Indian J Phys (January 2023) 97(1):17–23
https://doi.org/10.1007/s12648-022-02375-3

ORIGINAL PAPER

Variability of air quality and aerosol over Indian region during 2003–2012

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Received: 02 August 2021 / Accepted: 16 April 2022 / Published online: 22 May 2022

Abstract: The Indo-Gangetic basin is one of the biggest, most populated and polluted regions in the world. Satellite- and ground-based data show strong seasonal variability of aerosol loading with a maximum during pre-monsoon (summer) season. In this study, decadal (2003–2012) variability of aerosol optical depth (AOD) derived from AERONET measurements over Kanpur is presented which indicates maximum AOD during 2003 and minimum during 2012. The aerosol size distribution (ASD) exhibits an increase in the radius and a decrease in the width of distribution. It shows an enhancement during 2003 and 2010 and diminished values during 2004, 2007, 2009 and 2011. The ASD is found to show a peak during summer season (pre-monsoon) throughout 2003–2012 only except the years 2008 and 2011. Moreover, for years 2008 and 2011 ASD showed a peak value during a monsoon month (July). Changes in the spectrum of ASD are explained in terms of surface temperature and precipitation. Seasonal variation of aerosol radiative forcing and its climatic impacts have also been discussed.

Keywords: Aerosol size distribution; Aerosol optical depth; AERONET; Radiative forcing

1. Introduction

Atmospheric aerosols from both natural and anthropogenic sources affect air quality, radiation budget, environmental conditions and cloud phenomena, which ultimately results in climate change and human health/wealth [1–3]. A variety of sources including varying trends in aerosol loading and extreme heterogeneity in the temporal and spatial variability of optical and microphysical properties create much greater uncertainty in quantifying the climatic uncertainty than that of greenhouse gasses [3, 4]. Climatic forcing occurs both directly through the modification of atmospheric radiation budget and indirectly by acting as cloud condensation nuclei (CCN) which affect cloud albedo, cloud lifetime, precipitation rate, hydrological cycle, monsoon system, etc., and as a result climate change/radiative properties of the atmosphere [2, 5, 6]. The presence of heterogeneous mixture of optical, physical and chemical properties of dust in atmospheric aerosols poses considerable uncertainties in global radiation budget quantification [7]. Their spatial, vertical, and temporal distributions cause further complication which depends on regional variations of soil characteristics, mixing of dust with other aerosol species including dust/minerals transported through sand and dust storms from long-distance source region [8]. Aerosols do not only perturb the radiation energy balance of the Earth–atmosphere system, but also influence heating and stability of the atmosphere [9] and chemical and biological ecosystems [10]. The arid regions of Africa (Saharan Desert), Middle East, Arabian Desert, Afghanistan and Thar Desert (Rajasthan, India), located several thousand kilometres upwind of the western side of the Indo-Gangetic (IG) plains, are some of the hot spots of the world [8, 11] and add a large fraction of mineral dust compared with total annual loading of the regions.

The impact of any atmospheric change is commonly quantified by radiative forcing, the imposed change to the radiative energy balance. In particular, changes at the top of the atmosphere (TOA) are of interest because they summarize the overall impact on the Earth's climate. For

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aerosol, the radiative forcing changes in magnitude and even in sign depending on local aerosol and environmental properties. The overall (spectrally, daily and regionally integrated) impact (often a difference larger numbers) is highly uncertain even when integrating overtime.

Recently, several techniques such as ground-based measurements from Aerosol Robotic Network (AERONET) [12] and satellite-based measurement of Polarization and Directionality of the Earth's Reflectance's (POLDER), Total Ozone Mapping Spectrometer (TOMS), Moderate Resolution Imaging Spectroradiometer (MODIS) and Multi-angle Imaging Spectroradiometer (MISR)) have been used widely to study the optical properties of aerosol over the IG basin [11, 13–17].

