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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 during 2006–2010. The first measurement station was a background site in Mukteshwar, about 350 km northeast of New Delhi, in the foothills of the Indian Himalayas.

- ⁵ The second measurement site was located in Gual Pahari, about 25 km south of New Delhi. 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. Also the mean aerosol size decreased during the monsoon season. The size distribution at Mukteshwar was unimodal, with
- ¹⁰ a mode diameter at about 80 nm. In Gual Pahari, the ratio of Aitken and accumulation particle concentration was >1, due to wet deposition and new particle formation during the monsoon season. 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 around 3–4 μ m. The single scattering albedo varied from 0.73 to
- 0.93 during the monsoon season, being slightly lower in Gual Pahari than in Mukteshwar. The aerosol columnar properties, which were measured in Gual Pahari, showed a somewhat different seasonal behavior compared to the surface measurements, with the aerosol optical depth increasing to an annual maximum in the early monsoon season.

20 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 built 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 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.
- ²⁰ 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 modeling studies



of aerosol-monsoon interactions, and provides information of the aerosol properties during the monsoon.

2 Measurement sites and methods

2.1 Measurement locations

⁵ The measurement locations were introduced in detail in the companion paper (Hyvärinen et al., 2011). The measurement station in Mukteshwar about 350 km northeast of New Delhi is a background site, in a rural region at the Indian Himalayas. The Gual Pahari site was located in Gurgaon, about 25 km south of New Delhi.

2.2 Instruments

- The parameters measured at Mukteshwar included the particle number size distribution from 10 nm to 800 nm, particle mass concentration (<2.5 μm and <10 μm of particle aerodynamic diameter), aerosol scattering coefficient, black carbon concentration (absorption coefficient) and meteorological parameters including temperature, pressure, relative humidity and wind speed/direction. The measurements and instruments at the site are presented in more detail by Hyvärinen et al. (2009) and Komppula et al. (2009)</p>
- 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 the ground level and about 2 m above the roof of the station building.

In Gual Pahari, the air was sampled through three inlets. Two separate inlets with PM_{2.5} and PM₁₀ cut-off were used for sampling of the respective aerosol mass concentrations. The measurements which were conducted from the main inlet with a PM₁₀ cut-off were: particle number size distribution over the diameter range 4 nm–10 μm, aerosol black carbon concentration (absorption coefficient) and aerosol scattering coefficient. Meteorological parameter measured were: temperature, pressure, relative



humidity, wind speed/direction, and rain intensity/accumulation. 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 the 2008 (see Komppula et al., 2010) and a Cimel sunphotometer during the 2009 monsoon seasons. Cimel measurements were made as a part of the Aerosol Robotic Network, AERONET (e.g., Holben et al., 1998). In addition aerosol sampling for chemical characterization was performed during the 2008 monsoon season.

Data processing was explained in detail in the companion paper (Hyvärinen et al., 2011).

10 3 Results

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The monsoon characteristics were presented in detail in the companion paper (Hyvärinen et al., 2011). During the four measurement years there were contrasting monsoon seasons; with both excessive rain and draught. Rainfall was more intensive in Mukteshwar than in Gual Pahari, probably due to the mountain location. 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 (Tyagi et al., 2009). The weakest

monsoon occurred in 2006. The boundary layer height was identified as the main reason for the different annual

and diurnal variations of aerosol properties at these two stations (Raatikainen et al.,

20 2011). The explanation was based on mixing of clean air masses in Mukteshwar with polluted air masses from the plains below. This occurred, when the boundary layer was lifted above the Mukteshwar altitude (2180 m a.s.l.). During the monsoon season, the monthly average of boundary layer maximum heights did not exceed 1500 m (Fig. 1), so Mukteshwar resided mostly in the free troposphere.



