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Aerosol optical and radiative properties during summer and winter seasons over Lahore and Karachi

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ABSTRACT

The study of aerosol optical and radiative properties presented here focuses on a geographic region in which there exist significant gaps in our knowledge. These properties have been analyzed through the ground-based Aerosol Robotic Network (AERONET) over the two megacities of Lahore and Karachi for summer (April–June) and winter (December–February) of 2010–11. During the study period the monthly mean aerosol optical depth (AOD) at 500 nm over Lahore ranged from 0.57 ± 0.18 to 0.76 ± 0.38, and the monthly mean Ångström exponent (<alpha>) ranged from 0.39 ± 0.17 to 1.22 ± 0.13. Likewise, over Karachi the monthly mean AOD ranged from 0.33 ± 0.11 to 0.63 ± 0.28 and the <alpha> values varied between 0.29 ± 0.08 to 0.95 ± 0.22. The average AOD values in summer and winter are 0.66 ± 0.30, 0.50 ± 0.18 and 0.67 ± 0.40, 0.34 ± 0.12 in Lahore and Karachi respectively. The relationship between the Absorption Ångström Exponent (AAE) and the Extinction Ångström Exponent (EAE) provided an indication of the relative proportions of urban-industrial and mineral dust aerosols over the two sites. The volume size distributions were higher over Lahore than over Karachi during both seasons. The single scattering albedo (SSA) ranged from 0.83 ± 0.02 (440 nm) to 0.91 ± 0.05 (1020 nm) over Lahore and from 0.88 ± 0.02 (440 nm) to 0.97 ± 0.01 (1020 nm) over Karachi. The lower SSA values over Lahore suggest that absorbing aerosols are more dominant over Lahore than over Karachi. The average aerosol radiative forcing (ARF) values in summer at the surface and the top of atmosphere (TOA) are −101.6 ± 8.2, −63.3 ± 9.5 and −19 ± 4.35, −20 ± 3.1 over Lahore and Karachi respectively. Likewise, the average ARF values in winter at the surface and TOA are −90.3 ± 21.03, −57 ± 6.35 and −26 ± 7, −16 ± 2.3 over Lahore and Karachi respectively. The averaged aerosol ARF values over Lahore and Karachi for the entire period covered by the observations were −22.5 ± 5.9 W m^{-2} and −18 ± 2.2 W m^{-2} at the TOA and −96 ± 13 W m^{-2} and −60 ± 6.8 W m^{-2} at the surface, respectively, giving an averaged atmospheric forcing of 74.56 ± 16.8 W m^{-2} over Lahore and 41.85 ± 6.4 W m^{-2} over Karachi, which indicates significant heating of the atmosphere at both sites. The average heating rate during summer was 2.3 ± 0.1 and 1.2 ± 0.2 K day^{-1} and during winter was 1.8 ± 0.4 and 1.1 ± 0.1 K day^{-1} over Lahore and Karachi respectively.

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1. Introduction

Aerosol optical properties are one of the largest uncertainties in current assessments and predictions of global climate change (Intergovernmental Panel on Climate Change (IPCC), 2001). The optical properties of aerosols are responsible for many spectacular atmospheric effects, such as richly colored sunsets, halos around the sun or moon, and rainbows (Hinds, 1999). Aerosol particles also play an important role in global and regional climate change, both directly by backscattering and absorbing shortwave solar radiation, and indirectly through their fundamental role in cloud microphysics (Charlson et al., 1992; Jacovides et al., 1996). Aerosol particles therefore have a major effect on the solar radiation budget, both at the surface of the earth and within the earth's atmosphere, as well as affecting the hydrological cycle and precipitation rates (Charlson et al., 1992; Coakley et al., 1983; Lohmann and Feichter, 2005; Ramanathan et al., 2001a; Rosenfeld and Lensky, 1998; Twomey, 1977).

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There have been several field investigations into the physical and chemical properties of aerosols and their associated processes. These have included the ACE-1 (Bates et al., 1998), ACE-2 (Raes et al., 2000), ACE-Asia (Huebert et al., 2003), TARFOX (Russell et al., 1999), and INDOEX (Ramanathan et al., 2001b) programs. Other investigations have also been conducted into the optical properties of aerosols, their seasonal, spatial and temporal characteristics, and their influence on the solar radiation budget (Alam et al., 2011a, 2011c, 2010; Ge et al., 2010; Jayaraman et al., 2006; Liu et al., 2008; Prasad and Singh, 2007; Prasad et al., 2007; Ramanathan et al., 2001b; Singh et al., 2010; Smirnov et al., 2001; Tegen et al., 2004; Zheng et al., 2008). Despite these investigations aerosols remain a major source of uncertainty in the prediction of climate change due to the shortage of information on the spatio-temporal characteristics of aerosols over different locations within a particular region (Solomon et al., 2007). In recent years, however, a number of studies have been conducted that have helped to reduce the uncertainties in the direct aerosol radiative forcing (IPCC, 2007).

Satellite remote sensing is an essential tool for monitoring the global aerosol budget and the radiative effects that aerosols have on climate (Andreae, 1995; Charlson et al., 1992; Kaufman et al., 2002; Penner et al., 1992; Tripathi et al., 2005). However, satellite data is not able to provide a complete characterization of the optical properties of aerosols, or information on their other characteristics (Eck et al., 2005). A major advance in this respect has been the introduction of the AERONET (Aerosol Robotic Network) (Holben et al., 1998), which means that satellite remote sensing of aerosols no longer needs to be largely independent but can be tied in to this coordinated and harmonised ground data network. Ground-based remote sensing has become a powerful method for characterizing atmospheric aerosols (Dubovik and King, 2000) as it is able to present a clear picture of the optical properties of each of the aerosol species (Dubovik et al., 2002a; Cattrall et al., 2005).

There have been relatively few studies on aerosols in Pakistan (Alam et al., 2011a; 2011b, 2011c, 2010; Dutkiewicz et al., 2009; Ghauri et al., 2001, 1994). To the best of our knowledge no study has yet been conducted on optical properties of aerosols and aerosol radiative forcing in Lahore, and there has only been one such study to date in Karachi (Alam et al., 2011a). An improved understanding of the aerosol characteristics over Lahore and Karachi, and in particular their optical properties and radiative forcing, will enable a better understanding of both the regional and local behaviour of aerosols over the entire Pakistan region. In this study we have used AERONET data to analyze the aerosol optical properties in terms of aerosol optical depth (AOD), Ångström Exponent <alpha>, particle size distribution, single scattering albedo (SSA), and asymmetry parameter (ASY), together with the real and imaginary parts of the refractive index (RI) – $n(\lambda)$ and $k(\lambda)$, respectively. The aerosol radiative forcing (ARF) computations have been carried out using the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model (Ricchiazzi et al., 1998).

2. AERONET sampling sites and instrumentation

2.1. Site description

Lahore(31° 32′ N; 74° 22′ E) is the second largest city in Pakistan, with a population of approximately 10 million. It is situated along the Ravi River, close to the border with India (Fig. 1b). The climate in Lahore is hot and semi-arid, with relatively wet and extremely hot summers and dry, warm winters. The mean maximum temperatures in summer (April–June) range between 33 and 39 °C and in winter from 17 to 22 °C. Similarly the mean minimum temperatures in summer range between 22 and 28 °C and in winter from 7 to 12 °C.

The major industries in Lahore include the manufacture of motor cars and motorcycles, steel, chemicals, pharmaceuticals, engineering products, and construction materials.

The aerosols over this sampling site derive mainly from soil or road dust, industrial emissions, and vehicular emissions, or are secondary aerosols. Other anthropogenic sources include emissions from main highways, coal combustion and biomass burning (Biswas et al., 2008). The ground-based CIMEL sky radiometer (Lahore AERONET station) in Lahore has been operational since December 2006 under a collaboration between NASA and the Institute of Space Technology's Lahore office.

