Classification of Aerosols in an Urban Environment on the Basis of Optical Measurements

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ABSTRACT

The present study investigates various types of aerosol in the Lahore city of Pakistan using Aerosol Robotic Network (AERONET) data over a six year period from 2007 to 2012. Aerosol optical depths (AODs) observed was in the range 0.2–1.12. An analysis of seasonal variations in AOD has indicated that the highest AOD values occurred in summer and the lowest in winter. The urban aerosols of the study area were classified on the basis of optical parameters such as AOD, Extinction Angstrom Exponent (EAE), Absorption Angstrom Exponent (AAE), Single Scattering Albedo (SSA), Asymmetry Parameter (ASY) and Refractive Index (RI). The AAE values were in the range from 0.25 to 3.2. Real Refractive Index (RRI) and Imaginary part of the Refractive Index (IRI) values were in the range from 1.5 to 1.6 and 0 to 0.005 respectively. The major contributions to the atmospheric aerosols over Lahore were from urban industrial emissions, fossil fuel burning and road/soil dust. Higher RRI values reflected larger re-suspended road dust particles and long-range transported particles, while lower values reflected increased anthropogenic absorbing carbonaceous aerosols over the area. The AERONET retrieved SSA (0.80–0.89) and ASY (0.70–0.83) values suggested a predominance of urban industrial, vehicular and dust aerosols over Lahore. The Derivative of Angstrom Exponent (DAE) was derived at a wavelength of 500 nm and was found to indicate a predominance of fine aerosols across all seasons, particularly during summer and autumn seasons. Back-trajectory analyses using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model revealed that the major air masses over Lahore originated from India, Iran and Afghanistan.

Keywords: Aerosol; AERONET; Classification; AOD; SSA; DAE.

INTRODUCTION

Aerosols consist of a variety of liquid droplets and other particles which are solid in nature, and they remain suspended in the atmosphere, which contribute a lot to the earth-ocean-atmosphere system. Increased urbanization and industrialization and the increasing use of motorized transport are responsible for increased aerosol concentrations in the atmosphere. Aerosols are caused from both natural and man-made sources. High aerosol concentrations have a considerable effect on the climate, as well as on plants, animals and human beings (Dabbas et al., 2012; Hobaib et al., 2013). The properties of aerosol particles (such as size, shape, chemical composition, and optical thickness) are diverse due to their different source origins, and they have very broad temporal and spatial distributions (Wang et al., 2006, 2008). Some of the optical properties of aerosols (e.g., scattering cross section, extinction exponent, single-scattering albedo and phase function) and the microphysical properties (size distribution and, real and imaginary refractive index) are crucially important for determining the effect that aerosols have on the atmosphere (Clarke et al., 2007; Aissani et al., 2012). For the determination of the effects of the aerosols on the earth radiations and climate, remote sensing is an essential tool (Tripathi et al., 2005). However, comprehensive details of the properties related to the optical behavior and other characteristics of aerosols can’t be retrieved through satellite data (Eck et al., 2005). Therefore, Aerosol Robotic Network (AERONET) ground-based instrumentation can be used in this respect (Holben et al., 1998). Ground based remote sensing is a strong and reliable method for the determination of aerosol characteristics (Dubovik et al., 2000). This technique, however, cannot be employed to cover the whole globe (Dubovik et al., 2002).
One of the techniques used for aerosol characterization is to use the Aerosol Optical Depth (AOD), the amount of sunlight, which is either absorbed or scattered by aerosols (Alam et al., 2014). Many investigations have been conducted into aerosol characteristics and their associated properties, for example by Russell et al. (1999), Raes et al. (2000), and Huebert et al. (2003). Other researchers such as Ge et al. (2010), Liu et al. (2008), and Zheng et al. (2008) have investigated the seasonal, spatial, and temporal variations in the characteristics of aerosols. Higher AODs were reported over different parts of India in summer while lower AOD reported in winter (Ramanathan et al., 2001; Ranjan et al., 2007; Singh et al., 2010).

