

Atmospheric aerosols properties over Indo-Gangetic Plain: A trend analysis using ground – Truth AERONET data for the year 2009–2017

Akhilesh Kumar^{a,d}, Vineet Pratap^a, Sarvan Kumar^b, A.K. Singh^{a,c,*}

^a Atmospheric Research Laboratory, Department of Physics, Institute of Science, Banaras Hindu University, Varanasi 221005, India

^b Department of Earth and Planetary Sciences, V.B.S. Purvanchal University, Jaunpur 222003, India

^c DST - Mahamana Centre of Excellence in Climate Change Research, Banaras Hindu University, Varanasi, India

^d Kashi Naresh Government Post Graduate College Gyanpur, Bhadohi 221304, India

Received 11 June 2021; received in revised form 28 December 2021; accepted 31 December 2021

Available online 6 January 2022

Abstract

The aerosols play an important role in the modification of the regional radiation budget. Long-term trend analysis of properties of atmospheric aerosols is important for policymakers and also for the study of the climatic implications. The Aerosol Robotic Network (AERONET) provides continuous ground-truth aerosol data during cloud-free days since two decades in India. Long-term atmospheric aerosol properties have been studied over four different sites of Indo-Gangetic Plain (IGP): Gandhi College rural background, Kanpur, urban and highly industrial area, Jaipur urban and desert dust influenced region, and Lahore urban and agriculture rich region respectively using AERONET data. The yearly variation of aerosol optical depth (AOD) along with angstrom exponent (AE), single scattering albedo (SSA), size distribution parameters (volume concentration and effective radius), and aerosol radiative forcing (ARF) have been analyzed for nine years from 2009 to 2017. A positive trend of AOD over Kanpur (0.0074/year), a negative trend over Lahore (−0.0054/year), Jaipur − 0.0027/year) and Gandhi college (−0.0008/year) was found. Volume concentration shows the increasing trend of fine mode particles which may be due to increased anthropogenic activities in comparison to natural aerosols over Kanpur, Lahore, Gandhi College, and Jaipur respectively. SSA values over Kanpur and Jaipur show dominance of scattering while absorbing nature of particles over Lahore and Gandhi College were found. The trend of total atmospheric radiative forcing (ARF_{ATM}) over Kanpur and Jaipur locations was found to be decreased slightly but not so significantly while over Lahore and Gandhi College trend was found to increase in a significant way. The yearly trend of ARF_{ATM} over Kanpur, Lahore, Jaipur, and Gandhi College was found to be $-0.76 \text{ Wm}^{-2}/\text{year}$, $2.22 \text{ Wm}^{-2}/\text{year}$, $-0.08 \text{ Wm}^{-2}/\text{year}$, and $3.09 \text{ Wm}^{-2}/\text{year}$ respectively.

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Keywords: Aerosol optical depth; Angstrom exponent; Size distribution parameters; Single scattering albedo; Radiative forcing; AERONET

1. Introduction

Atmospheric aerosols are playing important role in the atmospheric processes. It can affect radiation budget, air

quality, human health, climate (Jethva et al., 2018; Taylor, 2010; Vaishya et al., 2017), and hydrological cycle (Rosenfeld et al., 2008). Aerosol particles are classified by fine modes as well as coarse mode particles. Fine mode aerosols include gaseous contaminants (NO_2 , SO_2 , etc.), viruses, tobacco smoke, soot, etc. Whereas coarse mode aerosols are sea salt, dust particles, etc. (Dubovik et al., 2002; Tiwari et al., 2019). Aerosol particles have direct as

* Corresponding author at: Atmospheric Research Lab, Department of Physics, Institute of Science, Banaras Hindu University, Varanasi, India.
E-mail address: singhak@bhu.ac.in (A.K. Singh).

well as indirect effects on the atmosphere. It can prevent incoming radiations towards the surface of the earth directly by absorbing and scattering process (Liou, 2002; Miller and Tegen, 1998). During severe dust storm days, long-range transportation of aerosol particles plays important role in aerosol loading, increased concentration of coarse mode particles, etc (Kumar et al., 2015; Tiwari et al., 2019). The dust particles mixed with fine mode anthropogenic aerosols are called brown carbon that increase the concentration of aerosol particles and can affect the biosphere, cryosphere, and eco-system of Earth (Gautam et al., 2013; Ramanathan et al., 2005; Srivastava and Ramachandran, 2013) and also cause global dimming (Stanhill and Cohen, 2001). Indo – Gangetic plain (IGP) is one of the most polluted and populated plains where aerosol loading is high round the year. It experiences frequent dust storms occurred in a mid-arid region, as a result, there is the transportation of dust particles from one place to another place (Tiwari et al., 2019). It is found that the central and lower parts of IGP are characterized by high aerosol loading with the dominance of mixed and fine aerosols (Kumar et al., 2018). In IGP there are dominance of only two types of aerosols, which are dust and anthropogenic aerosols. A major source of anthropogenic aerosols is urban-industrial areas, but polluted dust can be in a wide range (Masoumi et al., 2013). In a recent study, long-term aerosol characteristics over IGP for the year 2007–2017 were analyzed, and found an increasing trend of aerosol optical depth (AOD) and angstrom exponent (AE) suggesting an increasing concentration of fine mode particles (Kumar et al., 2021). Inhalable aerosols or particulate matters (PM₁₀ and PM_{2.5}) significantly affect human health through the respiratory system (Ren-Jian et al., 2012; Oh et al., 2020). The health effects of aerosols are also debatable in scenarios of the COVID-19 pandemic as its spread is likely to be more airborne and aerosols can spread at long distances (Kumar, 2020). Long-term trends of aerosols and their optical properties are of particular interest because their changes can have broad consequences on the Earth's climate system. Examining trends of different aerosol properties is important to understand the impact of anthropogenic activities on the earth-atmosphere system (Collaud Coen et al., 2013). Numerous studies have been done to understand the properties of aerosols and their radiative impacts (Kedia et al., 2018; Singh et al., 2016), but still, there is large uncertainty in earth's radiation budget (IPCC, 2013; Kaskaoutis et al., 2016; Kumar et al., 2015) since morphology and chemical composition of dust particles are intricate (IPCC, 2013; Kim et al., 2011; Lafon et al., 2006). Developments and anthropogenic activities over the Indian region are increasing continuously therefore air quality of this region is strongly affected. The effect of AOD on aerosol radiative forcing is well-known, but the size distribution, single-scattering albedo (SSA), and morphology also affect aerosol radiative forcing (ARF) (He et al., 2015; Stocker et al., 2013; Wu et al., 2016). An increasing trend

of aerosol optical thickness over rapidly populating areas in the Indian subcontinent was found (Kishcha et al., 2011). Several long term studies of aerosol optical thickness over the Indian region (Mehta, 2015; Prijith et al., 2018; Srivastava and Saran, 2017), middle east (Klingmüller et al., 2016), Eastern Mediterranean (Tutsak and Koçak, 2019), and over China (Zhao et al., 2017) were carried out. A long-term trend of aerosol parameters (like; AOD, SSA, AE) and their climatology are studied by several researchers over IGP (Kumar et al., 2021; Kumar et al., 2018; Lodhi et al., 2013). But, the trends of other parameters such as ARF and size distribution parameters (volume concentration, effective radius) are not yet completely analyzed. So, the long-term trends of these parameters are also important and need to be studied over IGP.