Measurements showed the high aerosol loading during the pre-monsoon season (April–June) is attributed to the large range transport of large influx mineral dust aerosols through dust storms from western arid regions as well as relatively larger amount of eruption of dust from earth surface [11, 16, 18–20]. Kumar et al. [21] validated ground-based AOD from AERONET with those from satellite (MODIS, MISR) in different seasons and found an increasing trend of AOD over IG basin during 2005–2009. The winter season is interesting one because dust storm activity is absent which is the major source for AOD loading in IG basin from non-local sources, and the anthropogenic component has dominance presence [14]. Previous workers have shown that AOD over the IG basin has been increasing since 2000 [11, 21, 22]. Using volume size distribution (VSD) technique, Zhang et al.[23] analysed AOD data collected at Chengdu, China, and showed that the fine-mode volume concentration was higher than the coarse mode on the annual basis. The fine-mode volume concentration and median radius were higher in summer and winter. The higher value of fine-mode single-scattering albedo suggests that the coarse-mode aerosols have a stronger absorbing effect on solar light than that of the small-sized aerosol particles.

In the present study, ground- and satellite-based data are analysed with a focus on the radical changes observed during different seasons in aerosol optical properties (AOD), aerosol size distribution (ASD) and aerosol radiative forcing (ARF) at Kanpur in the IG basin during a decade 2003–2012. Data used are briefly described in Sect. 2, results and discussion are given in Sect. 3, and finally, Sect. 4 summarizes the findings of this study.

2. Data used

Aerosol optical depth data have been obtained using level-3 MODIS (Moderate Resolution Imaging Spectroradiometer) Terra (MOD08_M3, in HDF format) as a monthly gridded average in $1^\circ \times 1^\circ$ spatial resolutions (http://modis.gsfc.nasa.gov/) [24–26]. The MODIS launched onboard the NASA's Earth Observing System (EOS) Terra and Aqua spacecrafts provides daily global data of aerosol characteristics using 36 spectral bands ranging from visible to thermal infrared (0.41–14.38 μm), with spatial resolutions of 1000 m, 500 m and 250 m (pixel size at nadir) in December 1999 and onboard EOS Aqua satellite in May 2002.

The AERONET (http://aeronet.gsfc.nasa.gov/) program is a general ground-based remote sensing aerosol network to evaluate aerosol optical properties and validate satellite-derived aerosol optical properties [27]. We have used the AERONET version 2, level 2 cloud-screened data in the analysis of optical properties during 2003–2012. Aerosol size distribution (ASD), peak and geometrical width of volume particle size distribution [dV(r)/dlnr ($\mu m^3/\mu m^2$)], derived from AERONET at Kanpur station, are used to recognize and describe trend during 2003–2012. The aerosol products are usually derived from AERONET data following the approach discussed by Dubovik and King [28] and Dubovik et al. [29]. In addition, we have also derived aerosol radiative forcing.

Water vapour and other meteorological parameters are considered from the Atmospheric Infrared Sounders (AIRS) onboard on Aqua satellite [30]. In this study, daily daytime ascending surface temperature (AIRS3STD_V06) with spatial resolution $1^\circ \times 1^\circ$ is taken from GIOVANI, NASA. The monthly rainfall and precipitation rate product with spatial resolution of $0.25^\circ \times 0.25^\circ$ is taken from Tropical Rain fall Measuring Mission (TRMM) (3B43 Version-6) from the website (https://giovanni.gsfc.nasa.gov/giovanni/).

3. Results and discussion

3.1. Optical properties of aerosol in the IG basin

Aerosol is usually characterized by amount, size, composition and shape. The equivalent optical property is aerosol optical depth (AOD) which is the vertically integrated attenuation due to scattering and absorption of solar radiation. Variation of aerosol optical depth at 550 nm derived from AERONET and MODIS measurements during 2003–2012 is shown in Fig. 1. The maximum AOD $\sim$ 1.6 from AERONET is noticed in the year 2003, whereas MODIS data showed a maximum during the year 2011. The discrepancy between the two data sets may be due to the difference in spatial and temporal resolution [21]. Trend analysis of both the data sets is shown in Fig. 2, which clearly indicates the increasing trend of AOD during the period 2003–2012 having the same slope of $\sim$ 0.001.

