smaller by 40–75 % compared to the pre-monsoon average concentrations, decreasing with increasing total local rainfall during the monsoon season.

In Mukteshwar, there were two major wet removal mechanisms: aerosol scavenging by falling rain drops and activation to cloud and mountain fog droplets. The former mechanism removed other than accumulation mode particles effectively, while the latter mechanism was especially effective in removing the accumulation mode and bigger particles. Thus, the monsoon removed particles in all size classes. In Gual Pahari, the main removal mechanism was scavenging by rain. The accumulation mode was less effectively removed than the smaller and larger particles. Consequently, in Mukteshwar, black carbon (being mostly in the accumulation mode) decreased effectively during the monsoon season, resulting in decreased absorption over scattering. In Gual Pahari, accumulation mode BC was less effectively removed. Thus, the single scattering albedo increased in Mukteshwar during the monsoon season, but decreased in Gual Pahari.

In addition to the loss processes, aerosol concentrations during the monsoon were affected by sources. In Gual Pahari, new particle formation was observed during the pre-monsoon and monsoon seasons. In both locations, mineral dust was observed. This resulted in an elevated PM_{10} concentration at both stations (Hyvärinen et al., 2011), and a dominating volume mode at 3–4 µm observed with the APS in Gual Pahari. The occurrence of mineral dust was further evinced by an increased scattering coefficient at both stations, and a refractive index ∼1.5 was observed with the Gual Pahari sunphotometer. The contribution of mineral dust during these months has been reported also previously (e.g., Dey and Tripathi, 2008; Gautam et al., 2009b, 2011; Ram et al., 2008, 2010). It is likely that the relatively high absorption coefficients in Gual Pahari (and corresponding low SSA values) during the monsoon season were partly caused by the pronounced anthropogenic combustion sources in the area.

The aerosol columnar properties, which were measured in Gual Pahari, had an annual maximum in the early monsoon season, which is in contradiction with the surface measurements. We concluded two likely reasons for this: (1) Columnar measurements were from the ambient aerosol (grown by water vapour), while the in situ measurements sampled dry aerosol. (2) The early monsoon dust events arrived from a higher altitude as indicated by the maximum extinction with the Lidar at around 1 km altitude.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys.net/11/8283/2011/acp-11-8283-2011-supplement.pdf.

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