In the present study, long-term atmospheric aerosol properties have been studied over four different sites of IGP: Kanpur, urban and highly industrial area, Gandhi College rural background, Jaipur urban and desert dust influenced region, and Lahore urban and agriculture rich region using AERONET ground truth data. The yearly variations of AOD, SSA, ARF, and size distribution parameters along with the long-term trend of the same parameter are studied. These aerosol inversion parameters (SSA, size distribution parameters, and ARF) also play an important role and have a significant contribution to the climate modeling projections.

2. Site description, data, and methodology

Four different locations of IGP are considered in this study. Different sites are shown in Fig. 1 with different symbols. The four different sites in the Indian subcontinent region: Gandhi College (25.87 °N, 84.13 °E), rural background, Kanpur (26.51 °N, 80.23 °E), urban and highly industrial area, Jaipur (26.91 °N, 75.81 °E) urban and desert dust influenced region, and Lahore (31.48 °N, 74.26 °E) urban, dust and agriculture rich region are chosen to study all the parameters (AOD and inversion parameters such as; SSA, size distribution parameters and ARF), using ground-truth AERONET observations. The long-term aerosol characterizations during the period 2009–2017 from the CIMEL sun/sky radiometer measurements were carried out. AERONET sun-photometer measures aerosol optical depth (AOD) at eight different wavelengths within the range from 0.34 to 1.02 µm (0.34, 0.38, 0.44, 0.50, 0.67, 0.87, 0.94, and 1.02 µm) and columnar water vapour (at 0.94 µm) using direct sun measurements. The phase function, asymmetry parameters (AP), refractive indices of aerosols, and SSA were retrieved with the help of the inversion method (Dubovik and King, 2000; Holben et al., 1998). In India, a CIMEL sun/sky radiometer is installed at different sites under the AERONET program of NASA, USA. The processing algorithms have evolved from Version 1.0 to Version 2.0 and now Version 3.0. The Version 3.0 databases are available from the AERONET and PHOTONS websites. Version 3.0 AOD

Table 1
Statistical parameters for the regression analysis in the yearly trend over Kanpur, Jaipur, Lahore and Gandhi College during 2009–2017. The statistically significant trends are taken at the 95% confidence level ($p < 0.05$).

Year (2009–2017)	Kanpur			Lahore			Jaipur			Gandhi College		
	Trend (/year)	(%)	p	Trend (/year)	(%)	p	Trend (/year)	(%)	p	Trend (/year)	(%)	p
AOD (500 nm)	0.0074	9.68	0.39	−0.0054	−7.40	0.66	−0.0027	−4.92	0.68	−0.0008	−0.98	0.97
AE(440–870 nm)	0.019	17.61	0.092	0.0218	20.71	0.052	0.0297	35.11	0.096	0.0361	32.04	0.033
SSA(440 nm)	0.0023	2.26	0.053	−0.0037	−3.72	0.040	0.0002	0.19	0.91	−0.0053	−5.24	0.25
SSA(1020 nm)	0.0004	0.39	0.87	−0.0039	−3.85	0.004	−0.0018	−1.72	0.51	−0.0078	−6.86	0.14
VolC-C	−0.0142	−60.56	0.012	−0.0107	−41.08	0.007	−0.0139	−57.70	0.015	0.0032	15.59	0.74
VolC-F	0.0031	35.82	0.004	0.0015	17.61	0.42	0.0016	33.05	0.004	0.0021	24.44	0.31
Reff-C	0.0223	8.58	0.045	−0.0084	−3.29	0.48	0.0225	9.13	0.17	0.0199	7.80	0.42
Reff-F	0.0042	24.06	0.006	0.0022	13.56	0.075	0.0017	10.86	0.058	0.0011	6.42	0.58
Radiative Forcing	−0.76	−11.62	0.35	2.22	31.24	0.078	−0.08	−1.70	0.94	3.09	38.30	0.22

data are computed for three data quality levels: Level 1.0 (unscreened), Level 1.5 (cloud-screened and quality controlled), and Level 2.0 (quality-assured) (Smirnov et al., 2000). In this study, we have used Version 3.0 and Level 1.5 data as this was the only available dataset for all the parameters. This level data is also used previously by others (Tiwari et al., 2013; Srivastava et al., 2011; Prasad et al., 2007) and the deviation of Level 1.5 AOD value from level 2.0 is found within the range of 0–5% over IGB (Tiwari et al., 2013). During the AOD calculation there is uncertainty which is less than ± 0.01 for higher wavelengths ($\lambda > 0.44 \mu\text{m}$) (under cloud-free condition) and less than ± 0.02 for shorter wavelengths; whereas columnar water vapor has high uncertainty which rises to 10 % (Dubovik et al., 2000; Eck et al., 1999).

The key parameters to study the characteristics of atmospheric aerosols are AOD and AE which can be simply calculated using the equation given by Ångström, (1964):

$$\tau_a(\lambda) = \beta \lambda^{-\alpha} \quad (1)$$

where λ is the wavelength, τ_a is AOD for the wavelength λ , α is angstrom exponent (AE) and β is the turbidity coefficient which is equal to columnar AOD at $\lambda = 1 \mu\text{m}$. It is observed that for shorter wavelengths AODs are higher whereas at longer wavelengths AOD values are relatively lower showing the presence of particle concentration from fine to coarse (Reddy et al., 2011; Tiwari and Singh, 2013). On the other hand, AE (α) tells about fine mode ($r < 1 \mu\text{m}$) as well as coarse mode ($r > 1 \mu\text{m}$) fraction (Tiwari et al., 2018). A higher value of α indicates the dominancy of fine mode particles while a lower value tells about coarse mode particles.

Single Scattering Albedo is one of the important radiative properties of the aerosol. It is the most significant parameter which can tell about the scattering and absorbing nature of different types of aerosols and has an important role in the calculation of atmospheric radiative forcing. SSA is defined as the ratio of scattering to the total extinction of light (Steinfeld, 1998). Its value lies between zero and one. Zero value indicates completely absorbing (black carbon or soot particles) while one indicates completely scattering particles like sulphate. It is observed that SSA also depends on the size and composition of aerosol particles (Bergstrom et al., 2007). In the presence of absorbing aerosols, the SSA value decreases with the increase of wavelength and in the presence of scattering aerosols, the SSA value increases with the increase of wavelength (Alam et al., 2012; Tiwari et al., 2013).