Many investigations have also been carried out into the possibility of using the properties of aerosols (optical and physical) to differentiate between different classes of aerosol. Some authors (e.g., Dani et al., 2010; More et al., 2013) have suggested that biofuel burning and fossil fuel consumption made a significant contribution to the aerosol loading over different Indian regions. The relation of AOD with Angstrom Exponent (AE) is used to identify dominant aerosol classes, source regions, and aerosol transport mechanisms (Giles et al., 2012). Similar information on particle type, size, and growth can be obtained from AE and the particle effective radius. Some of the major aerosol species were reported by Gobbi et al. (2007) through the relation between absorption and aerosol size parameters. Yu et al. (2009) showed extensive results regarding particle size, composition, and absorption parameter in order to classify natural and man-made aerosols. Hostler et al. (2008), and Reagan et al. (2004) used cluster analysis to generate aerosol classes. Two dimensional cluster analyses for the Scattering and Absorption Angstrom Exponent (SAE, AAE) were used by Clarke et al. (2007) to differentiate between different species of aerosols i.e., dust, fossil fuel burning and industrial emissions. Dust was distinguished from carbon (soot), using cluster analysis by Yang et al. (2009). Russell et al. (2010a) suggested using cluster analysis on AAEs and EAEs derived from AERONET to distinguish between different types of aerosol.

Previous investigations have revealed different types of aerosols over Lahore (Alam et al., 2012, 2014) but these investigations were only conducted over short periods of time, providing only short-term information on AAE, EAE, SSA, RI, and ASY values.

In this work we have used long-term (2007–2012) AERONET data in conjunction with cluster analysis to obtain detailed information on aerosol properties specially related to the optical behavior (such as AOD and AAE) and their relationships to other parameters such as EAE, SSA, ASY, RRI, and IRI. Optical parameters such as AOD, AAE, EAE, RI, SSA, and ASY have been used to identify various classes of aerosol over the mega city of Lahore.

DATA AND METHODS

Site Description

Pakistan is blessed with so many resources like high mountains in the north with thick forests, hilly areas to the west full of minerals, eastern regions with favorable monsoonal rainfall, and a southern coastline with harbors. Due to the topographical variation, Pakistan has got a unique location for environmental studies.

Lahore, the capital city of Punjab in Pakistan lies between 31°15′ and 31°45′ N, and between 74°01′ and 74°39′ E. The Ravi River is in the northern part of Lahore, and to the east of the city is the border with India. It is amongst the populated city in the world with approximately 10 million population. Its area is 404 square kilometers (156 square miles) and is still increasing with the passage of time. The climate is hot and semi-arid with extreme summer conditions prevailing during the months of April and June (with maximum temperatures in the range of 33 to 44°C and with cooler winter conditions lasting from December to February (with maximum temperature from 17 to 22°C). The rainy season starts in late June and lasts until August, bringing heavy rainfall to the city. The sources of aerosols over Lahore are derived mainly from local and long range transported dust particles, vehicular and industrial emission, biomass burning, and fossil fuel combustions (Biswaess et al., 2008; Alam et al., 2011).

Meteorological Conditions

Fig. 1 presents monthly averages of meteorological conditions over Lahore between 2007 and 2012, including data on wind speed and direction, minimum and maximum temperatures, and relative humidity obtained from the Lahore station of the Pakistan Meteorological Department. The highest average temperatures ranged from 16.6°C to 41.8°C, with the minimum average temperatures from 6.9°C to 28.7°C, and the average relative humidity from 20% to 68%, as shown in Fig. 1(a). The wind speeds ranged from 0.15 m s⁻¹ to 2.6 m s⁻¹, with the lowest speed in November 2007 and the highest in March 2012. During the study period, the winds were mainly from northwest (September–May) accompanied by small variations in southwest, southeast, and northeast, Fig. 1(b).

Instrumentation

Aerosol Robotic Network (AERONET)

AERONET is a network of ground-based instruments, that is used to obtain aerosol properties at wavelengths of 340, 380, 440, 500, 670, 870, 940, and 1020 nm (Dubovik et al., 2000). Uncertainties in AERONET AODs are of the order of ± 0.01 for wavelengths longer than 440 nm and ± 0.02 for shorter wavelengths (Alam et al., 2011; More et al., 2013). The error in particle size measurements is about 10% for small particles and as much as 35 to 100% for large particles (Alam et al., 2011). The uncertainties in SSA range between about 0.03 and 0.05 depending on the aerosol type and source (Dubovik et al., 2000). The real and imaginary parts of RIs have errors of 30–50% and ± 0.04 respectively (Dubovik et al., 2002).

Three level data are available from AERONET: Level 1.0 is unscreened data, Level 1.5 is cloud screened (Smirnov et al., 2000), and Level 2.0 is cloud screened and quality assured (Alam et al., 2011). The AERONET data are available online from NASA's AERONET website (http://aeronet.gsfc.nasa.gov/).
AERONET has been operational over Lahore since December 2006. In this study, we have used Level 2 data for the period from 2007 to 2012, making use of the properties of AOD, AE, AAE, EAE, RI, SSA and ASY.