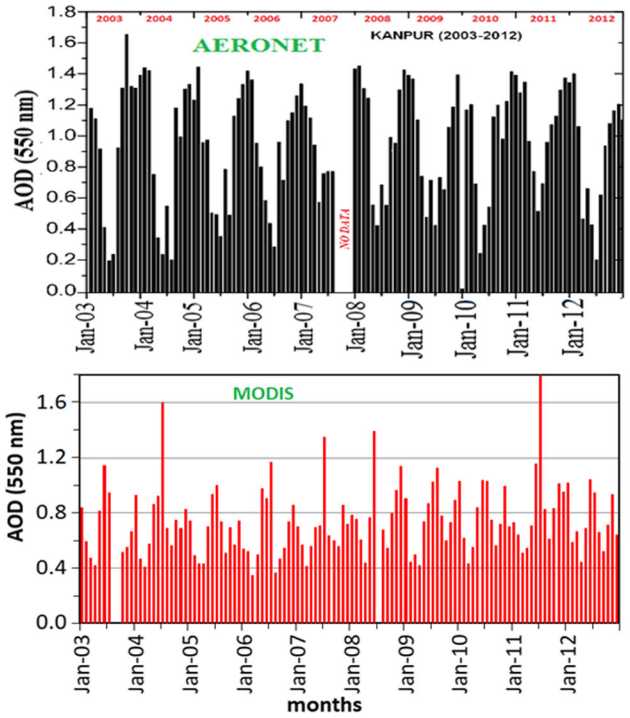

Fig. 1 Variation of aerosol optical depth at 550 nm observed from AERONET and MODIS during 2003–2012. AOD from AERONET is found maximum of ∼ 1.6 in the year 2003, but from MODIS found maximum during the year 2011

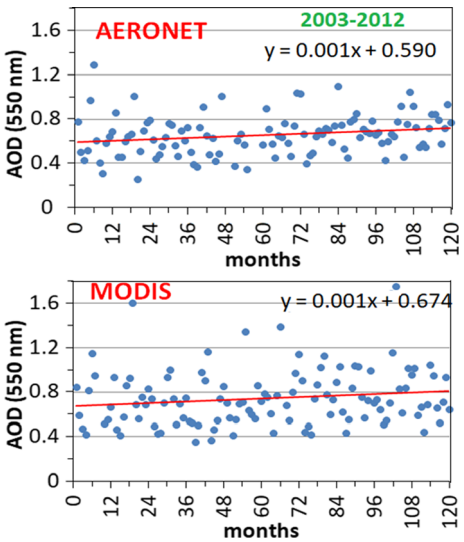

Fig. 2 Trend analysis of AOD from AERONET and MODIS at Kanpur during 2003–2012. Both the data show an increasing trend of AOD at Kanpur during 2003–2012 with the same small slope ∼0.001 and intercept of 0.59 and 0.67, respectively

Kumar et al. [21] have made trend analysis of AOD using data from AERONET, MODIS and MISR during 2005–2009 for the same station Kanpur and reported an increasing trend with little more slope 0.0018, 0.0024 and 0.003, respectively.

To study the seasonal distribution of aerosol, the year is divided into different seasons: summer (April–June), winter (November–February), monsoon (July–September) and spring (March). Value of AOD (in %) during each season is displayed by bar diagram in Fig. 3. It is clearly observed that AOD has maximum value in pre-monsoon summer season throughout 2003–2012 with exception for the years 2008 and 2011.The maximum value (59%) of AOD is observed during summer season of the year 2007. Moreover, for the years 2008 and 2011 seasonal maximum is observed during the winter season. The seasonal minimum occurred during the spring season throughout 2003–2012 except the years 2007 when the minimum is in monsoon season. Using AOD data for the period 2005–2009 from AERONET, MODIS and MISR at the Kanpur station, Kumar et al. [21] reported maximum AOD in summer season for all the three datasets.