Aerosol radiative forcing (ARF) is defined as the net difference in solar fluxes in the presence of aerosols and in the absence of aerosols whether it is at top of the atmosphere (TOA) or surface (BOA) respectively, mathematically;

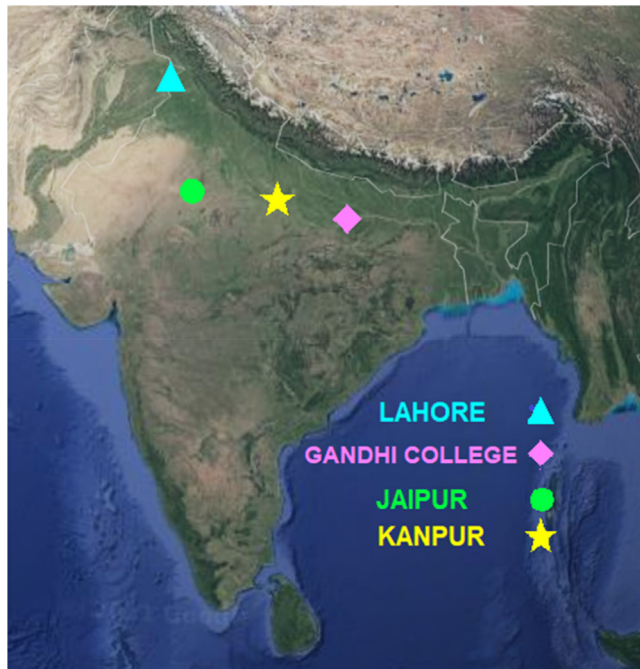


Fig. 1. Background map of study locations; Kanpur, Lahore, Jaipur and Gandhi College.

$$\Delta F = (F_{a\downarrow} - F_{a\uparrow}) - (F_{0\downarrow} - F_{0\uparrow}) \quad \text{in the unit of } W/m^{-2} \quad (2)$$

where ΔF is the irradiance, and $(F_{\downarrow} - F_{\uparrow})$ shows the net irradiance which is calculated in the presence of aerosol (F_a) and absence of aerosol (F_0) either at TOA or at BOA (Alam et al., 2012; Steinfeld, 1998). It is one of the key parameters in the atmosphere which tells about the heating or cooling of the climate system. In this study, ARF (at TOA and BOA) is taken from the AERONET. To calculate the radiative forcing, AERONET uses a radiative transfer model, GAME (Global Atmospheric Model) (Scott, 1974). Radiative forcing for TOA and BOA is defined as (García et al., 2012);

$$\Delta F_{TOA} = F_{0\uparrow,TOA} - F_{a\uparrow,TOA} \quad (3)$$

$$\Delta F_{BOA} = (F_{a\downarrow,BOA} - F_{0\downarrow,BOA}) \cdot (1 - SA) \quad (4)$$

where downward arrow (\downarrow) shows downward flux, an upward arrow (\uparrow) shows upward flux, and SA is defined as the surface albedo.

2.1. Trend analysis

A long-term trend analysis of aerosol optical properties was performed on time series from four sites situated in IGP. The trend analysis was applied to yearly mean data of various aerosol optical properties obtained from AERONET for all the locations during the period of 2009–2017, and the methodology of trend analysis is explained in Kaskaoutis et al. (2012) as;

$$x\% = \left(a * \frac{N}{\bar{x}} \right) * 100$$

where x is the variable, \bar{x} is the mean value, N is the whole number of days during the studied period and a is the slope value from the linear regression analysis. Further, the statistical significance of the slope was checked with the p -value, which was considered to be less than 0.05 for statistically significant variations at the 95% confidence level. Statistical parameters for the regression analysis in the yearly trend over Kanpur, Jaipur, Lahore and Gandhi College during 2009–2017 are tabulated in Table 1.

3. Results and discussion

3.1. Dependency of SSA on wavelength

Fig. 2 shows the wavelength dependence of SSA for March, April, and May over Kanpur for the year 2009. The SSA value is selected for four different wavelengths. Single scattering albedo is associated with the absorbing or scattering behaviour of aerosol particles, either they are coarse mode or fine mode particles in nature.

3.2. Aerosol optical depth (AOD) and angstrom exponent (AE)

Fig. 3 shows the yearly variation of AOD and AE over four different locations (Kanpur, Lahore, Jaipur, and Gandhi College) and their trends for the study period 2009 to 2017. Annual mean values of AOD and AE were found to be $(0.68 \pm 0.23$ and $0.97 \pm 0.30)$, $(0.67 \pm 0.20$ and $0.95 \pm 0.26)$, $(0.49 \pm 0.15$ and $0.78 \pm 0.33)$ and $(0.74 \pm 0.29$ and $1.03 \pm 0.27)$ over Kanpur, Lahore, Jaipur and Gandhi college respectively from year 2009–2017. On the basis of numerical values, AOD might be indexed in three classes: (a) dust dominated ($AOD > 0.5$; $AE < 0.5$), (b) mixed ($AOD > 0.5$; $AE = 0.5$ – 1.0) and (c) pollution dominated ($AOD > 0.5$; $AE > 1.0$) (Tutsak and Koçak, 2019). Over the Indo – Gangetic plain region (Kanpur, Lahore, and Gandhi College) high $AOD > 0.5$ and $AE > 0.5$ values suggest mixed aerosol loading. On the other hand, higher $AE > 0.5$ over the region suggests an abundance of fine mode particles. While comparatively low AOD values were found over Jaipur which suggests low aerosol loading during 2009–2017. Due to its seasonal variability, there is mixed aerosol loading over Indo – Gangetic plain (Kumar et al., 2012; Tiwari and Singh, 2012). The long-term trends over four sites also have been studied (Fig. 3). A slightly insignificant decreasing trend of aerosol optical depth (AOD) over Lahore ($-0.0054/\text{year}$, $p = 0.66$), Gandhi College ($-0.0008/\text{year}$, $p = 0.97$) and Jaipur ($-0.0027/\text{year}$, $p = 0.68$) and a slightly increasing trend of AOD over Kanpur ($0.0074/\text{year}$, $p = 0.39$) were found. While significantly increasing trend of angstrom exponent (AE) over Kanpur ($0.019/\text{year}$, $p = 0.092$),

