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Aerosol characteristics during winter fog at Agra, North India

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Abstract Simultaneous measurements on physical, chemical and optical properties of aerosols over a tropical semi-arid location, Agra in north India, were undertaken during December 2004. The average concentration of total suspended particulates (TSP) increased by about 1.4 times during intense foggy/hazy days. Concentrations of SO_4^{2-}, NO_3^-, NH_4^+ and Black Carbon (BC) aerosols increased by 4, 2, 3.5 and 1.7 times, respectively during that period. Aerosols were acidic during intense foggy/hazy days but the fog water showed alkaline nature, mainly due to the neutralizing capacity of NH_4 aerosols. Trajectory analyses showed that air masses were predominantly from NW direction, which might be responsible for transport of BC from distant and surrounding local sources. Diurnal variation of BC on all days showed a morning and an evening peak that were related to domestic cooking and vehicular emissions, apart from boundary layer changes. OPAC (Optical properties of aerosols and clouds) model was used to compute the optical properties of aerosols. Both OPAC-derived and observed aerosol optical depth (AOD) values showed spectral variation with high loadings in the short wavelengths (<1 μm). AOD value at 0.5 μm wavelength was significantly high during intense foggy/hazy days (1.22) than during clear sky or less foggy/hazy days (0.63). OPAC-derived Single scattering albedo (SSA) was 0.84 during the observational period, indicating significant contribution of absorbing aerosols. However, the BC mass fraction to TSP increased by only 1% during intense foggy/hazy days and thereby did not show any impact on SSA during that period. A large increase was observed in the shortwave (SW) atmospheric (ATM) forcing during intense foggy/hazy days (+75.8 W/m^2) than that during clear sky or less foggy/hazy days (+38 W/m^2), mainly due to increase in absorbing aerosols. Whereas SW forcing at surface (SUF) increased from −40 W/m^2 during clear sky or less foggy/hazy days to −76 W/m^2 during intense foggy/hazy days, mainly due to the scattering aerosols like SO_4^{2-}.

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1 Introduction

Aerosols influence the attenuation of solar radiation by scattering and absorption, depending upon their size as well as chemical composition. Submicron size aerosols have a potential role in the formation of fog and haze at any location (Lillis et al. 1999). Many parts of Northern India experience severe fog and haze during the winter season that frequently lead to the disruption of road and air traffic. In the Indo-Gangetic Basin (IGB) region, generally the prevailing low temperature and high humidity conditions, along with the availability of abundance of nuclei due to different anthropogenic activities; are favorable for the formation of fog and haze in the winter season. This region is one of the densely populated and very fertile areas of the world and is reported to be the significantly potential source for various types of pollutant species, especially those containing compounds of sulfur, nitrogen and carbon (Habib et al. 2006; Garg et al. 2001). These are mainly emitted by different anthropogenic activities carried out in this region, including biomass/biofuel burning for residential cooking, for agricultural purposes and fossil fuel burning by vehicular sources. Satellite observations have reported very high aerosol loadings over this region. The NASA-Terra MODIS satellite data reveal an extensive layer of aerosols covering the entire IGB and Himalayan foothills region with seasonal mean AODs of about 0.4 to 0.5 in the visible wavelengths (0.55 micron), which are among the largest seasonal mean dry season AODs for the tropics. (Ramanathan and Ramana 2005). The Indian Ocean experiment (INDOEX) which was conducted over the tropical Indian ocean during the winter season of 1998 and pre monsoon season of 1999, revealed the vital role of emissions (especially Black Carbon i.e. BC) from SE Asian regions (Neusuβ et al. 2002) and major contribution to it was attributed from fossil fuel burning (Novakov et al. 2000).

In December 2004, a month long land campaign (LC-II) was undertaken in the IGB region, covering eight different stations from Hissar in northwest India to Kharagpur in northeast India. The main aim of the campaign was to monitor the characteristics of aerosols and variations of major trace gases and to understand the impact of pollutant species on the prolonged foggy/hazy conditions experienced in north India, during winter season. Some important results relating to aerosol characteristics have been reported, using the data collected during the LC-II campaign (Tripathi et al. 2006; Tare et al. 2006; Ganguly et al. 2006; Ramachandran et al. 2006; Pant et al. 2006; Niranjan et al. 2006). In the present study, collocated measurements were undertaken on TSP, BC and aerosol optical depth (AOD) at Agra during December 2004. Depending upon the observed surface meteorological parameters and visual observations of visibility, the observational period was separated into two periods i.e. intense foggy/hazy days (11 to 13 and 18 to 25 December) and clear sky or less foggy /hazy days (1 to 10, 14 to 17 and 26 to 31 December).

Using the chemical composition of TSP and BC values in OPAC (Optical Properties of Aerosols and clouds) model, optical properties like AOD, SSA and asymmetry factor were computed, which were further used in SBDART (Santa Barbara Distort Aerosol Radiatite Transfer) model to compute the aerosol forcing at surface and top of the atmosphere at Agra. In this paper, we discuss the observed changes in the physical, chemical and optical characteristics of aerosols, especially in relation with fog and haze.

2 Description of the measurement site

Observations were conducted in the campus of Dayalbagh Educational Institute (DEI) in Agra (27° 10′ N, 78° 05′E, 169 m amsl), during 1 to 31 December 2004. Observations were carried out on the roof of the building, about 8 m above the ground. Figure 1 shows the location of Agra and the sampling site. Two third of surrounding regions of Agra are bounded by the arid regions of Thar Desert of Rajasthan. The soils are a mixture of sand and loam with excess amounts of salts. There are many industries, like ferrous and non-ferrous metal casting, rubber processing, tanneries, brick kilns, etc. The Mathura oil refinery is about 40 km to the NW of the study site and lie in the upwind direction as winds are mostly from W and NW sector during the winter season. In addition, the industrial areas of New Delhi lie in the NW, at about 250 km. Also, the majority of foundries and Ferozabad Glass- industries are at the distance of about 20 km to the E/NE. A road lies near the sampling site, where light traffic is observed and national highway (NH-11) is at about 2 km to the south, where traffic activity is more.