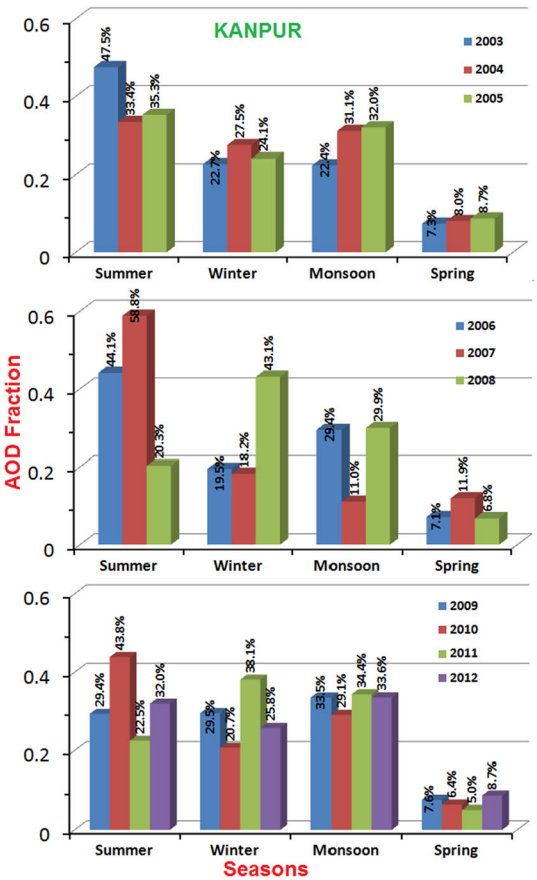

Fig. 3 Fraction of seasonal mean variation of AOD over Kanpur during 2003–2012. Seasonal mean AOD found maximum in pre-monsoon summer season throughout 2003–2012 only except the years 2008 and 2011 with a maximum for summer season (∼ 58%) of the year 2007. For 2008 and 2011, maximum AOD is noticed during the winter season

3.2. Aerosol size distribution

The aerosol size (volume) distributions (ASDs) for each month during the period 2003–2012 are shown in Fig. 4. The peak and width show the dominant size of aerosols, and the population of coarse-mode particles are generally higher (> 0.45) during 2003, 2005–2006, 2008, 2010 and 2012 and diminished value (< 0.3) during 2004, 2007, 2009 and 2011. Seasonal analysis of ASD shows peak during summer season (pre-monsoon) throughout 2003–2012 except the years 2008 and 2011 when a peak value shifted to monsoon (July) month. Major factors affecting the ASD are: aerosol strength, meteorological conditions such as surface temperature, rainfall, wind speed (transport by dust storm) and local production/industrial pollutions [8, 11]. Dust storms during the summer season directly enhance aerosol loading and harmfully affect the air quality over the IG plains [8, 15]. Dust storm-loaded aerosols usually have a lifetime of 3–4 days in the atmosphere. An interesting result of this study is the shift of ASD peak towards monsoon month which could be understood from the analysis of surface temperature and rainfall at Kanpur during 2003–2012. Figure 5(a) shows variation of surface temperature from AIRS satellite over Kanpur during 2003–2012. It is noticed that for years 2008 and 2011, temperature decreases significantly from May onwards until July. So, the local contribution to aerosol loading and hence ASD could not be significantly affected by temperature variation alone because even during the July months temperature remained almost same during the years 2008 and 2011 (Fig. 5a). The precipitation distribution at Kanpur during 2003–2012 is shown in Fig. 5(b). Large amount of precipitation rate as high as 0.4 mm/h during the pre-monsoon summer season (June) of the years 2008 and 2011 is recorded which is much higher than that of the other years. Precipitation removes aerosols from the atmosphere and in addition reduces the aerosol loading into the atmosphere from Earth's surface by enhancing its