**Hybrid Single Particle Integrated Trajectory (HYSPLIT)**

We used the HYSPLIT model to estimate back trajectories in order to determine the sources of the air masses over Lahore. The HYSPLIT model has a horizontal grid of 1.5° × 1.5° and a resolution of 500 × 500 m. HYSPLIT can be used to calculate up to 40 forward or backward trajectories at different altitudes, (Draxler and Rolph, 2003). The HYSPLIT website (http://ready.arl.noaa.gov/HYSPLIT.php) can be used to run this model, but to avoid computational saturation of the web server, this web version is configured with some limitations (Alam et al., 2010).

**RESULTS AND DISCUSSION**

**Variations in Monthly and Seasonal Averages of AERONET AODs**

The AOD is the degree to which transmission of light can be prevented by aerosols through scattering and absorption phenomenon. Aerosol concentrations show both spatial and temporal variations which affect their various optical properties. We have examined temporal variations in aerosol particle concentrations over Lahore using AERONET data over the six years from 2007 to 2012. Fig. 2 shows the monthly mean AODs over Lahore with the highest mean value was 1.07 ± 0.32. The seasonal variations in AOD and their STDs are summarized in Table 1. The AERONET AOD values were highest during autumn. Low AOD values were recorded during winter by both AERONET. Similar
HYSPLIT Back Trajectories

For this study, we computed backward trajectories over a few days at different altitudes during the investigated period (shown in Fig. 3) in order to determine the provenance of the air masses over Lahore. The aerosol patterns over Lahore were assessed by a 5-day back-trajectory analysis using the NOAA HYSPLIT model. Previous computations (Alam et al., 2011c) of air mass back trajectories for Pakistan have indicated different sources for summer and winter. Increased AOD values observed in northern India have previously been identified by back trajectories as being due to air masses originating from the Sahara, and the Thar Desert in Rajasthan (Kuniyal et al., 2009). The highest AOD values in India are also reported in summer because of winds bringing dust from the Horn of Africa and the Arabian Peninsula (Li., 2009; Ramanathan, 2002).

Some previous studies (Lodhi et al., 2009; Alam et al., 2011b) have identified local pollutants (due to dust, fossil fuel burning, and urban industrial emissions) over Pakistan. As shown in Fig. 4, High AOD values are due to the dust particles mainly from the Thar Desert in India that combine with local urban industrial emissions. Biswass et al. (2008) and Lodhi et al. (2009) also reported that local and neighboring western India were the major contributors of aerosols over Lahore. Other studies have reported that air masses are reaching Pakistan from the south-west and surrounding areas in winter by travelling long distances, but in summer the air masses have travelled shorter distances (Alam et al., 2011, 2014). The air masses shown in Fig. 3 arrive over Lahore from Dasht (Iran), the Sahara Desert, the Arabian Sea, and Afghanistan, increasing the aerosol concentrations in the region.

AAE and EAE

Fig. 4(a) shows a scatter plot of AAE values vs. EAE values over Lahore during the period from 2007 to 2012. The results indicate that industrial/vehicular emissions and road/soil dust aerosols over the city are similar to those obtained by Alam et al. (2012). The figure shows that AAE values ranged from 0.25 to 3.2. The AAE value was reported as 1 for black carbon (Russell et al., 2010b), diesel particle emissions (Schnaiter et al., 2003), urban pollution (Clarke et al., 2007), and motor vehicle emissions (Arnott et al., 2009). The carbon emissions in Lahore are due to industry and to vehicular exhaust (Lodhi et al., 2009; Stone et al., 2010). High AAE values (such as 1.45) have previously been linked to biomass burning (Russell et al., 2010b). A high value of 2.1 for biomass burning was reported by Clarke et al. (2007), and Arnott et al. (2009) observed higher values to a maximum of 3.5. Russell et al. (2010b) reported AAE values of 2.34 for dust from the Sahara Desert and 2.27 for a mixture of dust and pollution. The high AAE values (1–2) that we observed for Lahore reflect fossil fuel and agricultural residues burning in the nearby agricultural areas, and the values ranged from (2–3.2) for AAE represent mineral dust comprising both local and long-range transported dust.

Table 1. AERONET derived Seasonal average AODs and Standard Deviations over Lahore during 2007–12.

| Seasons | AERONET (AOD ± SD) |
|---------|---------------------|
| Winter  | 0.38 ± 0.18         |
| Spring  | 0.59 ± 0.27         |
| Summer  | 0.74 ± 0.26         |
| Autumn  | 0.93 ± 0.44         |

Fig. 2. Monthly average variations in AERONET AOD at 550 nm over Lahore between 2007 and 2012.