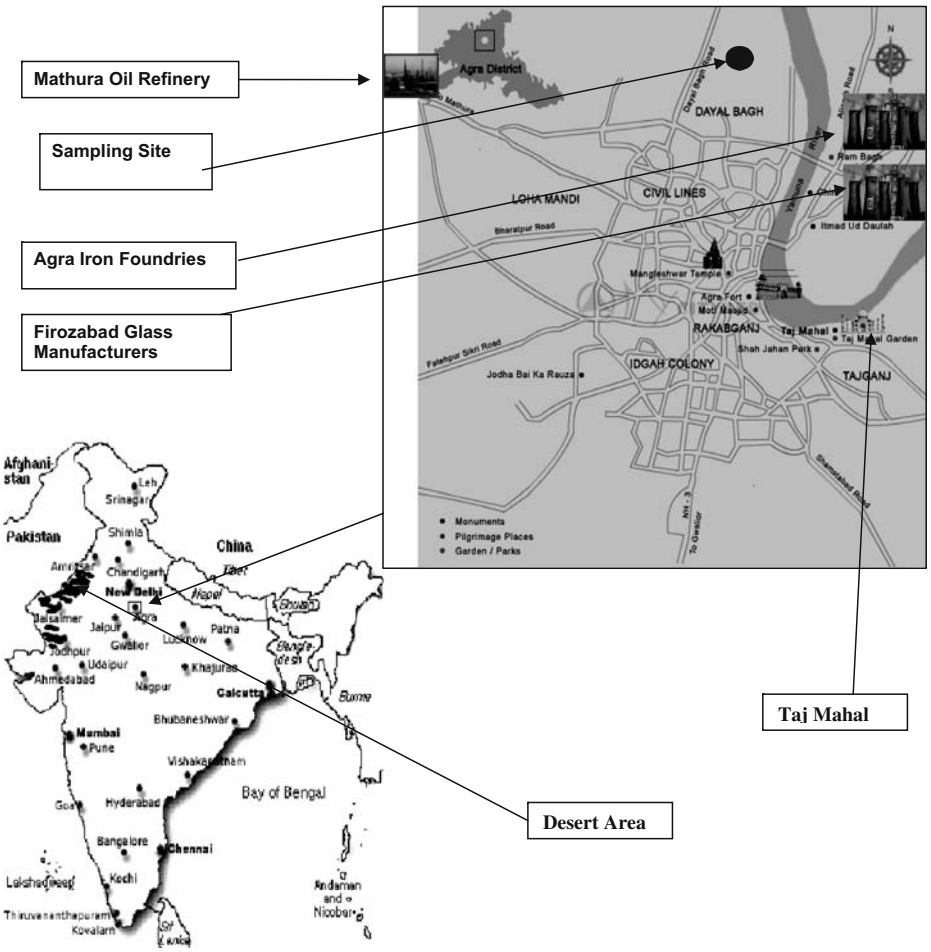


Fig. 1 Location of Agra and sampling site, along with surrounding major industries

3 Sampling and analysis techniques

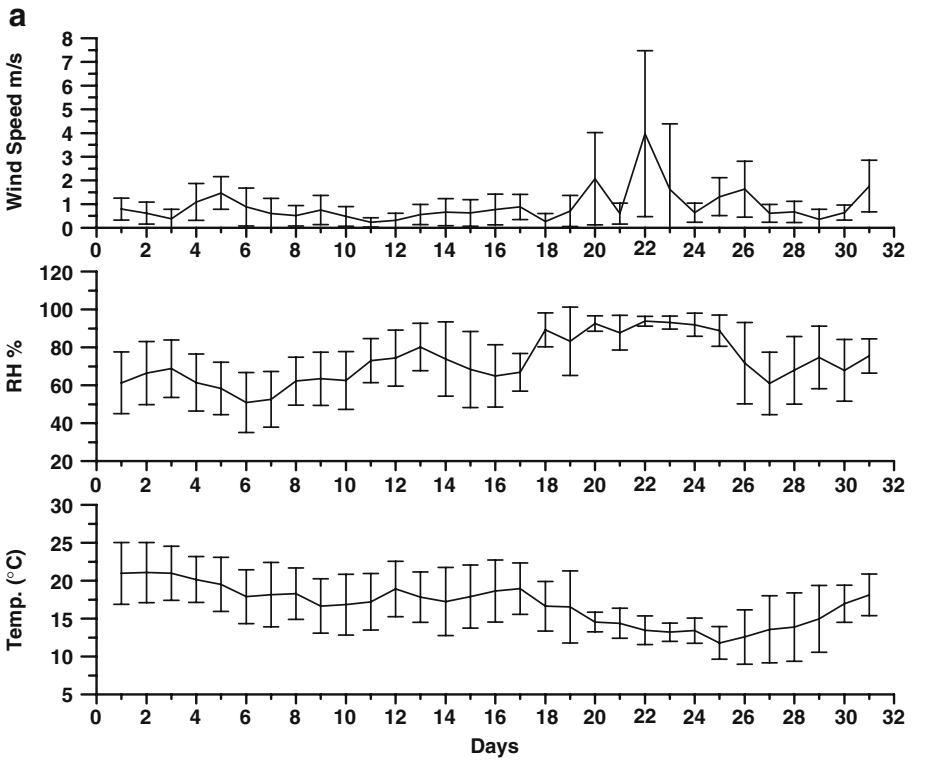
Samples of TSP were collected, during daytime period (0900 to 1700 hrs), using a High Volume Air Sampler (Envirotech Model 110). A few TSP samples were collected in the night too, during the first 2 weeks of December 2004, until the commencement of intense foggy/ hazy period. Whatman 41 filter papers with 8×10 inch size were used for the collection and the average rate of the air flow varied from 1 to 1.2 m^3/min. The filter papers were equilibrated for humidity, before and after weighing. After sampling, all the filter papers were extracted for chemical analysis. Analysis of anions (Cl^-, SO_4^{2-} and NO_3^-) was carried out with the help of Ion Chromatograph (Dionex 100), using analytical columns Ion Pac-AS4A- SC 4 mm, anion micro membrane suppressor ASRS -1, 1.8 mM Sodium Carbonate/1.7 mM Bicarbonate as eluent and triple distilled water as regenerant. Atomic Absorption Spectrophotometer (Perkin Elmer 373) was used for the analysis of cations (Na^+, K^+, Ca^{2+} and Mg^{2+}) and NH_4^+ was analyzed using spectrophotometric method (Weatherburn 1967). The detection limit for ion chromatographic analysis was about 0.02 ppm whereas, that for the atomic absorption spectrophotometric analysis varied from 0.0002 to 0.02 ppm. Field Blanks were taken and analyzed; using procedures similar to those adopted for samples and were found to be within the statistical limits. Detailed procedures of sampling and analysis techniques have been mentioned elsewhere (Safai 1999). Additionally, fog water samples were collected on five occasions, using a fog sampler. An active collector maintained at 15°C, inside a stainless steel cooling chamber was used for collection of fog droplets through the impaction technique. Details of this equipment are described elsewhere (Khemani et al. 1987; Ali et al. 2004).