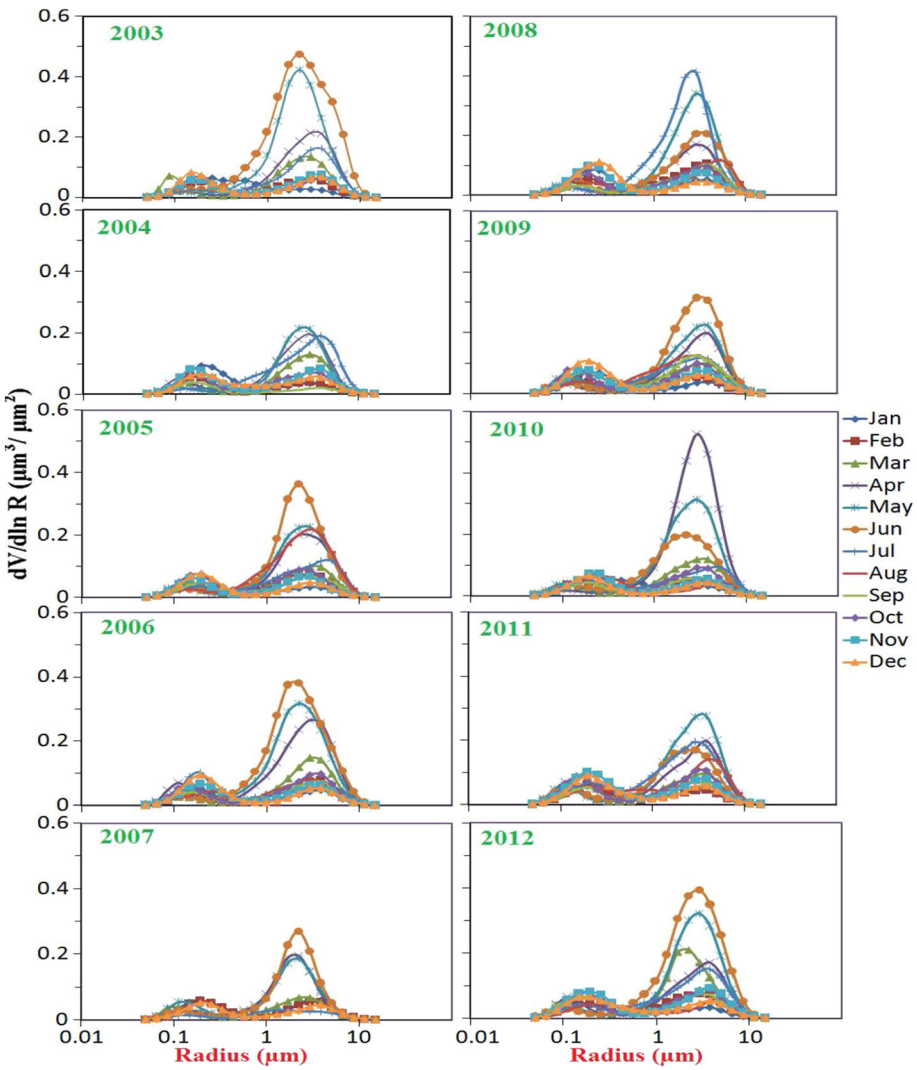

Fig. 4 Variation of peak and geometrical width of volume particle aerosol size distribution (dV(R)/dln R (mm3/mm2) against radius R of particle size at Kanpur during 2003–2012. The ASD shows peak during summer season (pre-monsoon) throughout 2003–2012 except the years 2008 and 2011