3.1 Aerosol size distribution

The average number size distribution during the pre-monsoon, monsoon, and postmonsoon seasons at Mukteshwar are presented in Fig. 2. In Mukteshwar, the number size distribution was unimodal at all times. The total particle concentrations decreased

- ⁵ by about 50–75% from the pre-monsoon to the monsoon season (Table 1), depending on the amount of rainfall. 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 ratio of Aitken-mode particles (25–75 nm) to accumulation mode (75–800 nm) particles, $N_{\rm ait}/N_{\rm acc}$ increased from the pre-monsoon average of 0.58 to 0.65. This illustrates that the accumulation mode particles were more effectively removed than the Aitken mode particles by wet deposition, owing to the larger size. In Mukteshwar, the particle number concentration in the nucleation mode was very low. During post-monsoon the average value for the mode diameter was ~108 nm and for $N_{\rm ait}/N_{\rm acc}$ 0.50.
- ¹⁵ Due to data coverage issues, a similar figure for the Gual Pahari observations is not available. Instead, Fig. 3 illustrates how the modal number concentrations change during the year. At the end of the pre-monsoon season (June), the accumulation mode concentration decreased. The Aitken mode concentration stayed relatively high, with the N_{ait}/N_{acc} ratio above unity during the rainy season. This behavior was similar as in Mukteshwar. The nucleation mode, however, behaved differently than in Mukteshwar, showing decreased, but still comparable concentrations to the Aitken mode during the monsoon. A closer inspection revealed that nucleation regularly occurred in Gual Pahari (in more than 75% when daily data was available), while in Mukteshwar practically no nucleation events occurred during the monsoon season. This is most likely
- to be related to the availability of nucleation precursors. Despite the wet deposition, nucleation precursors apparently existed in Gual Pahari during the monsoon season. On the other had in Mukteshwar, the boundary layer height resided mostly below 2 km height during this time, and precursors did not reach the site (Neitola et al., 2011).



During post-monsoon, especially the accumulation mode increased rapidly in Gual Pahari resulting in $N_{\text{ait}}/N_{\text{acc}}$ ratio <1.

Volume size distributions were available from Gual Pahari APS-measurements in the aerodynamic size range of 0.4–10 μm (Fig. 4) allowing the inspection of larger par-

- ⁵ ticles. 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 μ m during pre-monsoon and ~3 μ m during monsoon. This mode is strongly related to primary particles, indicating that these were a prominent source during these seasons. The occurrence of the coarse mode also illustrates that sources are an impor-
- tant factor affecting the variability of aerosols during monsoon, not only the wthe loss processes related to wet deposition. The contribution of mineral dust during summer months has been reported also previously (e.g., Dey and Tripathi, 2008). 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.

15 3.2 Aerosol scattering and absorption coefficient

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The optical properties (Figs. 5 and 6) generally followed the seasonal behavior of concentration levels decreasing between 40–75% from the pre-monsoon average during monsoon (Table 1). Similarly to PM and BC concentrations presented in the companion paper (Hyvärinen et al., 2011), the decrease was linearly dependent on the total rain accumulation.

In Mukteshwar, the scattering coefficient was highly variable during the monsoon season, and decreased less than the absorption coefficient. During post-monsoon, a clear difference between the years was seen at the two stations – at Mukteshwar, the concentrations decreased compared to pre-monsoon, while in Gual Pahari a substan-

tial increase was observed. This is related to the boundary layer evolution (Fig. 1), as most of the pollution was confined below the altitude of Mukteshwar. This is studied in more detail by Raatikainen et al. (2011).



We calculated the single scattering albedo, SSA from the scattering and absorption measurements from both stations. From Mukteshwar, the absorption at the Nephelometer wavelength of 525 nm was obtained directly from the Aethalometer channel 520 nm. However, at Gual Pahari, the MAAP measured the absorption at 637 nm (Müller et al., 2010). We converted this absorption coefficient to match 520 nm by as-5 suming 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). It 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 using values between 1 and 10 2. The effect on SSA was less than 5%. In Mukteshwar, the single scattering albedo had similar values of 0.75–0.90 during the monsoon season as during the pre- and post-monsoon seasons (Fig. 7). Some episodes, however, showed decreased SSA of about 0.70, indicating highly absorbing aerosol or the fact that loss processes favored

- ¹⁵ particles that scatter light. The SSA in Gual Pahari showed slightly lower values than in Mukteshwar, especially during the pre-monsoon, varying from 0.73 to 0.93. 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 similarly to Mukteshwar, the dom-
- inance of absorbing particles. 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. It's notable that even though the absolute concentrations showed a different annual variation at the stations, the properties of aerosols were substantially similar.

25 3.3 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. During the monsoon season the average AOD increased to about 0.70. It is noted that these are day-time values.