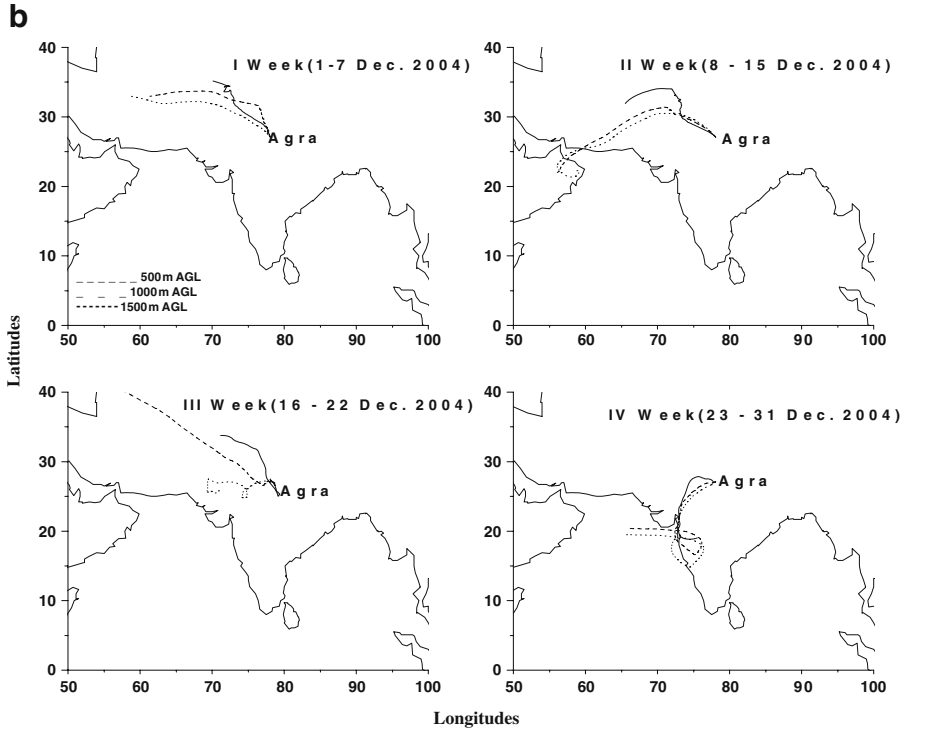
Continuous measurements of BC aerosols were carried out, using an Aethalometer (Magee Sci., INC, USA, and MODEL AE- 42). Measurements at 5 min interval were carried out at a flow rate of about 3 L/min. The instrument was calibrated by the manufacturer only about 6 months before the observational campaign, and uncertainty in measurements was reported up to ±2%. AOD measurements were carried out during clear sky conditions, using a multi-channel sun photometer (MICROTOPS II, Solar Light Co., USA), at six discrete wavelengths viz. 380, 440, 500, 675, 870 and 1,020 nm. The absolute uncertainty in derived AOD is less than 0.03 at all the wavelengths. AOD values were derived from measurements of direct solar irradiance, using a set of calibration constants for each wavelength. The detailed working principle and calibration procedure can be found elsewhere (Morys et al. 2001).

4 Meteorological conditions

Figure 2a shows the average day-to-day variation of meteorological parameters at Agra, monitored by using an automatic weather station during the campaign. Average temperature during intense foggy/hazy days (15.2±2.5°C) was about 2.5°C less than that during clear sky or less foggy/hazy days (17.7±3.8°C). Also, the average relative humidity (RH) during intense foggy /hazy days (86.2±8.5%) was about 21% more than that during clear sky or less foggy/hazy days (65±14.9%). Average wind speed (0.8 to 1.1 m/s) during all the days showed near calm conditions, which was conducive for the low ventilation of pollutants. Surface winds were predominantly from N/NW direction. It was observed from

Fig. 2 **a** Average day-to-day variation of meteorological parameters at Agra during December 2004. **b** Air mass back-trajectories at Agra in December 2004 (NOAA Hysplit Model) ▶


a
Wind Speed m/s
RH %
Temp. (°C)
Days


b
I Week(1-7 Dec. 2004)
II Week(8 - 15 Dec. 2004)
III Week(16 - 22 Dec. 2004)
IV Week(23 - 31 Dec. 2004)
Agra
500m AGL
1000m AGL
1500m AGL
Latitudes
Longitudes

meteorological data, reported by the Indian Daily Weather Report, (IDWR 2004) that average minimum temperature (8°C) was below the average dew point temperature (8.3°C) recorded at Agra, during intense foggy/hazy days. Also, the average visibility recorded was less than 500 m during these days (visual observations indicated that many times it was less than 50 m).

Air-mass trajectories for seven-day period at 500, 1,000 and 1,500 m above ground level were plotted using the Hybrid Single-Particle Lagrangian Integrated Trajectory Model (Draxler and Rolph 2003). Figure 2b shows the weekly variation of these trajectories at Agra.

5 Results and discussion

5.1 Day-to-day variation of TSP and BC

Figure 3 shows the day-to-day variation of TSP and BC concentrations at Agra. Concentration of TSP varied from 352 to 685 μg/m^3 during intense foggy/hazy days with an average of 475±101 μg/m^3, whereas it varied from 262 to 391 μg/m^3 during clear sky or less foggy/hazy days with an average of 338±38 μg/m^3. Concentration of TSP showed about 1.4 times increase during intense foggy/hazy days, which was significant at 95% confidence level. Similarly, concentrations of TSP showed an increase during hazy and foggy period at Hisar (Ramachandran et al. 2006), which is ascribed to the presence of fog combined with higher relative humidity. Enhancement in PM_{10} concentrations at Kanpur was reported (Tare et al. 2006) during foggy/hazy days (231.6±43.3 μg/m^3) as compared to those during clear days (175.1±33.1 μg/m^3).

The average BC concentrations at Agra during intense foggy/hazy days (17.34±6.58 μg/m^3) showed about 1.7 times increase as compared to those observed during clear sky or less foggy/hazy days (10.45±3.94 μg/m^3). The BC concentrations observed at Agra, compared well with other reported observations in the IGB region. Average BC concentration was reported to be 29±14 μg/m^3 with variation of 11 to 65 μg/m^3 during December 2004 at Delhi (Ganguly et al. 2006), which is about 140 km to the NE of Agra and where meteorological conditions are shown to be more or less similar to those at Agra. BC concentrations in the range of 6 to 20 μg/m^3 have been reported at Kanpur during December 2004 (Tripathi et al. 2006 and Tare et al. 2006), with more concentrations during foggy/hazy days (14.3±2.7 μg/m^3) as compared to those during clear days (9.8±3 μg/m^3). Similarly, increase in BC con-

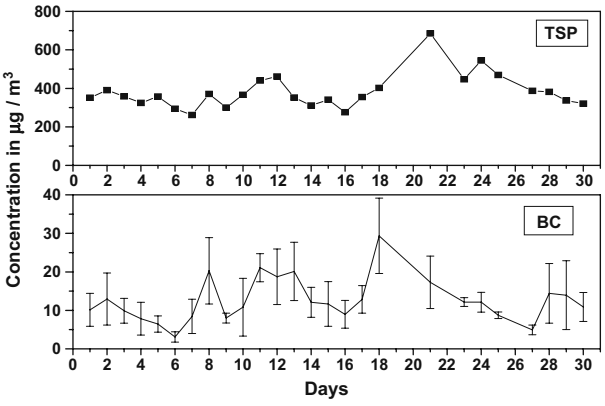


Fig. 3 Day-to-day variation of BC and TSP concentrations at Agra in December 2004

centrations during foggy/hazy days has been reported at Kharagpur (Niranjan et al. 2006) and Hisar (Ramachandran et al. 2006).