Karachi is the largest city in the country and is located in the south of Pakistan (24° 51′ N; 67° 02′ E). It has a population of more than 16 million and covers an area of around 3500 km^2. The climate in Karachi is relatively mild and subtropical to arid, with a low mean annual rainfall of 250 mm. The mean maximum temperature in summer (April–June) ranges from 33 to 36 °C and in winter (December–February) from 26 to 29 °C. Similarly the mean minimum temperature in summer ranges from 24 to 29 °C and in winter from 12 to 22 °C. The major industries include textiles, pharmaceuticals, cement factories, oil refineries, automobiles, chemicals, heavy machinery, shipbuilding, and a steel mill. In addition to industrial emission, vehicular emission from highways and emissions of sea salt particle from Arabian Sea are the major local sources of aerosol in this mega city. The AERONET sampling site is to the north–west of Karachi Airport, adjacent to the University of Karachi campus. The ground-based CIMEL sky radiometer at the Karachi AERONET station has been operational since August 2006, under the same joint collaboration between NASA and the Institute of Space Technology's Karachi office.

2.2. Instrumentation

The CIMEL sky radiometer is the standard AERONET instrument for taking measurements of the direct sun and diffuse sky radiances, within the 340–1020 nm and 440–1020 nm spectral ranges, respectively (Holben et al., 1998). The AERONET inversion algorithm (Dubovik et al., 2000) provides improved aerosol retrievals by fitting the entire measured field of radiances (sun radiance and angular distribution of sky radiances at four wavelengths: 440, 670, 870, and 1020 nm) to the radiative transfer model (Dubovik et al., 2002). The inversion algorithm is used to retrieve aerosol volume size distributions in the size range from 0.05 to 15 μm, together with spectrally dependent complex RI, SSA and ASY parameters from spectral sun and sky radiance data. The detailed aerosol properties retrieved are used for calculating broad brand flux within the spectral range from 0.2 to 4.0 μm. The AERONET data are available at three levels: level 1.0 (unscreened), level 1.5 (cloud screened – Smirnov et al., 2000), and level 2.0 (quality assured – Holben et al., 1998) and can be downloaded from the AERONET website (http://aeronet.gsfc.nasa.gov/). For this study we used AERONET level 1.5 (cloud screened) data, from both direct sun (AOD and <alpha>) and inversion products (SSA, ASY, RI, AAE, EAE), during 2010–2011 for the summer and winter seasons. AERONET level 2.0 data is not available over this period. The AERONET data for Lahore during 2007–2009 is not sufficient for the analysis of aerosol optical and radiative properties, therefore long term data can not be analyzed for this period.

The uncertainty in the retrieval of AOD under cloud free conditions is $<\pm0.01$ for wavelengths (λ) >440 nm and $<\pm0.02$ for shorter wavelengths, which is less than the ±5% uncertainty for the retrieval of sky radiance measurements (Dubovik et al., 2000). The errors for particle retrieval in the size range ($0.1 \leq r \leq 7$ μm) do not exceed 10% in the maxima of the size distributions but may increase to about 35% for the points corresponding to the minimum values of $dV(r)/d\ln r$ within this size range (Dubovik et al., 2002). However, for

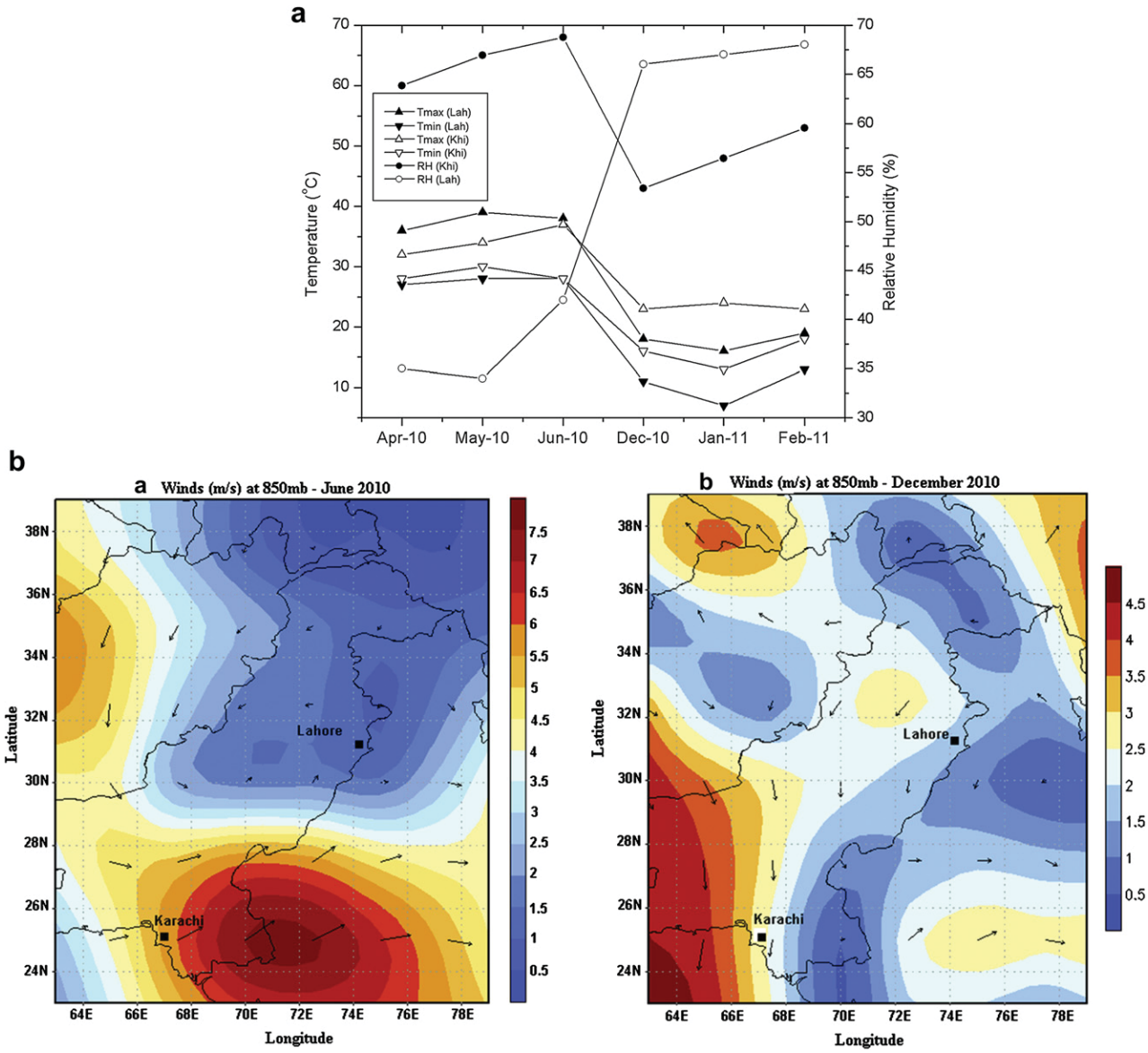

Fig. 1. a. Prevailing meteorological conditions for Lahore and Karachi during the study period. b Mean synoptic wind vector at 850 mb pressure level over the Pakistan region and its surroundings during (a) June 2010, (b) December 2010.

particles smaller than 0.1 μm or larger than 7 μm the accuracy of the size distribution retrieval drops significantly because of the low sensitivity of the aerosol scattering at 0.44, 0.67, 0.87 and 1.02 μm to particles in these size ranges. Aerosol particle size distributions ($dV(r)/d\ln r$) are known to typically have low values at the edges of their retrieval size intervals and the relatively high errors do not significantly affect the derivation of the main features of the aerosol particle size distribution (Dubovik et al., 2000, 2002). Single scattering albedos are expected to have an uncertainty of 0.03–0.05, depending on aerosol type and loading (Dubovik et al., 2000). The errors in RI are estimated at 30–50% for the imaginary part and ±0.04 for the real part (Dubovik et al., 2002). These estimated errors are for high aerosol loadings (AOD(440) ≥ 0.5) at a solar zenith angle greater than 50°.