Lahore (0.0218/year, $p = 0.052$), Jaipur (0.0297/year, $p = 0.096$), and Gandhi College (0.0361, $p = 0.033$) were found which shows dominance of fine mode particles which might be due to the increasing influence of anthropogenic aerosols. A positive trend of AOD and AE indicates fine mode aerosol loading which may be either absorbing or scattering in nature. On the other hand, the negative trend of AOD shows the scavenging of aerosol particles. A positive trend of AOD may depend on other factors like soil moisture, increasing temperature, and decreasing relative humidity leading to enhanced dust emissions and anthropogenic aerosols (Klingmüller et al., 2016). AOD has a direct relation to aerosol loading in the atmospheric column. A positive trend of AOD tells that aerosol loading is continuously increasing over the IGP, which may be the reason that industrialization and urbanization in IGP are increasing rapidly. Consequently, this aerosol loading is consistent with enhanced anthropogenic activities (Krishna Moorthy et al., 2013; Ramachandran et al., 2012; Srivastava and Saran, 2017). The population of India is increasing in an uncontrolled way and it was found to be increased by 181 million from 2001 to 2011 (Census 2001 and Census 2011). About 2.5 times vehicles and 27,600 new industries were found to be increased (Road transport yearbook 2009 – 2010 and 2010 – 2011, Gov. of India). Due to these activities, concentrations of black carbon, organic carbon, and other pollutant have enhanced (Lu et al., 2011; Pratap et al., 2020; Kumar et al., 2020). This may be a possible reason for the positive trend of AOD and AE respectively. IGP frequently experience an increasing number of dust storms in the summer season (April, May, and June) (Kumar et al., 2015; Tiwari et al., 2019) which have other important contribution and possible increasing trend of aerosol. Using satellite and ground-based observations similar results have also been found by the others (Babu et al., 2013; Ramachandran and Cherian, 2008). The trend of AOD over India and surrounding oceanic regions using satellite

observations have also been studied and increasing trends were found (Tiwari et al., 2015; Prijiith et al., 2018).

3.3. Single scattering albedo (SSA)

The yearly variation and trend of SSA (440 nm) and SSA (1020 nm) for the years 2009–2017 are shown in Fig. 4. Annual mean (\pm SD) of SSA (440 nm) and SSA (1020 nm) over Kanpur, Lahore, Jaipur and Gandhi College were found to be (0.91 ± 0.03 and 0.92 ± 0.03), (0.89 ± 0.03 and 0.91 ± 0.04), (0.90 ± 0.03 and 0.94 ± 0.04) and (0.91 ± 0.04 and 0.91 ± 0.05) respectively. Overall the four stations' relatively higher values of SSA were found (>0.9) which normally occurs during dusty days. SSA value was calculated over Kanpur during pre-monsoon dust storm days and it was found to be ≤ 0.95 (Kumar et al., 2015). A very high value of SSA (≥ 0.98) has been found during the severe dust storm that occurred on 5th July 2014 over Karachi (Iftikhar et al., 2018). A higher SSA value (0.89–0.976) over Jaipur was reported showing more scattering nature of aerosol particles (Payra et al., 2013). In our study SSA values are neither very high nor very low i.e. moderate values of SSA have been found overall the locations, which show moderately absorbing as well as moderately scattering nature of particles.

A positive trend of SSA (440 nm) and SSA (1020 nm) over Kanpur (0.0023/year, $p = 0.053$ and 0.0004/year, $p = 0.87$) has been found while a negative trend of SSA (440 nm) and SSA (1020 nm) over Lahore (-0.0037 /year, $p = 0.040$ and -0.0039 /year, $p = 0.004$) and Gandhi College has been found. But, over Jaipur station positive trend of SSA (440 nm) (0.0002/year, $p = 0.91$) and a negative trend of SSA (1020 nm) (-0.0018 /year, $p = 0.51$) has been found. It is reported that when SSA increases with increasing wavelength, it indicates the existence of scattering aerosols (coarse particles), whereas a decrease in SSA with increasing wavelength tells about the presence of absorbing aerosols (fine particles) (Dubovik et al., 1998; Dubovik et al., 2002; Sumit et al., 2012). The positive trends of SSA (440 nm) and SSA (1020 nm) over Kanpur show that scattering particles are abundant over Kanpur. In contrast, Lahore and Gandhi College SSA have decreasing trend which hints at the possibility of aerosol loading particles that are absorbing in nature. On the other hand trend of SSA (440 nm) over Jaipur is increasing and the trend of SSA (1020 nm) is decreasing which shows there is a mixing of aerosol particles. Recently, Shaik et al. (2019) examines the spatial, seasonal, and interannual variation of biomass burning and its impact on regional aerosol optical properties over Northern India where SSA was found to be 0.75 to 0.91 during pre-monsoon to post-monsoon which is absorbing in nature while in the winter season, it was found in the range 0.89 to 0.91 which is moderately absorbing. At Kanpur, aerosol particles in the pre-monsoon season were found to be scattering ($SSA > 0.9$) in nature (Kumar et al., 2015; Tiwari et al., 2019). The maximum value of SSA in

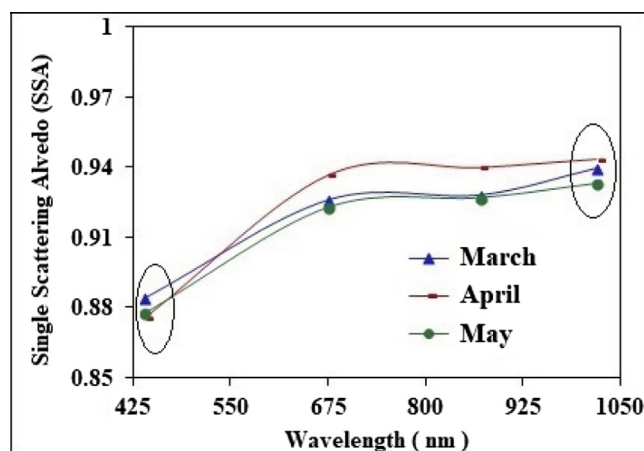


Fig. 2. Wavelength dependent SSA over Kanpur for the year of 2009.