BC mass fraction to TSP was observed to be between 3 to 4% at Agra. There was only 1% increase in BC mass fraction to TSP, even though the average concentration of BC increased by about 65% during the intense foggy/hazy days. About 1 to 4% BC fraction to TSP was reported at Hisar (Ramachandran et al. 2006). However, BC mass fraction was about 10% to TSP at Kanpur (Tripathi et al. 2006) and that at Delhi was about 4 to 15% (Ganguly et al. 2006). As stated earlier, in the present study, a few TSP samples were collected during the nighttime in the first 2 weeks of December. It was observed that the percentage contribution of BC to TSP during nighttime was significant (about 9%). One of the reasons for the comparatively more mass fraction of BC to TSP was the observed high BC concentrations during nighttime i.e. 1800 to 0600 hrs (21.7±8.7 μg/m^3 and 24.7± 9.4 μg/m^3 during clear sky or less foggy/hazy days and intense foggy/hazy days, respectively) than those during daytime i.e. 0600 to 1800 hrs (14.1±7.5 μg/m^3 and 16.7± 4.8 μg/m^3 during clear sky or less foggy/hazy days and intense foggy/hazy days, respectively). The average day/night BC ratio was about 0.65. More BC concentrations during nighttime are attributed to the increase in bio-fuel burning in the nearby surroundings during evening and night hours, especially for cooking as well as for room heating; or burning of grass, dry leaves, etc for getting warmth against cold weather. At the same time, average TSP concentration decreased during nighttime due to low frequency and intensity of other human activities that generally occur during the daytime.

From the air mass back trajectories, it was observed that during the first 3 weeks, north-westerly (NW) winds were predominant at Agra (Fig. 2b). Air masses traveled from as far as the Mediterranean region, crossing Iran, Afghanistan and Pakistan, sometimes from Gulf coast. The average BC concentration during this period was 19.3±9.7 μg /m^3. However, during the fourth week, winds were mostly from S/SW direction and during this week, the average BC concentration reduced up to 16.6±8.0 μg /m^3. This indicates towards the possibility of long range transport of BC from some potential source region in the NW direction of the sampling location. High BC concentrations were reported at Lahore (which lays to the upwind of Agra in NW), ranging from 5 to 110 μg/m^{3}during winter season of 2005; which were attributed to both fossil fuel combustion as well as biomass burning (Husain et al. 2007). In addition, there are certain other local sources in the surrounding areas of Agra including Mathura oil refinery and some other industrials installations in New Delhi. A phenomenon termed as Western disturbances, is reported to be the major reason for the occurrence of dust haze that spreads from West Asia into Pakistan and northwestern India, reducing the visibility in the northwest Indian regions, during winter season (Pasricha et al. 2003). During this period, the vertical lifting and dispersal of pollutants are virtually absent. There were about 8 Western disturbances during December 2004 (IDWR 2004), generated over Afghanistan and adjoining Pakistan, during regular interval from 1 to 30 December, which moved subsequently to Rajasthan and then to east north eastwards. Due to these prevailing conditions, there might be reduction in low-level inversion heights. Due to lack of availability of related altitude profiles of temperature and RH at Agra, we could not directly relate the effects of Western disturbances to change in inversion layers. However, at Delhi (which is about 250 km to the NW of Agra and where the prevailing meteorology can be considered to be similar to that at Agra), lidar observations revealed that during fog events, there was a subsidence of aerosols to an extremely dense and shallow atmospheric layer of <200 m height from surface (Ganguly et al. 2006). The intense foggy /hazy days reported by them do match with those considered at Agra, i.e. 11 to 13 and 18 to 25 December 2004. The transport from other regions along with the

lowering of inversion layer and thereby stagnation conditions together might have led to the observed increase of BC during intense foggy/hazy days at Agra.

5.2 Chemical composition of TSP

Figure 4a shows the average concentrations of chemical components of TSP. All the chemical components showed elevated concentrations during intense foggy/hazy days (Table 1). However, concentrations of SO_4^{2-}, NH_4^+, and NO_3^- showed more increase (4 times increase for SO_4^{2-}, 3.5 times for NH_4^+, and 2 times for NO_3^-), which was significant at 95% confidence level. Generally, these aerosols exist in the fine size due to their formation through gas to particle mechanism (Milford and Davidson 1987). Studies on chemistry of size separated aerosols at Agra (Ranjit Kumar et al. 2007) have shown significantly more contribution of fine size aerosols for SO_4^{2-}, NH_4^+, and NO_3^- at the same site in Agra. However, NO_3^- can exist significantly in coarse size too, through the reaction of nitric acid vapour with coarse sized sea-salt or crustal components (Wolff 1984; Li and Shao 2009). Parmar et al (2001) have reported coarse size NO_3^- at Agra. Even though the concentrations of aerosols that are generally observed in coarse size have also shown increase; it was comparatively less, i.e. Ca^{2+}, K^+, Mg^{2+}, Na^+ and Cl^- showed increase by about 1.2 to 1.6 times, during intense foggy/hazy days. Significantly high concentrations of NH_4^+ and NO_3^-, during foggy/hazy days at Kanpur were attributed to the increase in burning activity (Tare et al. 2006).

Nitrate formed more fraction of TSP (about 7% during clear sky or less foggy /hazy days and 10% during intense foggy/hazy days). It was followed by SO_4^{2-} during intense foggy/hazy days (7%) whereas, during clear sky or less foggy/hazy days, BC showed next highest percentage contribution to TSP (3.1%) and SO_4^{2-} followed it (2.6%). Concentrations of the secondary aerosols formed by their gaseous precursors (e.g. SO_4^{2-}, NO_3^-, and NH_4^+ aerosols from SO_2, NO_2 and NH_3, gases, respectively) were more due to the aqueous heterogeneous reactions occurring in the atmosphere, associated with high relative humidity; particularly during intense foggy/hazy days. In addition, there were other influencing factors such as, low ventilation coefficients as well as more domestic fossil fuel burning incidences. Enhanced concentrations of SO_2, NO_2 and NH_3 at the same location in Agra during winter have been reported earlier, which were attributed to high fuel usage and persistent inversion conditions (Ranjit Kumar et al. 2004).

BC, SO_4^{2-}, NO_3^- and NH_4^+ showed more or less similar feature in day-to-day variation (Fig. 4b). In addition, BC showed good correlation with SO_4^{2-} (r=0.50, p=0.01) and NO_3^- (r=0.54, p=0.004). This indicates towards the common source for these constituents of aerosols such as combustion related activities like biomass/fossil fuel burning.