2.3. Synoptic meteorology

The average prevailing meteorological conditions over the study period are shown in Fig. 1a. The monthly mean maximum and minimum temperatures and the relative humidity were obtained from Pakistan Meteorological Department. The mean maximum and minimum temperatures were in the range of 16–39 °C and 7–28 °C in Lahore, and 23–37 °C and 13–30 °C in Karachi, respectively. The mean ambient relative humidity ranged from 34 to 68% in Lahore and from 43 to 68% in Karachi. The surface wind flow patterns at 850mb were obtained from the U.S.A.'s National Center for Environmental Prediction (NCEP) and the National Center for Atmospheric Research (NCAR) reanalysis of monthly data (http://www.cdc.noaa.gov) over the Lahore and Karachi regions (along with other parts of Pakistan). Fig. 1b shows the wind flow pattern for the typical summer and winter months of June 2010 and December 2010, with the arrows showing wind directions and the lengths of the arrows representing the wind speeds (m s^{-1}). In June 2010 the region experienced strong winds that were predominantly directed in a north-westerly direction from the Arabian Sea towards Karachi and Lahore. In December the winds direction were predominantly from north towards south at Karachi and towards south-west at Lahore.

3. Result and discussion

3.1. Aerosol optical depth and Ångström Exponent

The monthly variations in AOD at a wavelength of 500 nm and <alpha> (440–870 nm) for summer (April, May, June) and winter (December, January, February) over Lahore and Karachi during 2010–2011 are shown in Fig. 2a. The error bars show the standard deviations for the averaged monthly values. <Alpha> is determined from the spectral dependence of the measured optical depth as suggested by Ångström (1961). <Alpha> is a good indicator of aerosol particle size. High values of <alpha> indicate a dominance of fine particles, while low values indicate a relatively lower concentration of fine particles (or dominance of coarse particles). AOD values were higher over Lahore than over Karachi in both summer and winter seasons. The average AOD values in summer and winter at Lahore are 0.66 ± 0.30 and 0.67 ± 0.40 and at Karachi are 0.50 ± 0.18 and 0.34 ± 0.12. <Alpha> values were in the range of 0.29–1.22, with the lowest value in June and the highest in January. There was an inverse relationship between AOD and <alpha>. The decrease in <alpha> (particularly over Karachi) between April and June was associated with dust loading. The highest mean AOD over Lahore was observed for the month of December (0.76 ± 0.35), when the corresponding <alpha> value was 1.20 ± 0.08. The high <alpha> (1.20 ± 0.08) corresponds to higher concentration of fine particles, which could be attributed to vehicular and industrial emissions (presumably containing diesel emission). Over Karachi the highest mean AOD (0.63 ± 0.28) was observed during the month of June, with a corresponding <alpha> value of 0.29 ± 0.08. This means that aerosol particles were larger during this period, which relates to dust aerosols (Alam et al., 2011c). Behnert et al. (2007) found that sea salt and Saharan dust were associated with low <alpha> values, represented by a high proportion of large particles. The lowest AOD (0.32 ± 0.11) over Karachi, with a corresponding <alpha> value of (0.95 ± 0.21), was observed in January (see Table 1), which is attributed to anthropogenic aerosols.

In order to investigate the origins of the air masses arriving in the studied sites due to long-range transport we performed back trajectory analyses based on the NOAA HYSPLIT (National Oceanic and Atmospheric Administration Hybrid Single Particle Langrangian Integrated Trajectory) model (Draxler and Rolph, 2003). These trajectories were computed at several altitudes (2500 m, 1500 m, and 1000 m) for 30th April 2010, 30th June 2010, 24th December 2010 and 28th February 2011. These trajectories are considered to be representative of the entire time period analyzed. Fig. 2b shows that different type of air masses reaching from various source regions at the study sites. The air masses reached Lahore and Karachi from the Dasht (Iran), Cholistan (Pakistan), Thar (India) deserts, Hamoun Wetlands (Afghanistan), and also from the Arabian Sea.

Fig. 2c shows the monthly mean values for the water vapor column (WVC), which ranged between 0.7 cm and 3.3 cm. The WVC started to increase from the month of April and reached its maximum value in June before decreasing to its lowest value in December. The WVC values were found to be high in summer and lower in winter, which is consistent with the general synoptic pattern for the region. Overall, the WVC values were higher over Karachi than over Lahore, as shown in Fig. 2c. The correlation coefficients between WVC and AOD were found to be 0.23 for Lahore and 0.94 for Karachi, which is clearly evident from Fig.2a and c and suggests that aerosol particles over Karachi are more hygroscopic than over Lahore (Alam et al., 2010).

Fig. 2d shows a scatter plot of Absorption Ångström Exponents (AAEs) vs. Extinction Ångström Exponents (EAEs) for wavelengths between 440 and 870 nm for Lahore and Karachi. This type of analysis is necessary in order to distinguish between aerosol types (Russel et al., 2010a). The plot represents the urban-industrial and mineral dust aerosol compositions over the two sites. Dust is frequently blown from southern Pakistan, across the Arabian Sea towards Karachi, resulting in a higher AOD in the Karachi region (Alam et al., 2010, 2011a, 2011c). The AAE values ranged from 0.53 to 2.35. Theoretical AAE values are close to 1 for black carbon, higher for biomass burning aerosols (through partially overlapping), and highest for Saharan dust aerosols (Russel et al., 2010b). Schnaiter et al. (2003) found that diesel particle emissions had an AAE of about 1. Clarke et al. (2007) reported that AAE values around 1 were due to urban pollution, and around 2.1 may have originated from burning biomass. Yang et al. (2009) investigated the relationship between the AAE and the Scattering Ångström Exponent (SAE) near Beijing and found that dust generally had an SAE close to zero due to its large particle size, while the AAE was high but variable. Clarke et al. (2007) analyzed the relationship between SAE and AAE in an attempt to distinguish dust from pollution plumes and biomass burning. However, the relationship between AAE and EAE measurements was unable to distinguish urban-industrial aerosols from those derived from biomass burning. More research is therefore required into improving the remote identification of aerosol types (Russel et al., 2010b).

3.2. Aerosol volume size distribution

The AERONET aerosol volume size distributions ($dV(r)/d\ln r$) are retrieved from spectral sun and sky radiance data using the Dubovik and King (2000) approach, with the following initial guess: $dV(r)/d\ln r = 0001$, $n(\lambda_i) = 1.50$, $k(\lambda_i) = 0.005$, where $dV/d\ln r$ denotes the aerosol volume size distribution, and $n(\lambda_i)$ is the real and $k(\lambda_i)$ the imaginary part of the refractive index (RI) at wavelength λ_i. The volume size distributions exhibit a bimodal structure, which can be characterized by the sum of two log-normal distributions as follows:

$$\frac{dV(r)}{d\ln r} = \sum_{i=1}^{2} \frac{C_{V,i}}{\sqrt{2\pi}\sigma_i} \exp\left[-\frac{(\ln r - \ln r_{V,i})}{2\sigma_i^2}\right]$$

where $r_{V,i}$ is the volume median radius, $C_{V,i}$ is the volume concentration and σ is the geometric standard deviation for mode i.

The aerosol volume size distributions over Lahore and Karachi during the 2010–2011 summer and winter seasons are shown in Fig. 3. For both sites the volume size distributions in the coarse mode are higher in summer and lower in winter. The higher values in summer are due to dust and also due to meteorological conditions, such as temperature, pressure, and relative humidity (Alam et al., 2011a). The volume size distributions in the accumulation mode are higher in winter than in summer for both sites, which is attributed to hygroscopic growth of ambient particles (Alam et al., 2010; Singh et al., 2004; Tripathi et al., 2005). The volume size distributions are consistently higher over Lahore than over Karachi. During the summer, aerosol volume size distributions over both Lahore and Karachi are 3 times higher than in the winter. Variations in aerosol volume size distributions over Lahore and Karachi occur in the coarse mode, whereas only minor variations are observed in the accumulation mode. Similar aerosol volume size distributions in the coarse mode have been found in Bahrain (Smirnov et al., 2001) and Karachi (Alam et al., 2011a).