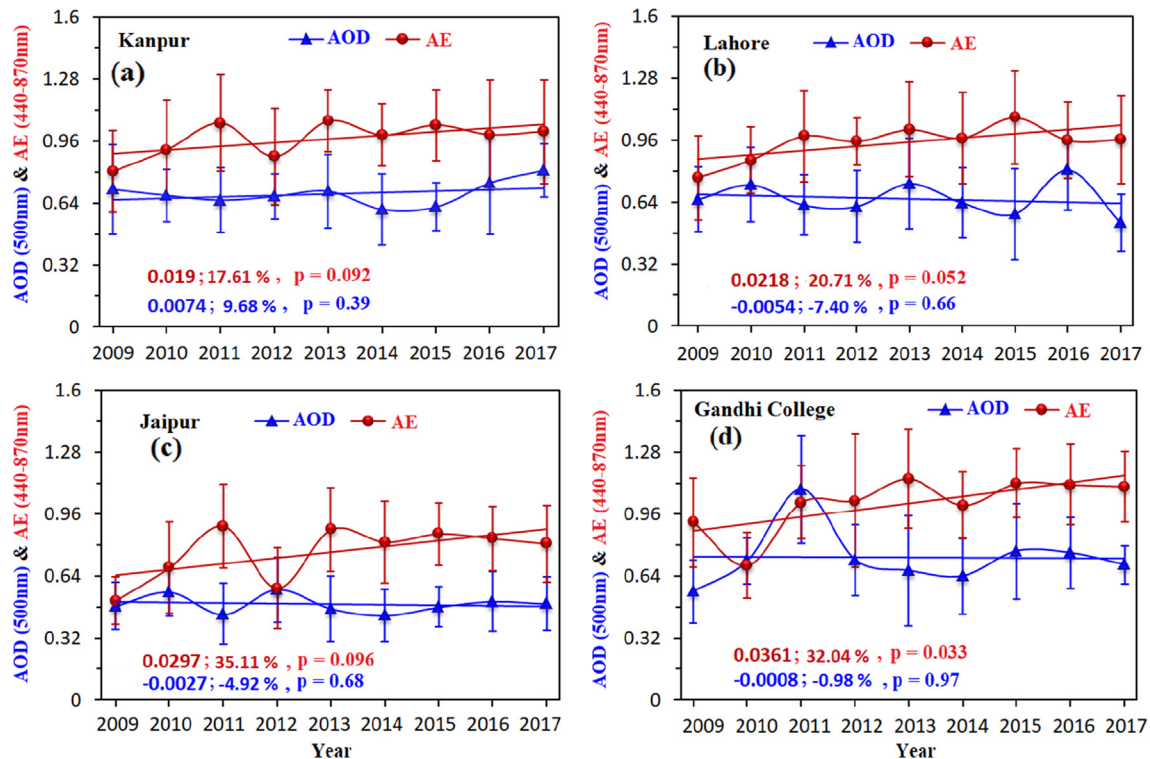


Fig. 3. Yearly variation and trend of AOD and AE for the year 2009–2017.

the summer season and minimum value of SSA in the winter season over Zanjan during 2006 – 2008 have also been reported by Masoumi et al. (2013). Seasonal variability of SSA and special events like dust storms, forest fires, and other anthropogenic activities play a significant role in the trend of single scattering albedo. Zhao et al. (2017) have done a similar study and investigated the trend of SSA over Eastern and Central China, Western Europe, and the Eastern United States during 2001–2015, and decreasing trend over the entire regions was found.

3.4. Size distribution parameters

Fig. 5 shows yearly variation and trend of volume concentration for fine mode, a coarse mode for the year 2009–2017 over Kanpur, Lahore, Jaipur, and Gandhi College respectively. Annual mean (\pm SD) of volume concentration (in the unit of $\mu\text{m}^3/\mu\text{m}^2$) for fine mode and coarse mode over Kanpur, Lahore, Jaipur and Gandhi College were found to be $(0.078 \pm 0.009$ and $0.21 \pm 0.05)$, $(0.08 \pm 0.01$ and $0.23 \pm 0.03)$, $(0.044 \pm 0.005$ and $0.22 \pm 0.05)$ and $(0.08 \pm 0.01$ and $0.18 \pm 0.06)$ respectively. From these observations, it can be seen that the mean value of volume concentration for coarse mode aerosols over Gandhi college was found to be relatively lower as compared to Kanpur, Lahore, and Jaipur. On the other hand, volume concentration for fine mode aerosols over Jaipur (near the Thar Desert) was found to be low as compared to Kanpur, Lahore, and Gandhi College. Interestingly, a negative trend of volume concentration for coarse mode

particles over all stations (except Gandhi College) (Fig. 5a, c, e, g) and a positive trend of volume concentration for fine mode particles over the regions (Fig. 5b, d, f, h) was found. Over Kanpur, Lahore, Jaipur, and Gandhi College trend of volume concentration (per year) for coarse mode / fine mode particles were found to be $(-0.0142 / 0.0032)$, $(-0.0107 / 0.0015)$, $(-0.0139 / 0.0016)$ and $(0.0032 / 0.0021)$ respectively. Overall the three locations (Kanpur, Lahore, and Jaipur) trends were found to be significantly decreasing having p-value 0.012, 0.007, and 0.015 over Kanpur, Lahore, and Jaipur respectively which shows that the concentration of coarse mode particles decreasing. The trend of volume concentration over Gandhi college was found to be slightly positive having a low significant value ($p = 0.74$). While, significantly increasing trend of fine mode volume concentration were found over Kanpur ($p = 0.004$), Lahore ($p = 0.007$), Jaipur ($p = 0.015$), and Gandhi College ($p = 0.31$) which suggest dominance of fine mode aerosols. Noticeably, overall the trend of the above region of volume concentration for coarse mode aerosol was found to be decreasing which shows that the concentration of coarse mode particles decreasing and increasing portion of anthropogenic aerosols continuously. In addition, a trend of the effective radius was also analyzed for fine mode and coarse mode aerosol particles.

Fig. 6 shows trend analysis of effective radius for fine mode as well as coarse mode aerosol particles. Over Kanpur location, a significantly increasing trend of R_{eff} for fine mode (with trend 0.0042/year and $p = 0.006$) and coarse mode (0.0223/year, $p = 0.045$) particles was found