Fig. 3. AERONET AOD at 500 nm over Lahore between 2007 and 2012.
Fig. 3. Five-days HYSPLIT back trajectories.
Figs. 4(b), 4(c), 4(d), and 4(e) show scatter plots of AAE values vs. EAE values for winter, spring, summer and autumn, respectively. Fig. 4(b) shows AAE values from 0.5 to 2.8, but most of the AAE values ranged from 1 to 1.5, representing carbonaceous aerosols (Schnaiter et al., 2003; Clarke et al., 2007; Russell et al., 2010a). A large proportion of the carbon aerosols over Lahore originate from the combustion processes (Liaquat et al., 2007). Local source emissions, e.g., motorized transport, brick kilns (Stone et al., 2010), power plants, industries (Alam et al., 2011b), and fertilizer factories in Multan (Qadir and Zaidi, 2006), are the main contributors of pollutants over Lahore, particularly during winter. The similar sources for winter emissions have also been reported by Biswass et al. (2008) for urban atmospheres in South Asia.

Fig. 4(c) shows AAE values (in spring) that ranged from 0.32 to 3.2, representing soil or road dust and pollution (Russell et al., 2008, Lodhi et al. (2009), and Stone et al. (2010) reported that dust particles were transported to Lahore from the neighboring city of Amritsar, and also from northern India.

The AAE values in summer ranged from 0.36 to 2.2 (Fig. 4(d)) reflecting an increase in aerosols from biomass burning and dust particles, and a reduction in the concentration of carbonaceous aerosols (Dutkiewicz et al., 2008). The area surrounding Lahore is largely agricultural and the burning of agricultural residues is practiced throughout the area; wood is also used for cooking and residential heating. Combustion of these types of bio-fuel can contribute significantly to aerosol particle emissions within the region.

Fig. 4(e) shows a scatter plot for AAE values vs. EAE values during autumn. The AAE values ranged from 0.5 to 3.2. The data for this season reveal the lowest concentration of carbonaceous aerosols and significant quantities of dust comprising both locally derived and long-range transported dust particles (Dutkiewicz et al., 2008; Russell et al., 2010b).

**RRI vs. AAE and IRI vs. AAE**

The complex refractive index has been analyzed in this study, providing information on scattering and absorbing types of aerosols (Alam et al., 1983; Sinyuk et al., 2003; Alam et al., 2011a, 2012).

Figs. 5(a)–5(e) show a scatter plot for RRI values vs. AAE values. The RRI values in this study ranged between 1.37 and 1.6. The highest RRI (1.5–1.6) and AAE (1–2) values represent larger dust particles, especially road dust and long-range transported dust particles (Liu et al., 2008; Alam et al., 2011a). Similar results were obtained by Tripathi et al. (2005) over Kanpur, in India, and our results are also consistent with those reported by Dubovik et al. (2002a) in Bahrain and Solar Village, Saudi Arabia.

Figs. 6(a)–6(e) show a scatter plot of IRI values vs. AAE values. The IRI values ranged from 0.0005 to 0.024 over Lahore. These values are consistent with soot (Dubovik et al., 2002a; Raut et al., 2006) which is a major component of urban carbonaceous aerosols (Arola et al., 2011). The IRI values and AAE values for Lahore show high level of anthropogenic absorbing carbonaceous aerosols. These aerosols derived mainly from traffic exhaust, industries, the combustion processes (Liaquat et al., 2007; Dutkiewicz et al., 2009; Lodhi et al., 2009; Stone et al., 2010; Alam et al., 2011). The results are consistent with that of Kanpur, India, by Singh et al. (2004) and Tripathi et al. (2005).

**SSA vs. AAE**

SSA is the ratio of the efficiency which shows the scattering to total efficiency known as extinction efficiency. An SSA value equal to unity implies that all particle extinction is due to scattering and an SSA value of zero implies that all extinction is due to absorption. The SSA is a key parameter in studies of aerosol radiative forcing (Dubovik et al., 2002; Alam et al., 2012). SSA values are greatly dependent on wavelength, increasing with increasing wavelength (Russell et al., 2010a, b).
Figs. 7 and 8(a)–8(d) show a scatter plot of SSA values vs. AAE values, with SSA values (0.80–0.98). Results with similar SSA values were also obtained by Ramanthan et al. (2001b). In Fig. 8 the lower values for SSA (0.80–0.89) indicate a predominance of urban and industrial anthropogenic aerosols over Lahore. These results are consistent with those obtained previously over Lahore, e.g., by Alam et al. (2012) and Ali et al. (2013). Zheng et al. (2009) obtained similar results over different areas in China for different absorbing locally derived urban aerosols. Our results are also consistent with the findings of Singh et al. (2004) over Kanpur and Singh et al. (2010) over Delhi. The larger SSA (0.90–0.98) and AAE (1–2) values can be attributed to combinations of absorbing aerosols and dust particles
Fig. 5. The same as Fig. 4, but for AAE versus RRI

(Dubovik et al., 2002; Ali et al., 2013). Bibi et al. (2016) discriminated aerosol into three major types (dust, biomass burning and urban/industrial) over four locations in the Indo-Gangetic Plains.