3.3. Single scattering albedo and asymmetry parameter

Single scattering albedo (SSA) and asymmetry parameter (ASY) are key quantities for the determination of the aerosol radiative forcing effects. Together with the aerosol optical depth (AOD) these

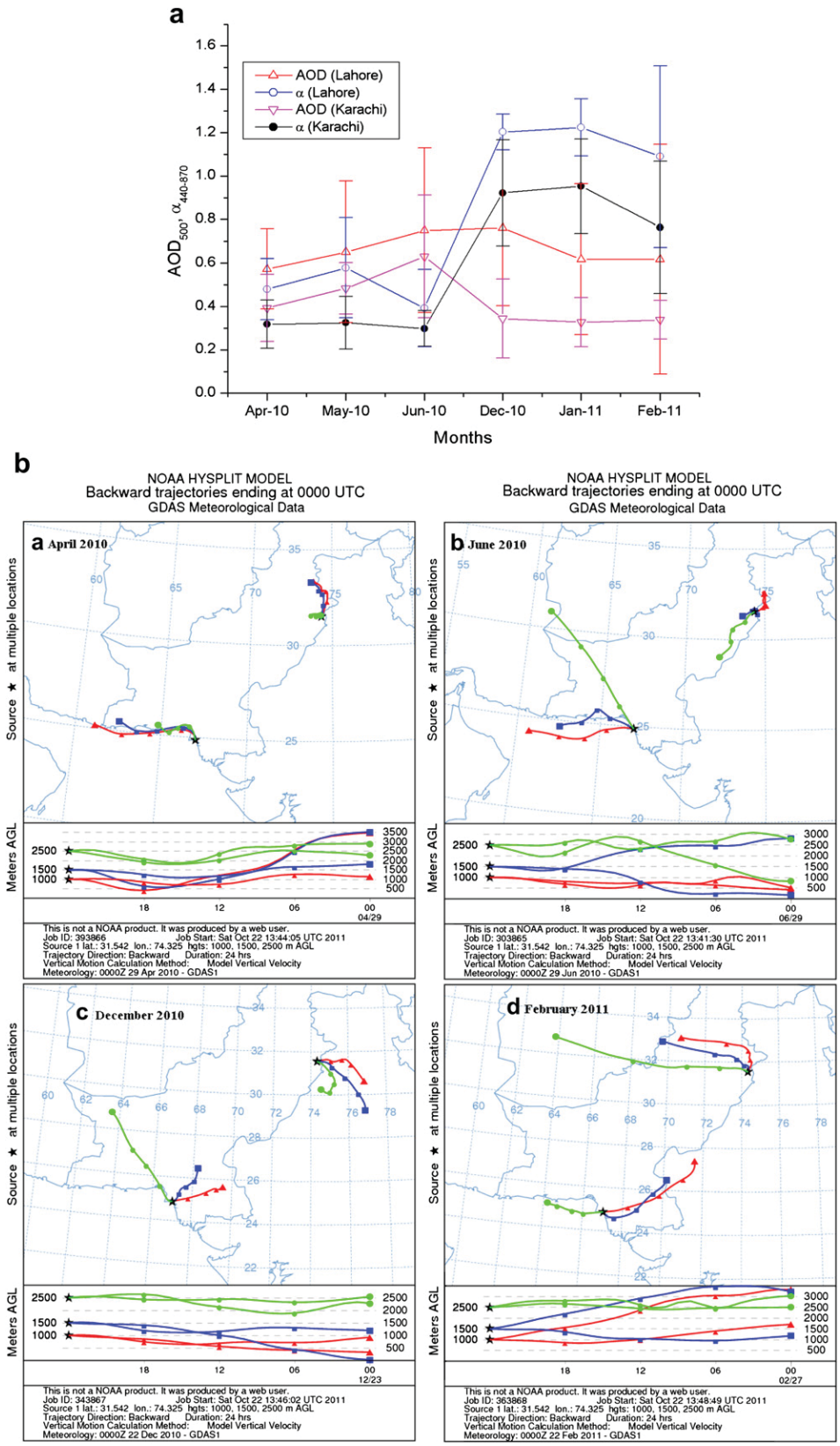

Fig. 2. (a). Monthly average variations in AOD at 500 nm and <alpha> (440–870) (b) HYSPLIT model back trajectories at 1000, 1500 and 2500 m on different days during 2010–11 (c) Water vapor column and (d) Scatter plot of AAE vs. EAE for Lahore and Karachi during summer and winter.

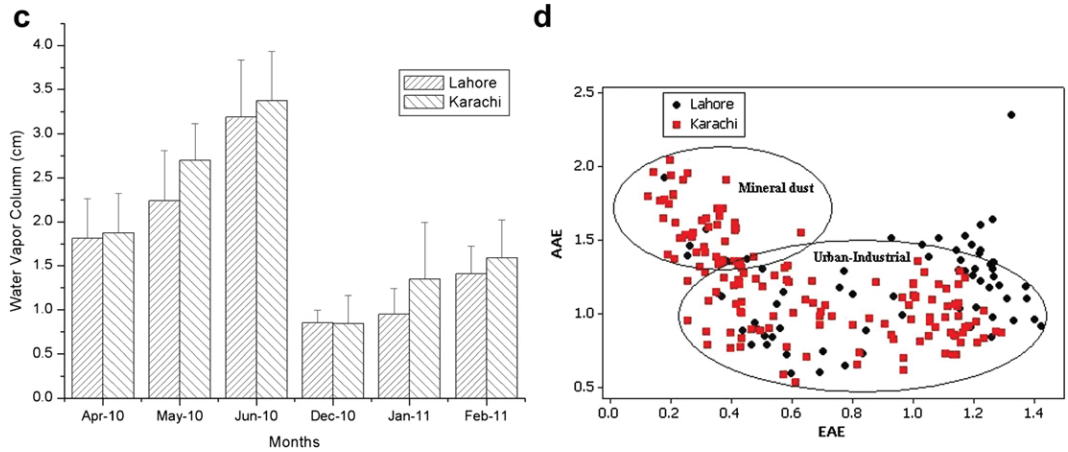

Fig. 2. (*continued*).

quantities determine the aerosol radiative forcing effects as explained in section 3.5 later on. The present paper focuses on solar heating effects in the heavily polluted urban environment of Karachi and Lahore. Therefore, column-averaged data from AERONET are used to characterize the variabilities of these radiatively relevant parameters during summer and winter. From the uncertainty discussion in section 2.2 it can be concluded that the AERONET derived time series for SSA, ASY and AOD currently provide the most reliable data source to perform such an assessment for the two megacities in Pakistan.

The single scattering albedo (SSA) is the ratio of scattering efficiency to total extinction efficiency and provides important information regarding the scattering and absorption properties of aerosols. The averaged SSA during summer and winter at 440 nm over Lahore and Karachi are 0.85 ± 0.02, 0.92 ± 0.02 and 0.88 ± 0.03, 0.89 ± 0.02 respectively. Monthly averaged SSA values and corresponding standard deviations at 440, 675, 870 and 1020 nm are shown in Fig. 4a–b. SSA was found to be wavelength dependent due to the influence of dust and anthropogenic activities during both the summer and winter seasons. Spectral variations in the SSA differ between dust and urban pollution, with the SSA tending to increase rapidly with increasing wavelength during dust events but to decrease during periods of increased urban pollution (Bergstrom et al., 2007; Dubovik et al., 2002). The SSA over Lahore between April and June increased with increasing wavelength (Fig. 4a) reflecting the dominance of dust particles over anthropogenic aerosols in the atmosphere during this period. Our results are consistent with the findings of other authors (Cheng et al., 2006b; Dubovik et al., 2002; Xia et al., 2005), namely that SSA slightly increases with increasing wavelength. The low SSA values in April and May (0.83 ± 0.02 and 0.84 ± 0.02 at 440 nm) are attributed to the admixture of dust aerosols with absorbing anthropogenic aerosols.