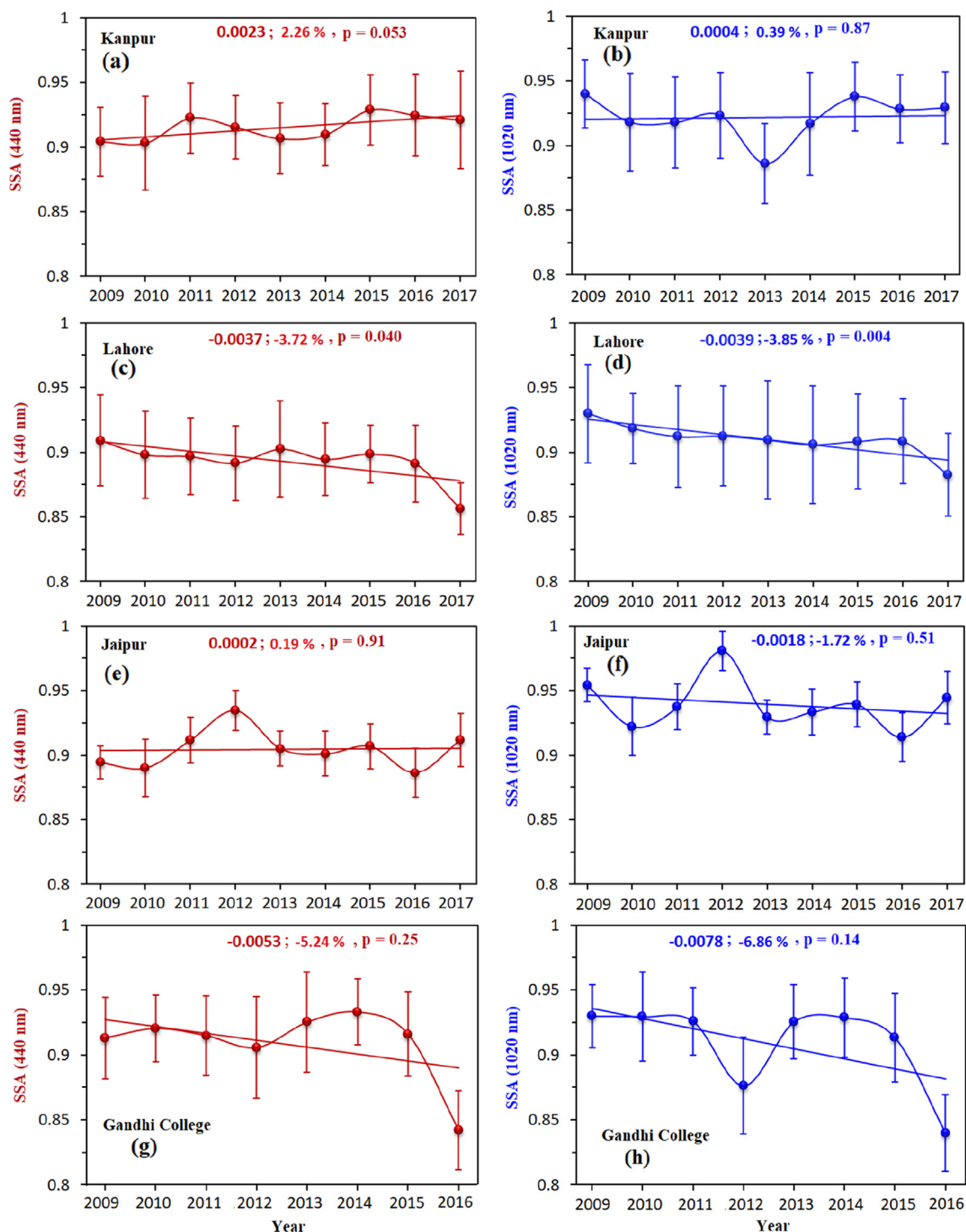


Fig. 4. Yearly variation and trend of SSA (440 nm) and SSA (1020 nm) for the year 2009–2017.

(Fig. 6a, b). For Lahore, a trend of R_{eff} for fine mode particles was found to be significantly increasing (0.0022/year, $p = 0.075$) while R_{eff} for coarse mode particles was found to be decreasing ($-0.0084/\text{year}$) but with low significance ($p = 0.48$) (Fig. 6c, d). Over the Jaipur region, it was found that the trend of R_{eff} for fine mode particles significantly increased (0.0017/year and $p = 0.058$) and the trend of R_{eff} for coarse mode particles increased slightly (0.0225/year

and $p = 0.17$) (Fig. 6e, f) while Jaipur is near the Thar Desert. It suggests there may be enhanced anthropogenic activities. Likewise, volume concentration, a trend of R_{eff} over Gandhi College was found to be slightly increased for coarse mode (0.0199/year, $p = 0.42$) and fine mode particles (0.0011/year, $p = 0.58$) (Fig. 6g, h). Since there is the event of dust storm during the pre-monsoon season (Alam et al., 2014; Kaskaoutis et al., 2019; Kumar et al., 2015;

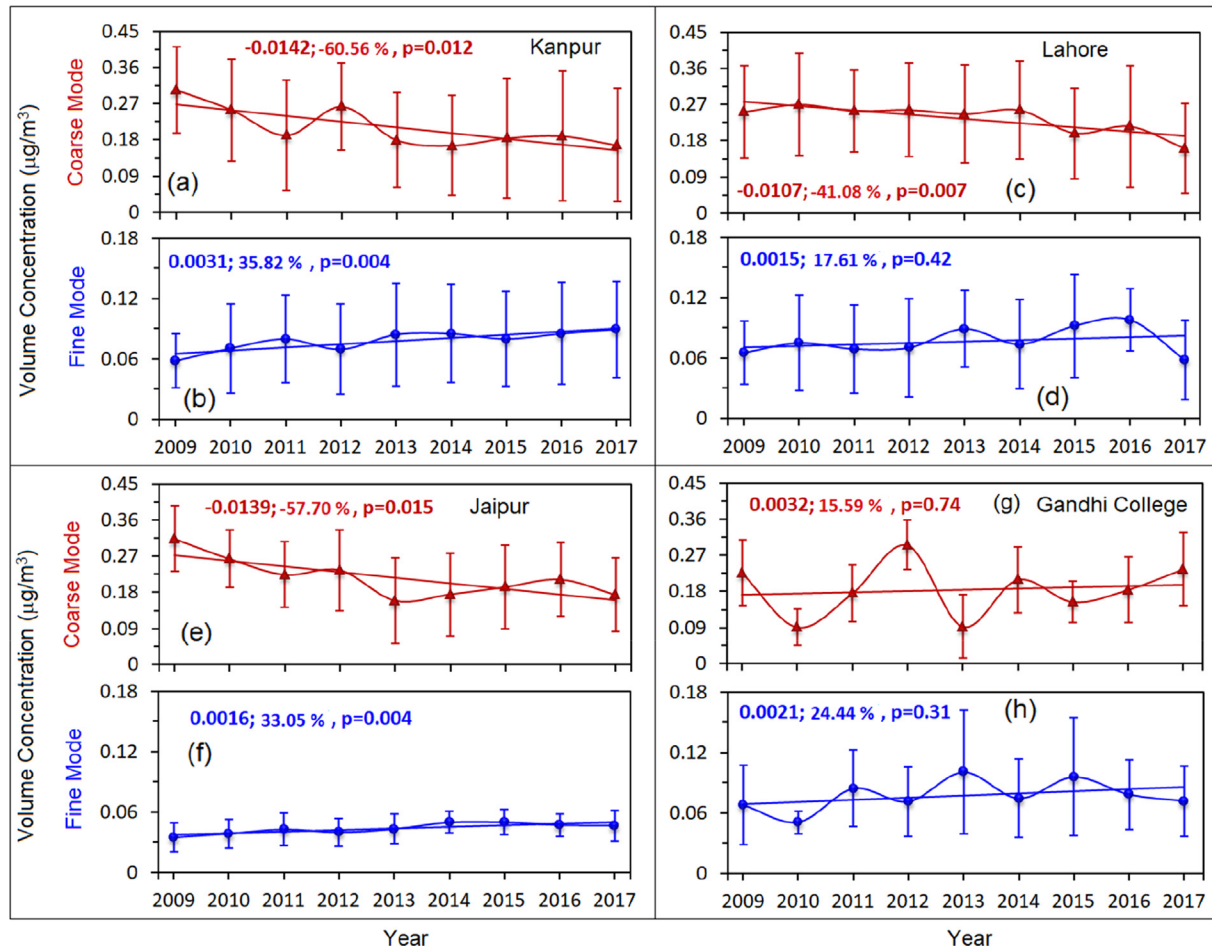


Fig. 5. Yearly variation of volume concentration for fine mode and coarse mode aerosol particles for the year 2009–2017.

Tiwari et al., 2019), biomass burning during the winter season (Shaik et al., 2019) which can modify the aerosol concentration. Due to the high rate of increase of urbanization, anthropogenic activities are rapidly and playing an important role in the modification of optical (AOD, AE, SSA, etc) and physical (size) properties of the aerosol.