- The aerosol columnar properties show a different seasonal behavior compared to the surface measurements, with the AOD having a maximum in the early monsoon season (Fig. 8). 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. 9). 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-
- ¹⁰ 2 km, and decreased monotonically with altitude. During post-monsoon season, a lowlevel but thick aerosol layer was observed. This corresponds well with the boundary layer height (Fig. 1), 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.
- ¹⁵ 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
- ²⁰ compared to the average values (Kusuma et al., 1991). However, comparing the in situ concentrations only when AOD was measured did not solve the difference in seasonal trends. 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
- ²⁵ altitude. However, the AODs during the early monsoon showed much higher values than during the mineral-dust laden pre-monsoon season, so this can be only half of the explanation.

Relative humidity may provide additional explanation in the observed discrepancies, as it affects the particle size. 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. 10). (University of Wyoming, department of atmospheric research, http://weather.uwyo.edu/upperair/sounding.html). It can be seen that the profiles are very similar, and the RH dependence of particle size could well contribute to the discrepancy between surface in situ and columnar measurements. However, to quantify the effect is difficult without state-of-the-art instruments such as the hygroscopic tan dem DMA or a humidified nephelometer.

4 Conclusions

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A 4-year data set from Mukteshwar, Indian Himalayas, and a 2-year data set from Gual Pahari, Indo-Gangetic plains, were 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, with a linear relationship with the total local rainfall during the monsoon season.

The monsoon rains removed particles in all size classes, with subtle differences compared to pre-monsoon and post-monsoon. During monsoon, the average geomet-

- ric diameter of the number size distribution was smaller at both stations, because the accumulation mode particles were more effectively scavenged than the Aitken mode particles. In Gual Pahari, new particle formation was observed during the monsoon season. Occasionally decreased single scattering albedos were observed together with elevated BC/PM_{2.5} fractions, especially in Gual Pahari and during the early half of
- the monsoon. This indicates that sources for absorbing material may be different than those for scattering particles, or that removal of scattering particles may at times be more effective. In addition to loss processes, aerosol concentrations during the early



monsoon were found to be affected by primary emissions, most likely dust from the Thar Desert. This resulted in an elevated coarse mode at both stations and a dominating volume mode at 3-4 µm observed with the APS in Gual Pahari (Hyvärinen et al., 2011). The contribution of mineral dust during summer has been reported also ⁵ previously (e.g., Dey and Tripathi, 2008).

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 vapor), 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.

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Table 1. Difference between the monsoon and pre-monsoon aerosol concentrations in Mukteshwar and Gual Pahari.

Year	Abs coeff.	Scat coeff.	N _{tot}	V _{tot} APS
	Mm ⁻¹	Mm ⁻¹	#/cm ⁻³	μm ³ cm ⁻³
Mukteshwar	520 nm	525 nm		
2006 Pre/Monsoon	15.3/5.2	62.5/29.8	3250/1430	-
% diff	–65.6	-52.3	–56.1	
2007 Pre/Monsoon	18.2/6.3	68.2/40.1	4060/1870	-
% diff	–65.4	–41.3	-54.0	
2008 Pre/Monsoon	17.7/4.9	94.4/24.2	4870/1350	-
% diff	–71.7	–74.4	–72.3	
2009 Pre/Monsoon	18.1/-	90.8/52.3	4510/2290	-
% diff	-	-42.4	–49.3	
Gual Pahari	637 nm	520 nm		
2008 Pre/Monsoon % diff	55.3/— —	-	-	68.0/- -
2009 Pre/Monsoon	53.0/24.0	229.7/-	-	44.1/25.7
% diff	-54.6	-		–41.8





Fig. 1. Annual variation for the averaged daily maximum boundary layer heights (PBL) obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF).





Fig. 2. Average size distributions for different seasons in Mukteshwar. Solid lines are averages and dotted lines represent the 10th and 90th percentiles.











Fig. 4. The volume size distribution in Gual Pahari during different seasons. Solid lines are averages and dotted lines represent the 10th and 90th percentiles.































Fig. 9. 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. 10. Seasonal averaged RH- profiles from soundings at New Delhi airport during 2008 for the same time periods the lidar data was available. The 10 and 90 percentiles are also shown.