In winter (December–February) the absorbing aerosols over Lahore were dominant, and the SSA consequently decreased with increasing wavelength. Singh et al. (2010) also found that the SSA decreased over Delhi with increasing wavelength during the winter, when local pollution is dominant, and a similar decrease in SSA with wavelength was reported by Zheng et al. (2008) in China. The SSA over Lahore varied between 0.83 ± 0.02 in May and 0.89 ± 0.03 in January (at 440 nm), and between 0.91 ± 0.06 in June and 0.86 ± 0.08 in February (at 1020 nm); these values are close to those from the Indian Ocean Experiment (INDOEX) which ranged from 0.86 to 0.9 at 550 nm (Ramanathan et al., 2001b). Over Karachi the SSA increased significantly with increasing wavelength for April, varying between 0.93 ± 0.02 at 440 nm and 0.97 ± 0.01 at 1020 nm (Fig. 4b). The SSA over Karachi varied between 0.88 ± 0.03 in December (at 870 nm) and 0.97 ± 0.01 in April (at 1020 nm). This is slightly lower than the SSA values reported by other authors for desert dust (0.95–0.99 nm: Dubovik et al., 2002; Kaufman et al., 2001), which suggests a possible combination of dust, urban-industrial particles, and sea salt aerosols over the Karachi region (Alam et al., 2011b). Alam et al. (2011a) found that the SSA

Table 1
Mean AOD at a wavelength of 500 nm and Alpha (440–870 nm) with corresponding standard deviation during 2010–11.

Months	Mean AOD (500 nm) ±SD		Mean Alpha (440–870 nm) ±SD	
	Lahore	Karachi	Lahore	Karachi
Apr 2010	0.57 ± 0.18	0.39 ± 0.15	0.48 ± 0.14	0.32 ± 0.11
May 2010	0.65 ± 0.32	0.48 ± 0.11	0.57 ± 0.23	0.32 ± 0.12
Jun 2010	0.75 ± 0.38	0.63 ± 0.28	0.39 ± 0.17	0.29 ± 0.08
Dec 2010	0.76 ± 0.38	0.34 ± 0.18	1.20 ± 0.08	0.92 ± 0.24
Jan 2011	0.62 ± 0.35	0.33 ± 0.11	1.22 ± 0.13	0.95 ± 0.22
Feb 2011	0.62 ± 0.52	0.34 ± 0.09	1.09 ± 0.41	0.76 ± 0.30

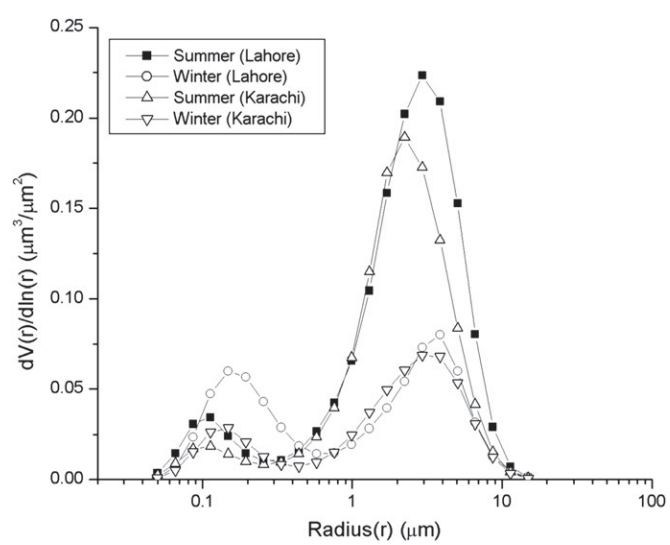

Fig. 3. Seasonal variations in AERONET retrieved aerosol size distributions during 2010–11.

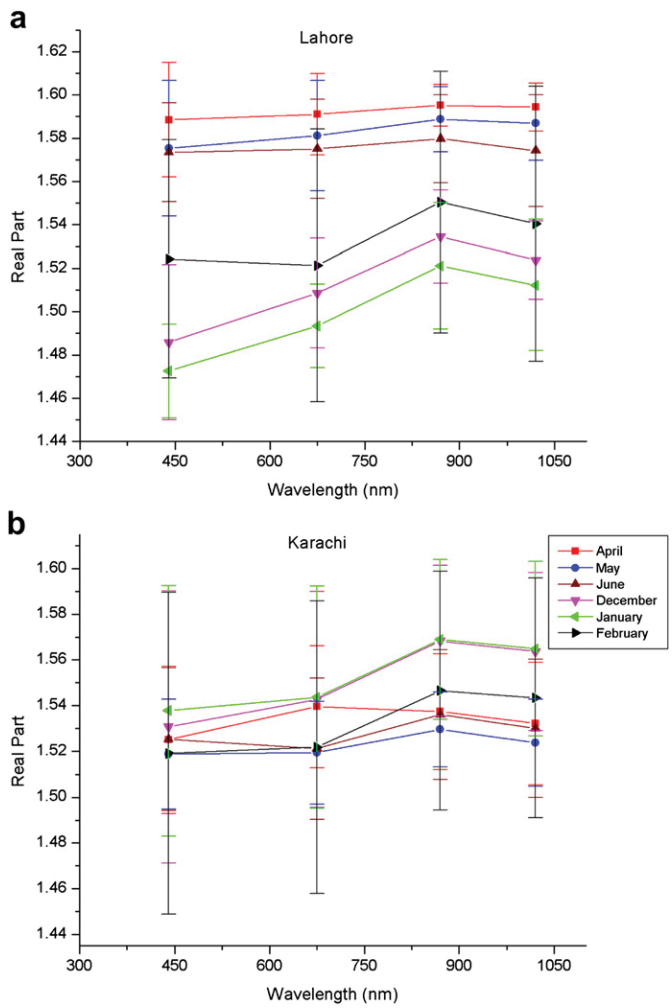

Fig. 4. Monthly average spectral variation of single scattering albedo (SSA) over (a) Lahore and (b) Karachi for summer and winter seasons during 2010–11.

increased significantly with increasing wavelength during the summer months, when dust aerosols are dominant over local pollution and the atmosphere contains more water-soluble particles, in conditions similar to those found by Singh et al. (2004) in Kanpur, India. In addition, for locations closer to the ocean the air is more humid during summer leading to enhanced water uptake of the water-soluble particle fraction. Our SSA values are comparable to those obtained between April and June by Prasad and Singh (2007) and Prasad et al. (2007) over the Indo-Gangetic Plains, India. Over Karachi the SSA during the winter was almost steady, reflecting a mixing of aerosols from different sources. Our results for winter over Karachi are similar to those obtained in a study conducted by Alam et al. (2011a) in Karachi during the winter of 2007. The overall differences in SSA variations with wavelength between Lahore and Karachi are due to differences in meteorological conditions, geographical locations, and aerosol types. Bergstrom et al. (2007) and Russel et al. (2010a, 2010b) analyzed the SSA variations in different regions of the world and concluded that SSA increases with increasing wavelength for locations dominated by desert dust, which in our study was the case over Karachi between April and June and over Lahore in June. They also found that SSA spectra decrease with increasing wavelength for urban-industrial and biomass burning aerosols, as was the case over Lahore during the winter of 2010–11.