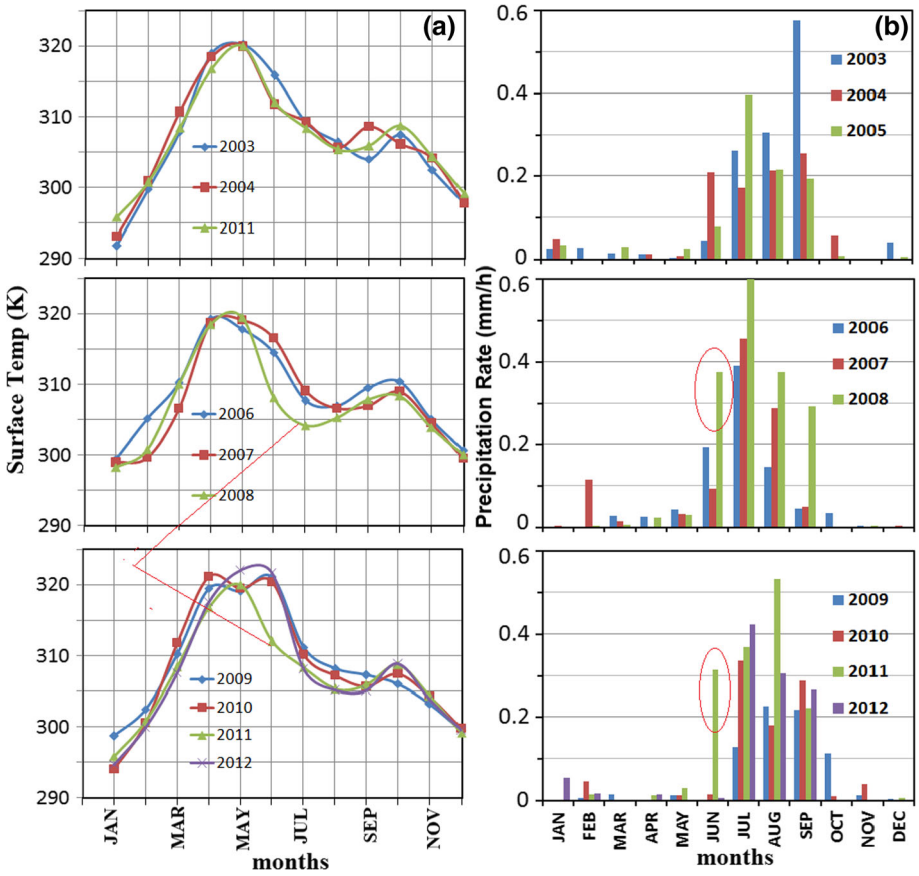

Fig. 5 (**a**) Variation of monthly mean AIR surface temperature at Kanpur during 2003–2012; (**b**) monthly mean precipitation rate (mm/h) from TRMM satellite. High precipitation rate is noticed in pre-monsoon summer month for the years 2008 and 2011

retaintivity. Thus, precipitations during summer month suppress ASD and make it largest during the monsoon (July) month. Apart from the coarse-mode aerosols, fine-mode aerosols also show volume size distribution with lower peak. The range and width of VSD for the coarse and fine modes are given in Tables 1 and 2. The peaks position shifts from season to season in a given year and also changes from year to year in a given season. This is quite evident in the case of fine mode, whereas there is lesser shift in the coarse mode. Tables 1 and 2 show peak months, peak radius, range and FWHM of coarse- and fine-mode aerosols. Usually, large-sized aerosols have peak concentration during April to June months except in the year 2008 and 2011 when peak radius occurs in the month of July. This is attributed to a change in weather condition specially rain. The largest peak radius (3.86 μm) was observed in

Table 1 Range and width (FWHM) of aerosol size distribution for coarse mode corresponding to peak for each year throughout 2003–3012

Year	Peak month	Peak radius (μm)	Range (μm)	FWHM (μm)
2003	June	2.240	0.99–6.60	5.61
2004	May	2.586	1.30–5.60	4.30
2005	June	2.240	1.70–3.85	2.15
2006	June	2.240	0.99–5.06	4.07
2007	June	2.240	1.70–3.85	2.15
2008	July	2.933	1.30–5.06	3.76
2009	June	2.933	1.07–5.06	3.99
2010	April	2.939	2.24–5.06	2.82
2011	July	3.857	1.70–6.64	4.94
2012	June	2.933	1.30–6.64	5.34

Table 2 Range and width (FWHM) of aerosol size distribution for fine mode corresponding to peak for each year throughout 2003–3012

Year	Peak month	Peak radius (μm)	Range (μm)	FWHM (μm)
2003	December	0.148	0.112–0.334	0.222
2004	January	0.194	0.112–0.439	0.327
2005	December	0.194	0.112–0.334	0.222
2006	July	0.194	0.112–0.255	0.143
2007	February	0.194	0.148–0.334	0.186
2008	December	0.255	0.112–0.439	0.327
2009	December	0.194	0.112–0.439	0.327
2010	November	0.224	0.148–0.439	0.291
2011	November	0.194	0.112–0.439	0.327
2012	November	0.194	0.112–0.334	0.222

July 2011. Fine-mode aerosols are usually present during the winter months when wind and temperature are lower. The boundary layer usually comes down to lower altitude. As a result, smokes and other pollutants from industry, vehicles and other sources are trapped and show their presence [33].