3.5. Aerosol radiative forcing (ARF)

The yearly variation of aerosol radiative forcing (ARF) at the TOA, BOA, and in the atmosphere (ATM) over Kanpur, Lahore, Jaipur, and Gandhi College is shown in Fig. 7. The average value of ARF over Kanpur during the year 2009–2017 was found to be $-27.51 \pm 6.62 \text{ W/m}^{-2}$, $-86.42 \pm 23.54 \text{ W/m}^{-2}$, and $58.91 \pm 21.02 \text{ W/m}^{-2}$ at TOA, BOA, and in the atmosphere (ATM) respectively. Over Lahore, the average value of ARF during the same period was found to be $-30.14 \pm 9.46 \text{ W/m}^{-2}$, $-94.86 \pm 26.52 \text{ W/m}^{-2}$, and $64.73 \pm 23.96 \text{ W/m}^{-2}$ at TOA, BOA, and in the atmosphere (ATM) respectively. The average value of ARF over Jaipur for the year 2009–2017 was found to be $-25.56 \pm 6.86 \text{ W/m}^{-2}$, $-67.67 \pm 15.63 \text{ W/m}^{-2}$, and $42.11 \pm 14.41 \text{ W/m}^{-2}$ at TOA, BOA, and in the atmosphere

(ATM) respectively, and, over Gandhi College, the average value of ARF during the same period was found to be $-26.99 \pm 9.76 \text{ W/m}^{-2}$, $-89.81 \pm 25.28 \text{ W/m}^{-2}$, and $62.81 \pm 26.42 \text{ W/m}^{-2}$ at TOA, BOA, and in the atmosphere (ATM) respectively.

Positive radiative forcing in the atmosphere (ATM) indicates warming and negative forcing shows cooling of the atmosphere. Overall the four regions ARF at the TOA and the BOA were found to be negative showing a cooling effect. As a consequence, ARF value in the atmosphere (ATM) was found to be positive which shows a warming effect on the atmosphere. Radiative forcing is event-specific i.e. it is also affected by a special event like dust storms, biomass burning, forest fires, etc. Recently, ARF_{ATM} values over Kanpur were reported during dust storms by Tiwari et al. (2019) which was found to be 124.40 W/m^2 on 17 May 2018 and 84.94 W/m^2 on 14 June 2018 respectively. While, relatively low value of ARF_{ATM} over Kanpur during a dust storm on 20 April 2010 (75.11 W/m^2), 28 May 2010 (60.65 W/m^2), and 2 June 2010 (37.13 W/m^2) were reported (Kumar et al., 2015). But all the above values during the dust storm days were found to be higher than the normal days. During the dust event, the daily averaged value ARF_{ATM} was found to be

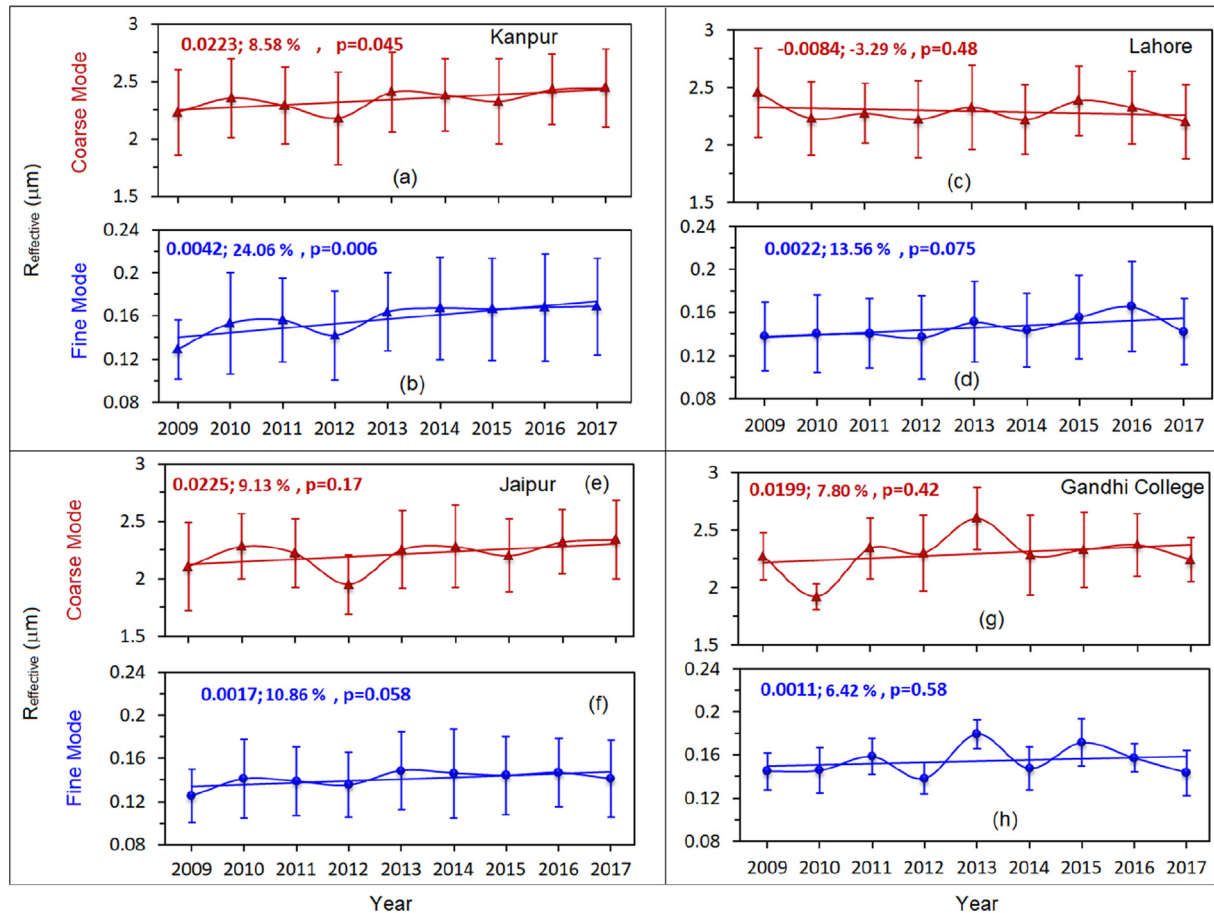


Fig. 6. Yearly variation of effective radius ($R_{\text{effective}}$) for fine mode and coarse mode aerosol particles for the year 2009–2017.

25–52 W/m^2 over Karachi and 47–77 W/m^2 over Lahore respectively while during the haze event it was found to be 17–67 W/m^2 (Iftikhar et al., 2018). Radiative forcing also shows seasonal dependency and it was reported by Ramachandran and Kedia, (2012). During the pre-monsoon season the maximum value of ARF_{ATM} ($>30 \text{ W/m}^2$) over Gandhi College, was found for the year 2006–2008. While for the rest seasons (winter, monsoon, and post-monsoon) it was found to be $< 30 \text{ W/m}^2$.