The spectral-dependent ASY values over Lahore and Karachi are given in Table 2. They vary between 0.61 ± 0.01 at 1020 nm in December and 0.72 ± 0.02 at 440 nm in June over Lahore, and between 0.64 ± 0.04 at 870 nm in December and 0.73 ± 0.01 at 440 nm in June over Karachi. Our ASY values over Lahore are comparable with those obtained by Srivastava et al. (2011) over Kanpur and Gandhi College in India between April and June, but their values are lower than ours from Karachi. In general, the ASY decreases with increasing wavelength over both Lahore and Karachi. The greatest decrease occurs during the winter at both sites, suggesting that absorbing anthropogenic aerosols are dominant during the winter season.

3.4. Refractive index

The real $n(\lambda)$ and imaginary $k(\lambda)$ parts of the refractive index (RI) provide an indication of highly scattering or highly absorbing types of aerosols, with higher $n(\lambda)$ values corresponding to the scattering types and higher $k(\lambda)$ values corresponding to the absorbing types (Bohren and Huffman, 1983; Sinyuk et al., 2003). The monthly averaged $n(\lambda)$ values at 440, 675, 870 and 1020 nm over Lahore and Karachi are shown in Fig. 5a–b. The $n(\lambda)$ values are greater at higher wavelengths than at shorter wavelengths due to higher absorption by coarse particles in the near infrared band (Cheng et al., 2006a, 2006b). In our investigations the $n(\lambda)$ values ranged between 1.47 ± 0.02 (in January) and 1.59 ± 0.01 (in April) over Lahore, and between 1.52 ± 0.02 (in May) and 1.56 ± 0.02 (in January) over Karachi. The $n(\lambda)$ values over Lahore were higher between April and June and lower between December and February. The higher values during the summer months can be attributed to the coarse (dust) particles and the lower values during the winter months may be attributable to anthropogenic particles in the atmosphere, since the $n(\lambda)$ values of dust aerosols are greater than those of anthropogenic aerosols (Liu et al., 2008). In contrast, the $n(\lambda)$ values over Karachi were highest in the winter months and lowest in the summer months. The high values in winter were due to a mixture of aerosols in the region. Our results for the summer months at both sites are consistent with those obtained by Prasad and Singh, (2007), Prasad et al., (2007) and Tripathi et al. (2005) over Kanpur, in India. Our results for $n(\lambda)$ are also comparable with results discussed by Dubovik et al., (2002a) for Bahrain in the Persian Gulf and the Solar Village in Saudi Arabia.

Table 2
Monthly averaged Asymmetry Parameter with corresponding standard deviations for Lahore and Karachi during summer and winter 2010–2011.

Month	Asymmetry Parameter (Lahore)				Asymmetry Parameter (Karachi)			
	440 nm	675 nm	870 nm	1020 nm	440 nm	675 nm	870 nm	1020 nm
Apr 2010	0.71 ± 0.01	0.69 ± 0.01	0.69 ± 0.02	0.70 ± 0.01	0.73 ± 0.02	0.71 ± 0.01	0.70 ± 0.01	0.70 ± 0.01
May 2010	0.71 ± 0.02	0.69 ± 0.02	0.69 ± 0.02	0.69 ± 0.02	0.73 ± 0.01	0.71 ± 0.01	0.70 ± 0.01	0.71 ± 0.01
Jun 2010	0.72 ± 0.02	0.70 ± 0.02	0.70 ± 0.01	0.70 ± 0.01	0.73 ± 0.01	0.71 ± 0.01	0.70 ± 0.01	0.71 ± 0.01
Dec 2010	0.71 ± 0.01	0.64 ± 0.01	0.62 ± 0.01	0.61 ± 0.01	0.70 ± 0.02	0.66 ± 0.02	0.64 ± 0.04	0.65 ± 0.03
Jan 2011	0.71 ± 0.01	0.65 ± 0.02	0.62 ± 0.03	0.62 ± 0.03	0.70 ± 0.02	0.66 ± 0.02	0.65 ± 0.03	0.65 ± 0.03
Feb 2011	0.70 ± 0.03	0.65 ± 0.03	0.62 ± 0.03	0.62 ± 0.03	0.71 ± 0.02	0.68 ± 0.02	0.67 ± 0.03	0.67 ± 0.03

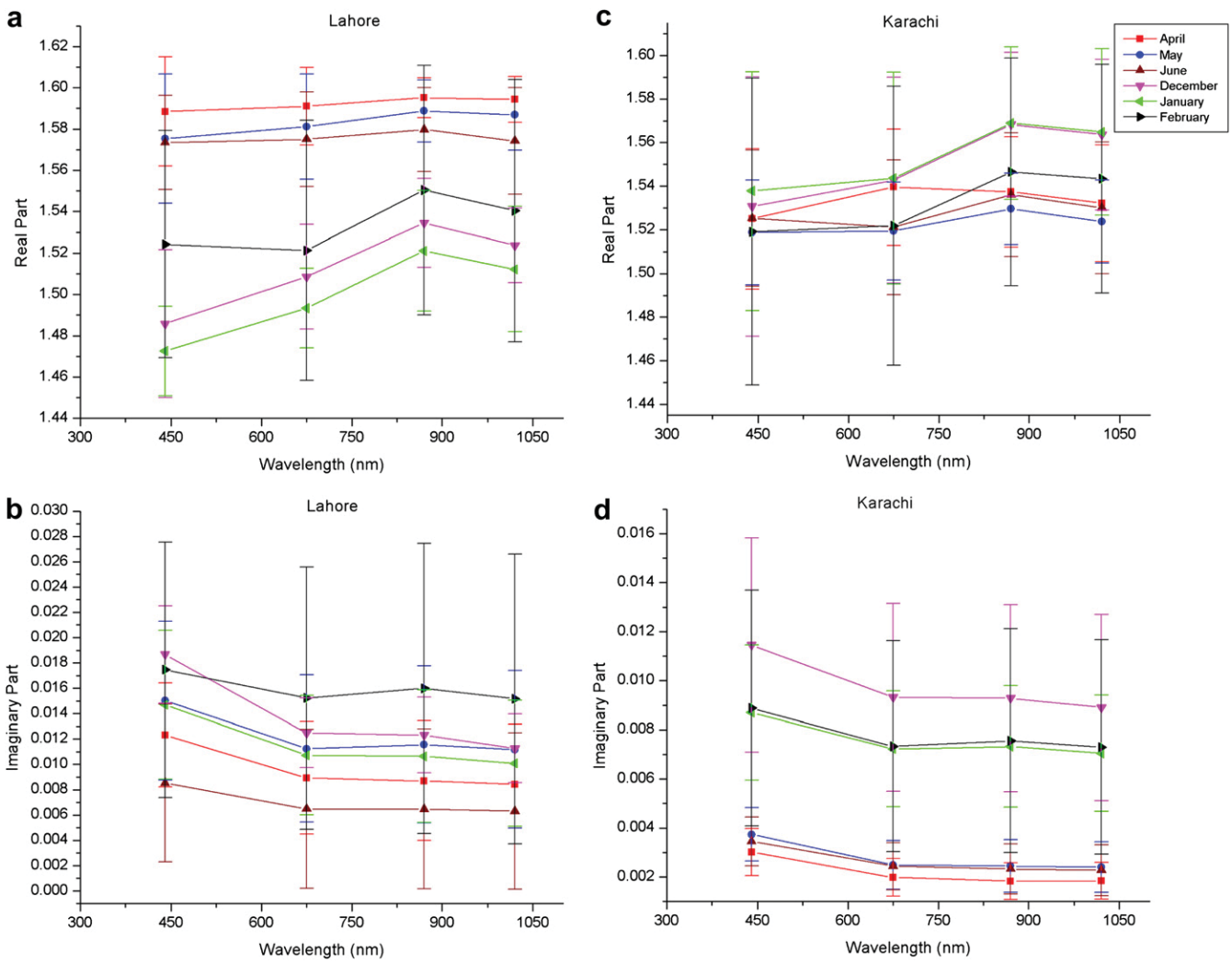

Fig. 5. Spectral variations of the real parts at (a) Lahore and (b) Karachi, and imaginary parts at (c) Lahore and (d) Karachi of the refractive index during the study period.