**ASY vs. AAE**

The Asymmetry Parameter (ASY) describes the scattering of light interacting with aerosol particles. Figs. 9(a)–9(e) show a scatter plot of ASY values vs. AAE values. The ASY values ranged from 0.70 to 0.83. The lower ASY values (0.70–0.74) represent absorbing aerosols that result from anthropogenic activities. Alam et al. (2011a, 2012) have previously reported ASY values of between 0.61 and 0.72 for Lahore. The ASY values calculated in this study are also close to the values reported by Srivastava et al. (2011) for the area of India. The larger ASY values (0.73–
Fig. 6. The same as Fig. 4, but for AAE versus IRI.

0.83) reflect a predominance of dust particles over Lahore. Similar values have been observed over different areas in China (Yu et al., 2006, 2009, 2011).

**DAE vs. AOD**

The Derivative of the Angstrom Exponent (DAE) reflects the dependence of the AE on wavelength. The DAE was calculated using the following formula (Eck et al., 1999):

\[
AE' (\lambda_i) = \frac{d\text{DAE}}{d\ln \lambda} = -\left( \frac{2}{\ln \lambda_{i+1} - \ln \lambda_{i-1}} \right) \times \left[ \frac{\ln r_{i+1} - \ln r_i}{\ln \lambda_{i+1} - \ln \lambda_i} \right] \times \left[ \frac{\ln \lambda_{i+1} - \ln \lambda_{i-1}}{\ln \lambda_{i} - \ln \lambda_{i-1}} \right] \tag{1}
\]

In this work the DAE was calculated for a wavelength
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Figs. 7 & 8. The same as Fig. 4, but for SSA versus AAE.

of 500 nm, using AOD values observed at 340 nm, 500 nm, 675 nm and 870 nm.

A DAE greater than zero denotes fine aerosol particles, which are related to anthropogenic activities such as biomass burning (Eck et al., 1999, 2001), while a negative DAE denotes coarse aerosol particles. Moderate to high AE values have previously been reported for some regions of India by Sing et al. (2010). In summer the DAE values approach zero or are negative, but in the spring both negative and positive DAEs have been reported (Kaskaoutis et al., 2006, 2007). In our findings, Fig. 10(a) shows the overall temporal trend of DAE values, which indicates a predominance of fine mode aerosols over the entire study period. Figs. 10(b), 10(c), 10(d), and 10(e) show the seasonal variation in DAE with respect to AOD at a wavelength of 500 nm. The same dominance of fine mode aerosols continues across all
seasons, in particular, the DAE values are nearly positive in the autumn and winter seasons. Similar trends have also been reported previously for New Delhi (Soni et al., 2011).

CONCLUSION

The AODs over Lahore from AERONET data, together with aerosol optical characteristics from both direct and inversion products, were used to classify aerosol over the 6 year period from 2007 to 2012. The salient features of the present study are summarized in the following lines:

- Higher monthly average AOD values were reported during the summer months and lower values in winter.
- Results for AAE and EAE indicated the presence of particles derived from vehicular and industrial emissions, biomass burning, and re-suspension of road dust.

Fig. 9. The same as Fig. 4, but for ASY versus AAE.
Fig. 10. Temporal trend of the DAE over Lahore (a) The whole period (b). Scatter plot of AOD vs. AE over Lahore during the Summer (c) Spring (d) Autumn (e) Winter seasons between 2007 and 2012.

- The SSA values were high representing the presence of dust over Lahore. High RRI, low IRI, and large ASY values can be attributed to large dust particles and anthropogenic absorbing carbonaceous aerosol particles.
- A back-trajectory analysis has revealed that the air masses over Lahore originated mostly from the Thar Desert in India, Dasht in Iran, and from Afghanistan.
- A variety of sources contribute to the overall pollution in Lahore, including particles from vehicular and industrial emissions, biomass burning, and locally derived dust, as well as a substantial amount of dust from neighboring regions.
- The DAE results indicated the predominance of fine mode aerosols across all seasons.
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