5.3 Acidity of aerosols

To check the effect of foggy and hazy conditions on the acidity of aerosols, we have examined the ionic balance and ratio of neutralization to acidic potential of aerosol chemical composition. The ionic balance (Total cations—Total anions or $\Delta\Sigma = \Sigma^+ - \Sigma^-$) is one of the widely used criteria to test the nature of aerosols i.e. if this balance is positive, then cations (the alkaline components e.g. NH_4^+, Na^+, K^+, Ca^{2+}, and Mg^{2+}) are dominating, which infers towards alkalinity. Whereas, if the ionic balance is negative, then

Fig. 4 **a** Average chemical composition of TSP at Agra during December 2004. **b**. Day-to-day variation of BC, SO_4^{2-}, NO_3^- and NH_4^+ at Agra during December 2004 ▶

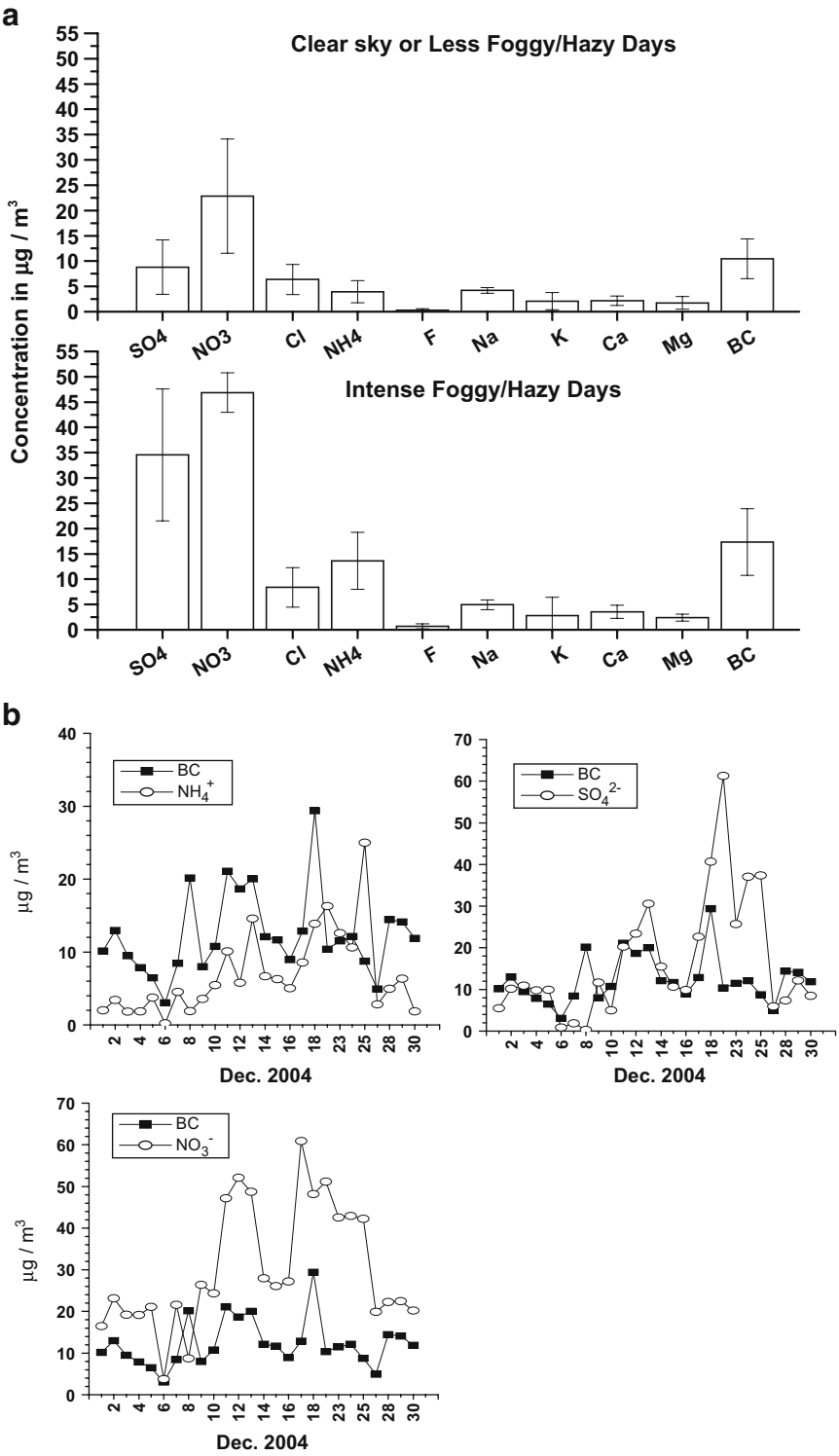

a
Clear sky or Less Foggy/Hazy Days
Concentration in µg / m3
SO4
NO3
Cl
NH4
F
Na
K
Ca
Mg
BC
Intense Foggy/Hazy Days
b
BC
NH4+
µg / m3
Dec. 2004
BC
SO4 2-
Dec. 2004
BC
NO3-
µg / m3
Dec. 2004

Table 1 Chemical composition of TSP along with BC concentrations (μg/m^3) during intense foggy/hazy days and clear sky or less foggy/hazy days at Agra during December 2004

Component	Intense foggy/hazy days		Clear sky or less foggy/hazy days	
	Mean	SD	Mean	SD
SO_4^{2-}	34.55	13.02	8.81	5.36
NO_3^-	46.87	3.90	22.83	11.3
Cl^-	8.39	3.90	6.36	2.98
NH_4^+	13.61	5.63	3.94	2.19
F^-	0.70	0.50	0.28	0.30
Na^+	4.95	0.95	4.20	0.58
K^+	2.80	3.64	2.07	1.69
Ca^{2+}	3.55	1.30	2.16	0.95
Mg^{2+}	2.40	0.68	1.74	1.24
BC	17.35	6.58	10.45	3.94

anions (the acidic components e.g. Cl^-, SO_4^{2-}, NO_3^- and F^-) are dominating, that shows the acidic nature (Hidy and Countess 1982; Khemani 1989). At Agra, the ionic balance of aerosols was negative showing acidic nature of aerosols. However, the magnitude of acidity was more during intense foggy /hazy days ($\Delta\Sigma$=−351 μeq/L) as compared to that during clear sky or less foggy/hazy days ($\Delta\Sigma$=−47 μeq/L).