The spectral variations in the $k(\lambda)$ parts of the RI over Lahore and Karachi are shown in Fig. 5c-d. The $k(\lambda)$ value is also wavelength dependent and generally decreases as the wavelength increases. The $k(\lambda)$ values ranged between 0.006 at 1020 nm (in June) to 0.019 at 440 nm (in December) over Lahore, and between 0.001 at 1020 nm (in April) to 0.011 at 440 nm (in December) over Karachi. The $k(\lambda)$ values were higher over Lahore than over Karachi in both summer and winter. The averaged values for $n(\lambda)$ and $k(\lambda)$ (1.47 ± 0.02 and 0.014 ± 0.005) over Lahore during the month of January are rather similar to those obtained by Dubovik et al. (2002a) over Mexico City. The highest $k(\lambda)$ value (0.019) was recorded over Lahore during the month of December. The higher $k(\lambda)$ values at the two shortest wavelengths (440 nm and 670 nm) are attributed to the absorption of organic carbon/black carbon (Arola et al., 2011). The $k(\lambda)$ values were generally higher in winter and lower in summer, with the higher values relating to absorbing anthropogenic aerosols and the lower values to dust aerosols (Alam et al., 2011a). Our results are consistent with those from other investigations conducted at Kanpur, in India (Singh et al., 2004; Tripathi et al., 2005). The higher values for the imaginary part of the refractive index for both cities are attributed to the influence of enhanced levels of anthropogenic absorbing particles released during wintertime (cf. residential heating using coal and wood during winter, see also Dutkiewicz et al., 2009).

3.5. Aerosol radiative forcing

The aerosol radiative forcing at the top of the atmosphere (TOA) or at the surface is defined as the difference in the net (down minus up) solar flux (solar plus long wave; in W m^{-2}) with and without aerosol, i.e.,

$$\Delta F = (F_{a\downarrow} - F_{a\uparrow}) - (F_{0\downarrow} - F_{0\uparrow})$$

where ΔF denotes the irradiance (downwelling or upwelling, W m^{-2}) and $(F\downarrow - F\uparrow)$ denotes the net irradiance (downwelling minus upwelling) computed with aerosol (F_a) and without aerosol (F_0) at either the TOA or the surface.

In our study we have computed the net flux at the TOA and at the surface separately, within the wavelength range from 0.3 to 4.0 μm, both with and without aerosols, using the SBDART model (Ricchiazzi et al., 1998). This model was developed by the atmospheric science community and is widely used for radiative transfer calculations. It is run at 1 h intervals over a 24 h period and the integrated average ARF is estimated during clear-sky days in summer (April–June) and winter (December–February).

Based on the prevailing weather conditions and measured parameters, we used the mid-latitude summer and winter atmospheric model. In order to have a better representation of relevant

atmospheric parameters and improve the accuracy of the estimated ARF, we used daily mean values for columnar water vapor and total column ozone concentrations, obtained from the sun/sky radiometer and the Ozone Monitoring Instrument (OMI) on board NASA's Aura satellite. The optical parameters crucial for ARF estimations are AOD, SSA, ASY and surface albedo. Other input parameters in the model include solar zenith angle, which is calculated using a small code in the SBDART model by specifying a particular date, time, latitude and longitude. The ozone concentration and surface albedo values were obtained from the Aura OMI version 3 reflectivity data through the Giovanni online data system, developed and maintained by the NASA GES DISC (http://disc.sci.gsfc.nasa.gov/giovanni). The AOD, SSA, and ASY used for the ARF calculations were taken from the Lahore and Karachi AERONET sites.

The monthly average ARF variations at the TOA, surface, and within the atmosphere during the study period are shown in Fig. 6a–b. The ARF at surface was found to be between −70 and −112 W m^{-2} for Lahore, and between −52 and −73 W m^{-2} for Karachi, while at the TOA the ARF was found to be between −16 and −31 W m^{-2} over Lahore and between −14 and −21 W m^{-2} over Karachi. Likewise the ARF within the atmosphere was between 51 and 82 W m^{-2} over Lahore, and between 33 and 51 W m^{-2} over Karachi. The averaged ARF at the surface in summer and winter periods were −101 ± 8.2 W m^{-2} and −90.3 ± 21.03 W m^{-2} for Lahore and −63 ± 9.5 W m^{-2} and −57 ± 6.35 W m^{-2} for Karachi, while at the TOA these were −19 ± 4.3 W m^{-2} and −26 ± 7 W m^{-2} over Lahore and −20 ± 3.1 W m^{-2} and −16 ± 2.3 over Karachi, giving rise to averaged atmospheric forcing of 82.6 ± 5.5 W m^{-2} and 64.3 ± 11.2 over Lahore and 43.33 ± 8.4 W m^{-2} and 40.6 ± 2.3 W m^{-2} over Karachi and indicating significant heating of the atmosphere at both sites. The forcings calculated in this study are consistent with those calculated by Alam et al. (2011a) over Karachi.

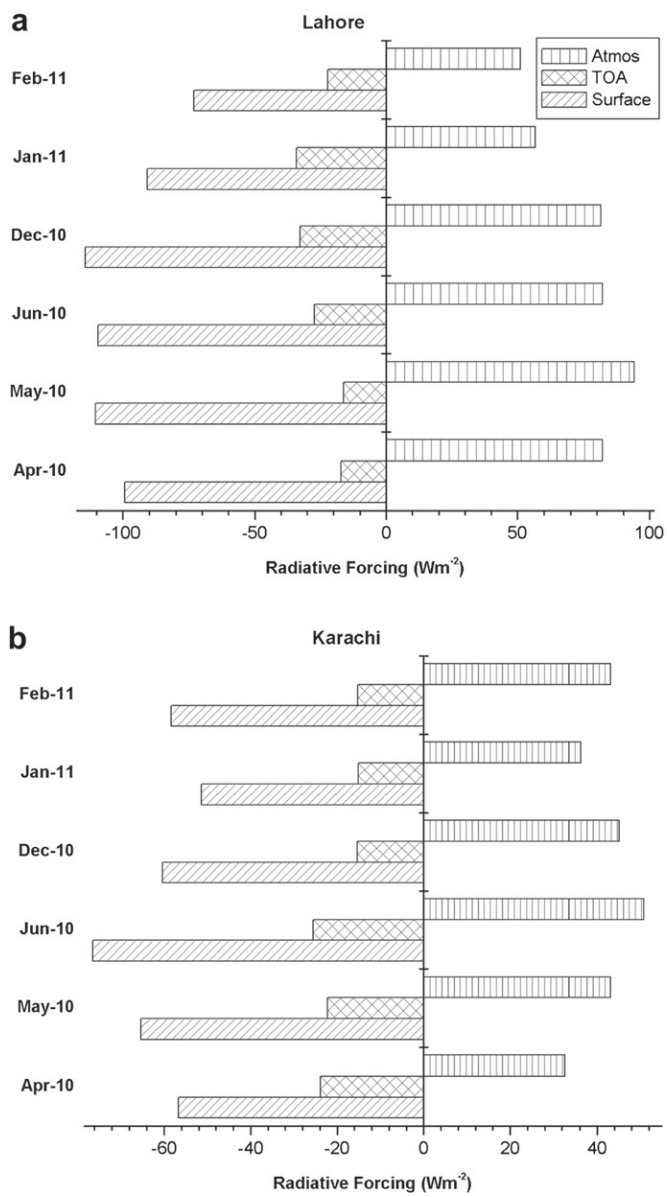

Fig. 6. Monthly averaged variations of simulated aerosol radiative forcing (W m^{-2}) over TOA, surface and in the atmosphere at (a) Lahore (b) Karachi during the study period.