3.3. Aerosol radiative forcing

Aerosol radiative forcing arises as both direct and indirect effects. Direct effect includes absorption and scattering of solar radiation by aerosols present in the atmosphere. Aerosols acting as cloud condensation nuclei involved in the cloud microphysical processes cause a change in radiation budget of the atmosphere which is termed as indirect effect [34]. The atmospheric radiative forcing (ARF) is the difference of radiation fluxes at the top of the atmosphere (TOA) and bottom of the atmosphere (BOA). Monthly mean aerosol radiative forcing at Kanpur during 2003–2012 is shown by 3-D bar diagram in Fig. 6. ARF is found positive in all the months during 2003–2012 indicating the heating effects. The highest values are observed during the pre-monsoon season for the years 2003, 2005–2007 and 2010, whereas in the remaining years of analysis the maximum is observed during winter season. Factors which may affect the ARF are aerosol concentration, its chemical compositions and aerosol cloud interaction. During the summer season, aerosol loading is enhanced from local sources, water vapour and dust storm events over IG basin [8, 35]. Water vapour transported from Arabian sea by dust storms and its effect on radiative forcing have also been discussed in previous papers [8, 36–38]. AOD plays a major role in ARF computation which is expected to be large during summer season due to the presence of dust storm activities [8, 11, 39]. In support to this, a strong correlation at surface (R^2 = 0.85) between AOD (500 nm) and ARF has been reported during three major dust storms in pre-monsoon summer season of the year 2010 [8, 40]. They have further shown that ARF is more sensitive to the changes in SSA and refractive index controlling the absorbing ability of dust aerosols. An enhanced level of AOD is likely to affect the monsoon system. In the winter months, an enhanced anthropogenic activity along with bio-mass and crop-residual burning could contribute to the AOD loading in the IG basin [41, 42] and hence can contribute to the aerosol radiative forcing during the winter season. Artaxo et al. [43] have shown that fine-mode aerosol particles emitted from bio-mass and biogenic burning could affect the global atmosphere.

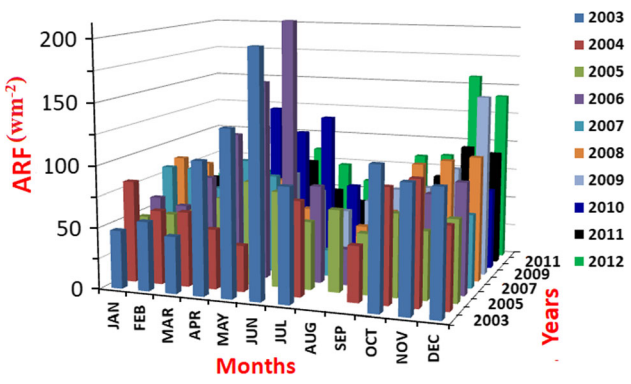

Fig. 6 Aerosol radiative forcing at Kanpur during 2003–2012. ARF found highest during the pre-monsoon season for the years 2003, 2005–2007 and 2010, but maximum during the winter season for the years 2004, 2008–2009 and 2011–2012

4. Conclusions

Aerosol data along with ASD and ARF from AERONET and satellite-based measurements over Kanpur for the years 2003–2012 are analysed, and seasonal variations of AOD are studied. The main results are summarized as follows:

1. AOD from AERONET and MODIS during 2003–2012 at Kanpur shows a constant increasing trend with a slope 0.0012. Seasonal analysis showed maximum AOD loading during the summer season throughout 2003–2012 except during the years 2008 and 2011. The highest value is ~ 59% during the summer season of 2007. For the years 2008 and 2011, AOD loading is maximum in the winter season. Seasonal minimum is noticed during the spring season throughout 2003–2012 only except the year 2007.
2. The aerosol size distribution (ASD) exhibits large variations in radius and width of distribution and shows an enhanced value during the years 2003 and 2010 and a diminished value during the years 2004, 2007, 2009 and 2011. Further, ASD shows peak during summer season (pre-monsoon) throughout 2003–2012 except in the years 2008 and 2011. Moreover, for the years 2008 and 2011 peak values appear in the monsoon (July) month. High precipitation rate as well as rainfall in June month of 2008 and 2011 may have suppressed the ASD during summer and make it largest in July (monsoon). The role of surface temperature seems to be overtaken by the precipitation.
3. Analysis of aerosol radiative forcing (ARF) at Kanpur is found largest in pre-monsoon (summer) season for the years 2003, 2005–2007 and 2010, whereas it is largest in winter months (November and December) for the years 2004, 2008–2009 and 2011–2012. During

pre-monsoon summer season of 2003 and 2006, ARF is noticed almost double from the average level of previous years. Enhanced ARF alters the atmospheric heating rate which is known to further affect the circulation/monsoon system as well.

Acknowledgements This work was supported by CSIR-New Delhi under Scientist Pool scheme (13(9049-A)/2019-POOL). The authors also thank NASA team for supporting operation of Kanpur AERONET sites. The authors would like to thank MODIS, AIRS and TRMM science data teams for processing the data via Giovanni website (https://giovanni.gsfc.nasa.gov/giovanni/). This work is supported under the collaboration program of IITM, Pune, and BHU, Varanasi, India.

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