The ratio of Neutralization Potential (NP) to Acidic Potential (AP) shows the acid neutralizing capacity of aerosols. It is computed as:

$$\mathrm{NP/AP} = \left(\left[\mathrm{nss\ Ca^{2+}}\right] + \left[\mathrm{NH_4^+}\right] + \left[\mathrm{nss\ Mg^{2+}}\right] + \left[\mathrm{nss\ K^+}\right]\right) / \left(\left[\mathrm{nss\ SO_4^{2-}}\right] + \left[\mathrm{NO_3^-}\right]\right)$$

Where, nss indicates the concentration of non-sea salt fraction of the respective chemical component. The details of calculating non-sea salt fraction are given elsewhere (Safai 1999). The NP/AP ratios were 0.78 and 1.03 respectively, for the intense foggy/hazy days and clear sky or less foggy/hazy days. This indicates that NP was less than AP during intense foggy/hazy days, which means more acidic potential than neutralizing potential of aerosols. However, the neutralizing potential slightly dominated over the acidic potential during the clear sky or less foggy/hazy days, indicating alkaline nature of aerosols. The presence of acidic nature of aerosols during intense foggy/hazy days can be mainly attributed to the enhanced concentrations of two major acidic components i.e. SO_4^{2-} and NO_3^-. Combustion generated BC aerosols have been reported to play an important role in the formation of SO_4^{2-}. It acts as a catalyst and enhances the oxidation of SO_2 that quickly hydrolyses to form SO_4^{2-} (Chang et al. 1982; Brodzinsky et al. 1980; Disselkamp et al. 2000). Similar results were reported for fog water composition at Berkeley, California where the increased SO_4^{2-} concentrations in interstitial particles were attributed to acid forming oxidation reactions when fog mixed with anthropogenic pollutants like BC (Gundel et al. 1994). At Agra, it is to be noticed that concentrations of BC also increased (about 65% more) during intense foggy/hazy days, which could have helped in the formation of SO_4^{2-} and thereby catalyzed the increase in acidity of aerosols. However, this mechanism may be a minor possibility in the formation of SO_4^{2-} aerosols. SO_4^{2-} may be formed mainly in aqueous chemistry reactions, particularly during high humid conditions in intense foggy/hazy days. Conversion of S (IV) to S (VI) in the atmosphere involves aqueous chemistry through oxidation by H_2O_2, O_3, and/or O_2 (Pandis and Seinfeld 1989; Reilly et al. 2001).

The acidic nature of aerosols is not reflected in the chemistry of fog water. About six samples of fog water were collected at Agra during the campaign period. The fog water was alkaline (average pH 6.3), mainly due to the neutralizing capacity of NH_4^+ aerosols. Concentration of NH_4^+ was significantly high and its source could be related to the cattle stockyard situated close to the sampling site. The ionic balance was +156.6 μeq/L, indicating dominance of cations (especially NH_4^+ and Ca^{2+}). Also, the ratio of neutralization potential to acidic potential was 2.14, indicating complete neutralization of acidity. Alkaline nature of fog at Delhi was reported during 2000 to 2003 with major neutralization of acidity by NH_4^+ (Ali et al. 2004).

5.4 Diurnal variation of BC

Figure 5 shows the diurnal variation of BC during intense foggy/hazy days and clear sky or less foggy/hazy days. The observed morning and evening peaks during both these periods are attributed to local burning activities, especially for cooking during these hours and also due to the emissions from mobile sources, whose density was observed to be generally more during these hours. In addition to it, diurnal features in local boundary layer also play an equally important role in this feature (Allen et al. 1999; Babu and Moorthy 2002a; Husain et al 2007). Similar diurnal pattern for BC was observed at New Delhi (Ganguly et al. 2006).

However, there was a difference in the magnitude and time of occurrence of morning and evening peaks during intense foggy/hazy days and clear sky or less foggy/hazy days at Agra. While the morning peak during intense foggy/hazy days (24 μg / m^3) was observed at 1000 AM, the same was observed two hrs before at 0800 AM during clear sky or less foggy/hazy days (27.9 μg / m^3) . However, the evening peak showed opposite feature with peak values during clear sky or less foggy/hazy days (32.5 μg / m^3) occurring at 1100 PM that is two hrs after than that during the intense foggy/hazy days (37.5 μg /m^3) at 0900 PM. The late occurrence of peak BC concentrations during morning and early occurrence during evening in the intense foggy/hazy days could be due to the increase in intensity of local traffic sources during the respective time periods, as the foggy and hazy conditions

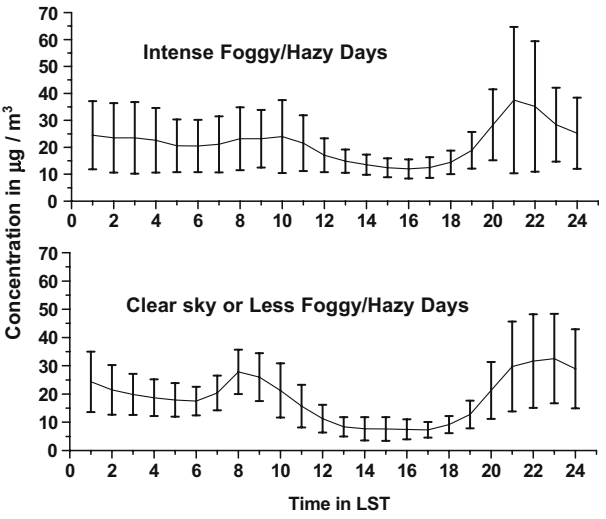


Fig. 5 Diurnal variation of BC at Agra, during December 2004

prevailed right from early evening to late morning. Also, the difference between morning peak (P1) and evening peak (P2) of BC concentration (ΔP=P2−P1) was significantly more during intense foggy/hazy days (13.5 μg/m^3) than during clear sky or less foggy and hazy days (4.6 μg/m^3).

In addition, during intense foggy/hazy days, observed values of AOD were high that might have prompted a considerable reduction in the solar heating and thereby delayed the onset of convective mixing or reduced the surface convection and the boundary layer depth and consequently the ventilation. This could be the reason for the observed prolonged morning peak of BC. Similarly, the impact of delayed/reduced solar heating during intense foggy/hazy days was observed on temperature and RH. Maximum temperature during afternoon hours in the intense foggy/hazy days was 19°C whereas RH dropped down up to 70% during that period, but during clear sky or less foggy-hazy days, maximum temperature during afternoon hours was 24°C and during that period RH decreased up to 40%.

Average BC concentrations during daytime (0600 AM to 0600 PM) were about 23% more in the intense foggy/hazy days (17.5±4.8 μg/m^3) than those in the clear sky or less foggy-hazy days (14.2±7.7 μg/m^3). Whereas, the nighttime (0600 PM to 0600 AM) BC concentrations were about 14% more in the intense foggy/hazy days (25.7±5.8 μg/m^3) as compared to those in the clear sky or less foggy-hazy days (22.6±6.0 μg/m^3). Also, the minimum concentrations, observed during the afternoon (0200 to 0500 PM) were about 65% more during intense foggy/hazy days (12.6±3.7 μg/m^3) than those during clear sky or less foggy-hazy days (7.53±3.6 μg/m^3).