The global mean clear-sky ARF at the surface and at the TOA have been found by previous authors to be negative (Ge et al., 2010; Kim and Ramanathan, 2008; Yu et al., 2006). Ge et al. (2010) found that ARF at surface to be between −7.9 and −35.8 W m^{-2} over north-western China, which are lower than our values. Our calculated TOA ARF over Karachi and Lahore are comparable with the TOA ARFs calculated by Huizheng et al. (2009) for Yulin (China); their surface ARF was higher than the surface ARFs for Karachi but in a similar range to those for Lahore. Kim et al. (2005) estimated an ARF between −13 and −43 W m^{-2} for three ground sites in Eastern Asia, which is lower than the range obtained from our results.

Prasad et al. (2007b) reported a surface ARF between −19 and −87 W m^{-2} and a TOA ARF between +2 and −26 W m^{-2} during the whole of the dust period from April to May, 2005, which is comparable to the ARF found in our own study. Pandithurai et al. (2008) reported a surface ARF between −39 W m^{-2} (in March) and −99 W m^{-2} (in June), and an atmospheric forcing between +27 W m^{-2} (in March) and +123 W m^{-2} (in June) over New Delhi in 2006. Bierwirth et al. (2009) reported that ARF at the surface varied between −45 and −65 W m^{-2} over Morocco, which is comparable to our own ARF values over Karachi.

The large differences between TOA and surface forcing (Fig. 6a–b) demonstrate that solar radiation is being absorbed within the atmosphere, and as result the atmosphere get warmer but the earth's surface gets cooler (Alam et al., 2011a; Ge et al., 2010; Miller and Tegen, 1999). This can substantially alter atmospheric stability and influence the dynamic system of the atmosphere (Li et al., 2010). Overall the TOA forcings at Lahore and Karachi are comparable, but the surface forcings are stronger (more strongly negative) at Lahore than for Karachi.

The atmospheric heating rate can be estimated by following Liou (2002) as

$$\frac{\partial T}{\partial t} = \frac{g}{C_P}\frac{\Delta F_{\mathrm{Atmos}}}{\Delta P}$$

where $\partial T/\partial t$ is the heating rate (K day^{-1}), g is the acceleration due to gravity, $\Delta F_{\mathrm{Atmos}}$ is the atmospheric heating, C_P is the specific heat capacity of air at constant pressure and ΔP is the atmospheric pressure. The average heating rate (K day^{-1}) during summer was 2.3 ± 0.1 and 1.2 ± 0.2 and during winter was 1.8 ± 0.4 and 1.1 ± 0.1 over Lahore and Karachi respectively. The high heating rates over both sites during summer and winter were attributed to high atmospheric absorption. During summer, moderately absorbing dust often gets mixed with black carbon (BC), whereas during winter absorbing BC has the highest concentration resulting in high atmospheric heating (Dey and Tripathi, 2008).

Fig. 7a–b shows comparisons between ARF at the surface from AERONET retrievals and from SBDART calculations over Lahore and Karachi. There is good general agreement between the AERONET retrieval and the SBDART calculated forcing at the surface.

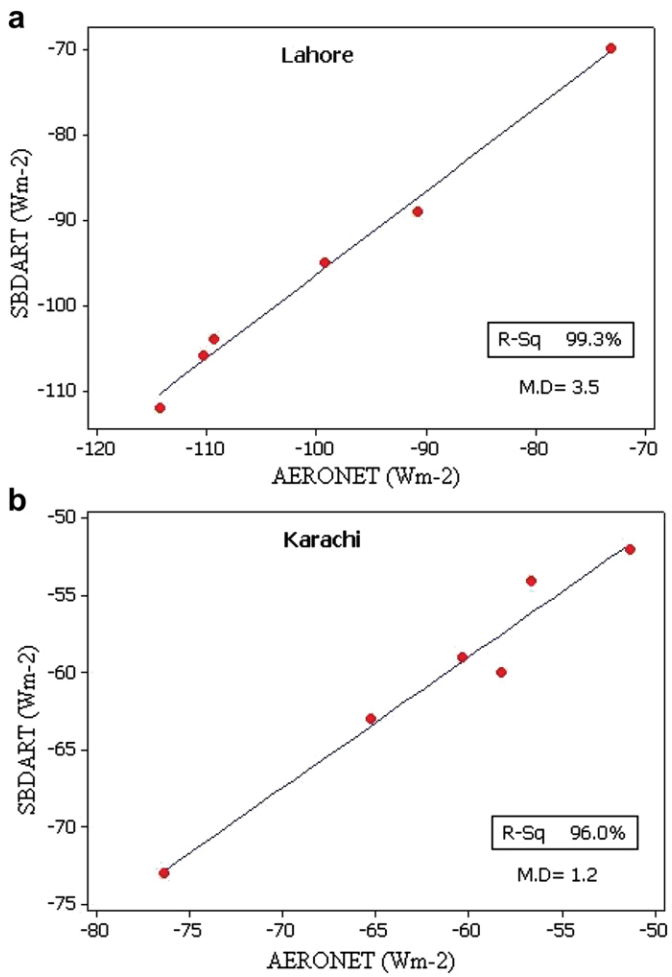

Fig. 7. Comparison of the AERONET derived and SBDART aerosol radiative forcing (W m^{-2}) at the surface of the atmosphere over (a) Lahore and (b) Karachi during the study period.

The AERONET-SBDART correlation coefficients and mean differences are 0.99 and 3.5 W m^{-2} for Lahore, and 0.98 and 1.2 W m^{-2} for Karachi. The good correlation between AERONET and SBDART surface forcing demonstrates that the input parameters, particularly the atmospheric profiles, are an appropriate representation of the prevailing atmospheric conditions. Uncertainties in our ARF calculation may have arisen due to (a) uncertainty in the retrieval of AOD (b) uncertainty in SSAs, (c) uncertainty in the surface albedo values, and (d) uncertainty in ozone concentrations. The overall uncertainty in the estimated forcing due to deviations in simulation has been previously found to be between 10 and 15% (Alam et al., 2011a; Prasad et al., 2007).

4. Conclusion

Aerosol optical properties and ARF have been analyzed over the two megacities of Lahore and Karachi for two seasons (summer and winter) during 2010–11. To the best of the authors' knowledge this has been the first comprehensive assessment of aerosol characteristics for these two megacities in Pakistan. The highest AOD values during the study period were found over Lahore. High AOD values over Lahore during December were largely due to absorbing anthropogenic activities, while high AOD values in June were attributed to mineral dust aerosols. Similarly high AOD over Karachi in June were attributed to dust aerosols, while lower values in winter were due to anthropogenic activities. The relationships between AAE and EAE suggest that urban-industrial and mineral dust aerosols were dominant over other aerosol types at the two investigated sites. The volume size distributions were higher over Lahore for both seasons than over Karachi. The lower SSA and higher $k(\lambda)$ values over Lahore suggest a greater dominance of absorbing aerosols over Lahore than over Karachi. The ARF values were higher for Lahore than for Karachi: the averaged ARF at the surface and at the TOA were -96 ± 13 W m^{-2}, and -22.5 ± 5.9 W m^{-2} for Lahore, and -60 ± 6.8 W m^{-2} and -18 ± 2.2 W m^{-2} for Karachi, giving rise to atmosphere forcing of 74.56 ± 16.8 W m^{-2} for Lahore and 41.85 ± 6.4 W m^{-2} for Karachi. In future studies the long term AERONET-derived aerosol characteristics for the region will be compared to satellite-derived aerosol properties, such as those from the "Cloud and the Earth's Radiant Energy System" (CERES) experiment, and from Moderate Resolution Imaging Spectrometer (MODIS) measurements. Such validations based on satellites, AERONET, and SBDART will provide a better understanding of the regional and local behavior of aerosol optical properties and radiative forcing over this region.

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