The long-term trend of aerosol radiative forcing (ARF) in the atmosphere (ATM) over the IGP region (Kanpur, Lahore, and Gandhi College) and Jaipur in North-western India for the year 2009 to 2017 is shown in Fig. 7. The trend of ARF_{ATM} over Kanpur, Lahore, Jaipur, and Gandhi College were found to be $-0.76 \text{ Wm}^{-2}/\text{year}$, $2.22 \text{ Wm}^{-2}/\text{year}$, $-0.08 \text{ Wm}^{-2}/\text{year}$, and $3.09 \text{ Wm}^{-2}/\text{year}$ with p – values 0.35, 0.078, 0.94 and 0.22 respectively from 2009 to 2017. Since ARF_{ATM} directly depends on AOD, so positive trend of AOD leads to the increasing trend of ARF_{ATM} (Kumar et al., 2015). The trend of ARF_{ATM} over Lahore and Gandhi College was found to be higher than those of Kanpur and Jaipur respectively. A less negative value of TOA forcing is due to low surface albedo (low SSA) which leads to the higher

value of ATM forcing, TOA forcing depends on SSA as well as AOD (Ramachandran and Kedia, 2012). So, a higher value of trend over Lahore and Gandhi College may be due to decreasing trends over the same region.

4. Summary and conclusions

The present study mainly focused on the long-term variation and trend of aerosol parameters like AOD, AE, SSA, size distribution parameters, and aerosol radiative forcing over Kanpur, Lahore, Jaipur, and Gandhi College respectively. All the parameters were taken using ground-based AERONET measurement for the years 2009 to 2017. The key findings of the present study are summarised as

- The yearly variations of AOD, SSA, ARF, and size distribution parameters along with the long-term trend of the same parameter were studied.
- Over the IGP region at three sites (Kanpur, Lahore, and Gandhi College) high AOD > 0.5 and AE > 0.5 values with a positive trend suggest mixed aerosol loading. On the other hand, higher AE > 0.5 over the region suggests an abundance of fine mode particles. While com-

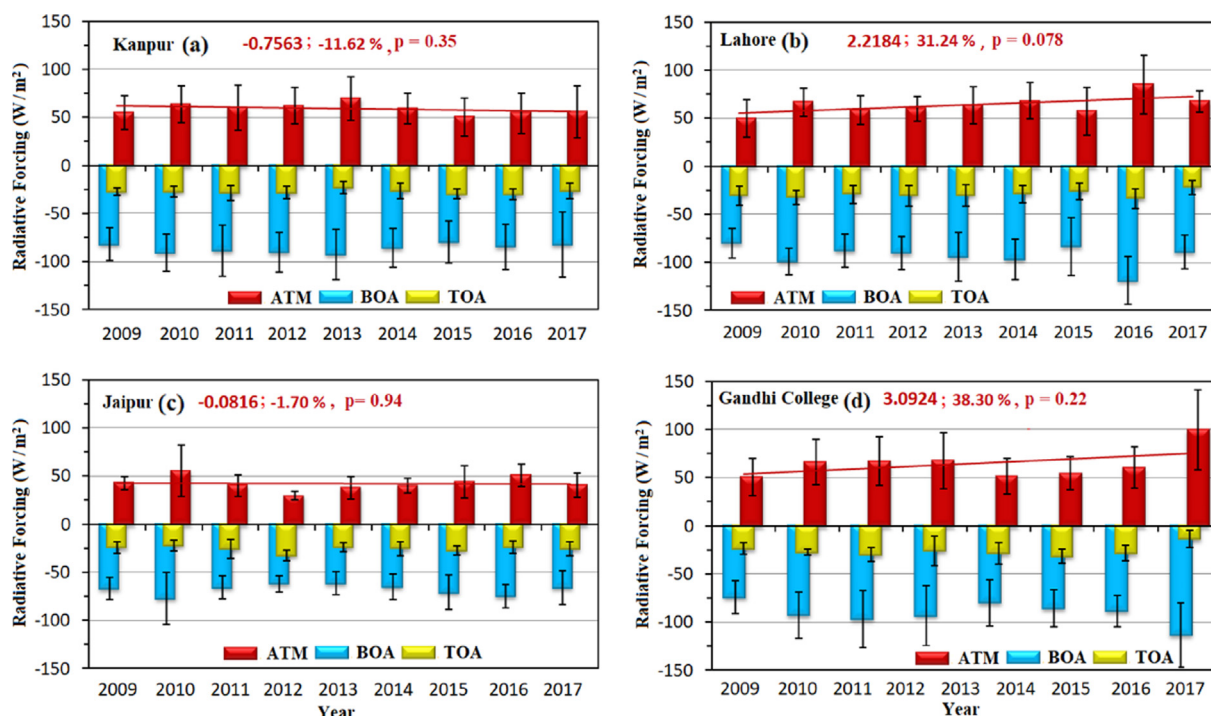


Fig. 7. Yearly variation of ARF (TOA, BOA and ATM) for the year 2009–2017.

paratively low AOD value (0.49 ± 0.15) was found over the Jaipur site with a negative trend which suggests low aerosol loading during 2009–2017.

- The trend of the aerosol volume concentration was found to be negative (decreasing) for coarse mode particles and slightly increasing (positive) for fine mode particles over all the regions of study. The increasing trend of fine mode particles may be due to the increase of anthropogenic activities.
- SSA shows the scattering and/or absorbing nature of particles, the positive trend of SSA over Kanpur indicates that scattering particles are abundant over Kanpur. In contrast, Lahore and Gandhi College SSA have decreasing trend which hints at the possibility of aerosol loading particles that are absorbing in nature. On the other hand trend of SSA (440 nm) over Jaipur is increasing and the trend of SSA (1020 nm) is decreasing which shows there is the mixing of aerosol particles i.e. they are both scatterings as well as absorbing in nature.
- The trend of ARF_{ATM} over Kanpur, Lahore, Jaipur, and Gandhi College were found to be $-0.76 \text{ Wm}^{-2} / \text{year}$, $2.22 \text{ Wm}^{-2} / \text{year}$, $-0.08 \text{ Wm}^{-2} / \text{year}$, and $3.09 \text{ Wm}^{-2} / \text{year}$ respectively from 2009 to 2017. A positive trend of radiative forcing over Lahore and Gandhi College suggests an increase of local climate by the definite value of ARF_{ATM} per year and a negative trend over Kanpur and Jaipur suggests cooling which can play a significant role in the atmospheric phenomenon (like monsoon circulation and hydrological cycle).

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

We are thankful to AERONET NASA scientific research team for providing aerosol data. Mr. Akhilesh Kumar is also thankful to the Department of Science and Technology, New Delhi for providing INSPIRE research fellowship. The work is partially supported by the Institute of Eminence (IoE) (Scheme No: 6031) to BHU.

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