5.5 Optical characteristics of aerosols

Aerosol properties are highly variable in space and time and it is rather difficult to model aerosol climatology at any location. However, an attempt is made in OPAC model, which reduces the variability of naturally occurring aerosols and clouds to typical cases, without neglecting possible fluctuations (Hess et al. 1998). This model is widely used and reported by many studies, particularly from Indian region (Babu et al. 2002; Tripathi et al. 2005; Ramachandran et al. 2006; Ganguly et al. 2006; Satheesh et al. 2006). In the present study, the chemical composition of TSP and observed soot concentrations have been used in conjunction with OPAC model to determine certain optical properties of aerosols like AOD, SSA and asymmetry factor. These parameters are essential for the radiative forcing calculations.

There are ten different aerosol models available in OPAC and we have used Continental Polluted type, consisting of three aerosol components i.e. water soluble, insoluble and soot or BC. From the TSP mass concentration and chemical composition, water soluble and insoluble components were obtained whereas soot was obtained from Aethalometer data. Keeping the measured BC mass constant, the number densities for water soluble and insoluble components of TSP were adjusted without changing the total TSP as obtained from high volume sampler, until the spectral AODs estimated from OPAC were matched with measured AODs within ±5% at common wavelengths. The average AOD values at 0.5 μm from observation and OPAC-derived methods were 0.63 and 0.62, respectively, during clear sky or less foggy/hazy days; whereas, those during intense foggy/hazy days were 1.22 and 1.21, respectively. Also, the average AOD values at 0.55 μm from OPAC and MODIS-derived methods were 0.55 and 0.52, respectively, during clear sky or less foggy-hazy days; whereas, those during intense foggy/hazy days were 1.08 and 0.95, respectively. Thus, the OPAC-derived AOD values matched well with the observed and MODIS AOD values.

5.5.1 Spectral variation of AOD

Figure 6a shows the spectral variation of OPAC-derived SSA and AOD values at Agra during intense foggy/hazy days and clear sky or less foggy/hazy days. The average AOD values at visible wavelengths (<1 μm) were 0.56 and 1.09 during clear sky or less foggy/hazy days and intense foggy/hazy days, whereas those for longer wavelengths (1 μm<λ< 3 μm), were 0.10 and 0.20 during clear sky or less foggy/hazy days and intense foggy/hazy days, respectively. This indicates the dominance of fine size particles. Similar results have been reported at Kanpur, where average fine mode AOD during the observation period was 0.62 and the average coarse mode AOD was 0.16 and the AOD values at 0.5 μm ranged from 0.35 to 1.6 during the month of December 2004 (Tripathi et al. 2006). Average AOD values at 0.5 μm, reported at Hisar, the location to the NW of Agra, were 0.33, 0.44 and 0.58 on clear, hazy and foggy days, respectively; during December 2004 (Ramachandran et

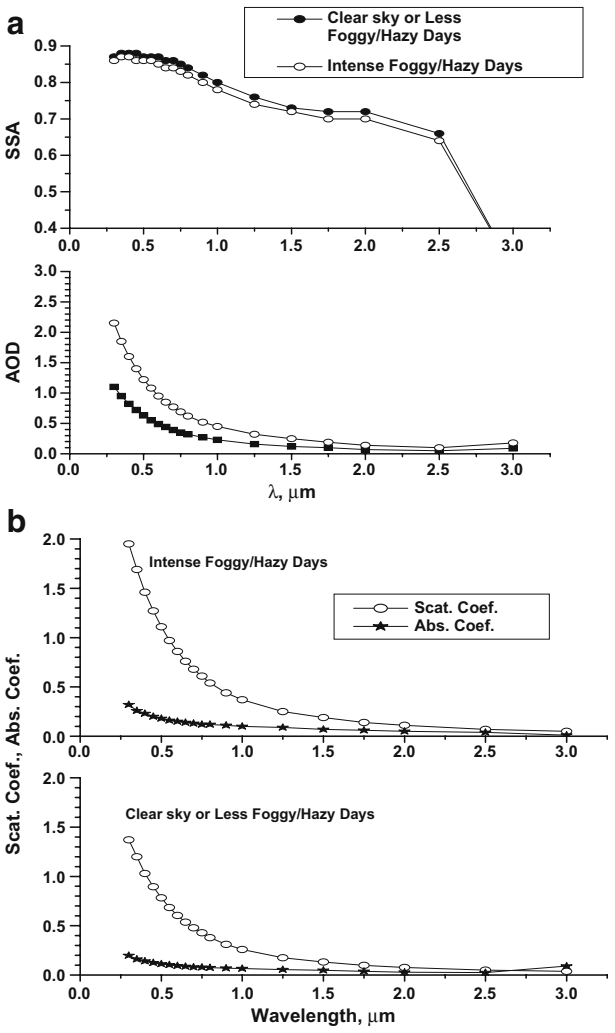


Fig. 6 **a** Spectral variation of SSA and AOD at Agra during December 2004. **b** Spectral variation of Scattering and Absorption Coefficients at Agra during December 2004

al. 2006). Doubling of AOD at 500 nm during hazy sky days (0.76), as compared to that during clear sky days (0.38) has been reported at Kharagpur, during December 2004 (Niranjan et al. 2006).

5.5.2 Spectral variation of SSA

As shown in Fig. 6a, SSA values were higher at visible wavelengths (0.84 and 0.86 during intense foggy/hazy days and clear sky or less foggy/hazy days) suggesting presence of the fine size scattering type aerosols whereas at longer wavelengths, they were lower (0.63 and 0.65 during intense foggy/hazy days and clear sky or less foggy/hazy days) suggesting the presence of coarse size absorbing type aerosols. It is in agreement with biomass burning aerosol model (Dubovik et al. 2000). SSA values were reported to be 0.88, 0.86 and 0.76 at 0.5 μm during clear, hazy and foggy conditions, respectively during December 2004 at Hisar (Ramachandran et al. 2006). At Kanpur, SSA values varied in the range of 0.87 to 0.97; showing strong spectral dependence, similar to the present observations at Agra (Tripathi et al. 2006). At Kharagpur, the OPAC derived SSA value at 500 nm was high (0.88) during hazy sky days than on clear sky days (0.85), which was attributed to the increase in large particles that scatter solar radiation more effectively (Niranjan et al. 2006). At Delhi, it was reported that SSA value during December 2004 varied between 0.6 and 0.8 with an average of 0.68 at 525 nm (Ganguly et al. 2006).

The Angstrom exponent (α), derived from OPAC model was 1.16 (350 to 500 nm) and 1.44 (500 to 800 nm) during both the intense foggy/hazy days and clear sky or less foggy/hazy days at Agra. This also indicates the major contribution by fine size aerosols, as also shown by the spectral dependence of AOD. Using OPAC results, it was observed that during both the intense foggy/hazy days and clear sky or less foggy/hazy days, the rate of decrease in scattering coefficient with wavelength was higher than that of absorbing coefficient (Fig. 6b). This feature shows the higher concentrations of fine size scattering type aerosols. However SSA values did not show much decrease during intense foggy/hazy days, even though the absorbing aerosols like BC increased during that period. This could be attributed to the simultaneous increase in scattering type aerosols like SO_4^{2-} along with absorbing type aerosols during intense foggy/hazy days, at Agra. The net increase in scattering aerosols exceeded that of absorbing aerosols (as also seen from chemical composition of TSP), leading to a meager reduction (0.02 units) in SSA value, even during the intense foggy/hazy days.

5.6 Aerosol short wave radiative forcing estimation

OPAC derived optical properties in the 0.3 to 3.0 μm wavelength ranges were used in the SBDART (Santa Barbara Discrete-ordinate Atmospheric Radiative Transfer) model to compute aerosol shortwave radiative forcing for intense foggy/hazy days and clear sky or less foggy/hazy days (Ricchiazzi et al. 1998). The other input parameters included the MODIS-derived column precipitable water, spectral surface albedo, and TOMS-derived total column ozone. Both white sky and black sky albedo at local solar noon in 7 bands (λ covering 0.305 to 2.80 μm) were used to compute actual surface albedo at Agra and the spectral average albedo was found to be 0.2. Diurnal average aerosol radiative forcing is estimated by computing the difference between net (down-up) radiative fluxes 'with aerosols' and 'aerosol free' conditions. Detailed methodology can be found elsewhere (Pandithurai et al. 2004).

Figure 7 shows the shortwave (SW) radiative forcing at surface (SUF), top of the atmosphere (TOA) and atmosphere (ATM). During the clear sky or less foggy/hazy days, the SW forcing at SUF was −40 W/m^2 whereas that at the TOA was −2 W/m^2. Therefore, the SW forcing at ATM was +38 W/m^2. During the intense foggy/hazy days, SW forcing at SUF was doubly increased and was −76 W/m^2 whereas there was a very meager forcing at the TOA (−0.2 W/m^2) and as such, forcing at ATM also almost doubly increased (+75.8 W/m^2). The enhanced surface cooling during intense foggy/hazy days at Agra is attributed to the increased concentrations of both scattering and absorbing type of aerosols. Similarly, a large negative forcing at surface (−40 to −86 W/m^2) with an average of −66 W/m^2 was reported at Delhi (Ganguly et al. 2006). Also at Kharagpur, large increase in negative forcing at SUF during hazy days (−80 W/m^2) as compared to that during clear days (−54 W/m^2) was reported and also, the forcing at ATM was 50 and 76 W/m^2 during clear and hazy days, respectively which was attributed to the combined effect of enhanced BC mass fraction (from 8% during clear days to 12% during hazy days), hygroscopic growth of aerosols and their confinement in lower altitudes due to decrease in mixing height (Niranjan et al. 2006). Significant increase in SW forcing at ATM during foggy days (49 W/m^2) as compared to the clear days (18 W/m^2) was reported (Ramachandran et al. 2006). Similarly at Agra also, SW forcing at TOA also changed from −2 W/m^2 during clear sky or less foggy/hazy days to −0.2 W/m^2 during intense foggy/hazy days, indicating more atmospheric absorption. The estimated atmospheric forcing efficiency (ratio of forcing at ATM to unit AOD) was found to be 60 and 63 Wm^{-2} during clear sky or less foggy/hazy days and intense foggy/hazy days respectively, indicating very little difference because there is not much change in SSA during these two periods. As already stated in Section 5.1, even though BC concentrations showed about 1.7 times increase during intense foggy/hazy days, their mass fraction to TSP increased by only about 1% and therefore this meager enhancement in BC mass fraction to TSP did not show much effect on SSA. A good correlation was shown between BC mass fraction and forcing efficiency over the Arabian Sea (Babu et al. 2004).

Overall, as observed from the present study at Agra and from the other reported studies on aerosol radiative forcing over the IGB region during winter season of 2004, the atmospheric SW forcing was enhanced significantly during foggy/hazy days mainly due to the increase in absorbing aerosols like BC whereas, a major increase in negative forcing was observed at the surface that can be attributed to the increase in scattering aerosols like SO_4^{2-} and NO_3^-. Thus, the impact of increase in the burning activities as well as that of prevailing meteorological conditions, especially low mixing heights and possible air-mass

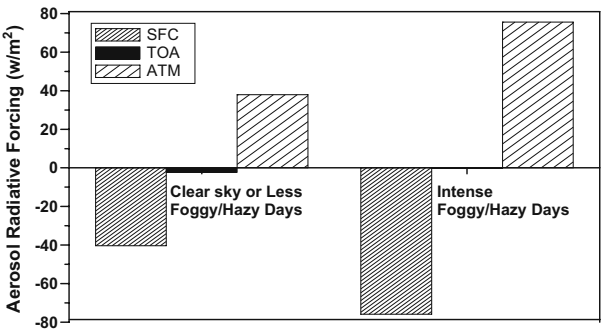


Fig. 7 Short wave aerosol radiative forcing at Agra during December 2004

transport due to western disturbances; is manifested during the winter months, which helps in formation and sustenance of fog and/or haze over this region.

6 Conclusions

During the intense foggy/hazy days, concentrations of TSP along with its all measured chemical components showed increase, especially SO_4^{2-}, NH_4^+ BC and NO_3^-. Aerosols were acidic in nature during intense foggy/hazy days. However, fog water was alkaline, mainly due to the neutralizing effect of NH_4^+. Trajectory analyses showed that air masses were predominantly from NW direction, which might be responsible for transport of BC from distant and surrounding local sources. Diurnal variation of BC on all days showed a morning and an evening peak that were related to domestic cooking and vehicular emissions, apart from boundary layer changes. AOD value at 0.5 μm wavelength was significantly high during intense foggy/hazy days (1.22) than during clear sky or less foggy/hazy days (0.63). OPAC-derived Single scattering albedo (SSA) was 0.84 during the observational period, indicating significant contribution of absorbing aerosols. The BC mass fraction to TSP increased by only 1% during intense foggy/hazy days and thereby did not show any impact on SSA during that period. SW forcing at the atmosphere (ATM) increased from +38 W/m^2 during clear sky or less foggy/hazy days to +75.8 W/m^2 during intense foggy/hazy days. The increase in forcing at ATM is mainly ascribed to the enhancement in absorbing aerosols during intense foggy/hazy days. Whereas SW forcing at surface (SUF) increased from −40 W/m^2 during clear sky or less foggy/hazy days to −76 W/m^2 during intense foggy/hazy days, mainly due to the scattering aerosols like SO_4^{2-}.

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