Indian Journal of Radio & Space Physics
Vol 46, March 2017, pp 5-14

A review of potential radiative effect of aerosol on climate

Raj Kumar[a,b] & Raj Paul Guleria[a]*

[a]Department of Higher Education Himachal Pradesh, Shimla 175 001, India

[b]Career Point University, Kota 325 003, India

Received 7 July 2016; revised 27 December 2016; accepted 17 January 2017

The study of physical and chemical properties of aerosol is of significant importance, because their radiative effects exert strong impact on Earth's climate. Aerosols scatter and absorb solar radiation. Backscattering of solar radiation towards space results loss in surface reaching solar radiation leads to cooling of the climate system. Absorption of solar radiation is associated with heating within the aerosol layer, thereby modifies the vertical temperature profile, and this also results loss in surface reaching solar radiation. Such processes alter the radiative balance of Earth directly so-called direct effects. A subset of aerosols also alters the radiative balance of the Earth by modifying microphysical and radiative properties of clouds *via* so-called indirect effects. Based on observations and models studies present work suggest that the regional radiative perturbations are several Wm^{-2} due to changes in aerosol emissions. Furthermore, if the black carbon emission is checked out may lead to a sudden change in the normal pattern of warming/cooling. This paper summarized the various potential radiative mechanisms associated with aerosol-climate interaction.

Keywords: Atmospheric aerosol, Black carbon, Radiative effect, Climate

1 Introduction

The small size particles which drift between surface and stratosphere are called aerosols[1-4]. They widely distributed in the atmosphere through various atmospheric processes. The most common atmospheric processes are atmospheric transport, evaporation and convection (Fig. 1a,b). Aerosols emitted into the atmosphere or produced in the atmosphere from variety of sources[1,2,5-7]. Those emitted directly into the atmosphere are called primary aerosols and those produced in the atmosphere from precursor gases called secondary aerosols[1,8]. The most common sources of aerosols are desert dust, oceanic spray, biomass burning, and industrial emission. The biomass burning, incomplete combustion of fossil fuels, sea spray, mineral dust, volcanic eruptions etc. are the examples of primary aerosol sources[5-8]. The secondary aerosols are mixtures of compounds; the main components are sulfate and nitrate[7,8]. They are nuclei in size (< 1 μm); the cloud processing is an important atmospheric phenomenon for the production of nuclei particles[9]. On the basis of their source strength they classified as fine-mode ($r < 0.1$ μm), accumulation-mode (0.1 μm $\leq r < 1.0$ μm) and coarse-mode ($r \geq 1.0$ μm)[1].

When we think about climate change we usually talk about greenhouse gases, because they go simultaneously hand-in-hand (see Fig. 1c). But, here we are concerned about aerosols and their effects. Because, aerosol effect human health[10], alter Earth's radiation budget[11-14], alter hydrological cycle[15,16], effect weather[14], influence major monsoons of the world[15,17,18], alter atmospheric circulation[19,20], accelerate glacier melting rate[21,22]. Moreover, it is hardly to say whether aerosol cooling or warming the atmosphere[11,12,14]. The increasing burden of atmospheric aerosol particles is imposing a serious threat to the world weather in terms of human health or climate[23-26] and hence compelling world-wide scientists acquiring extensive investigations from different parts of the globe. If we give a look on IPCC climate models prediction results (see Fig. 1d). We can clearly notice that in 2012 the actual temperature was much below their predictions. This error was mostly due to poor understanding of aerosols, which are not represented accurately in global climate models. These shortcomings in understanding of aerosols are partly due to their small size and varying chemical composition, making them difficult to study[27]. Chemical composition of aerosols determines their complex refractive index. The aerosol size, structure, and refractive index are important parameters in

*Corresponding author (E-mail: rg68319@gmail.com)

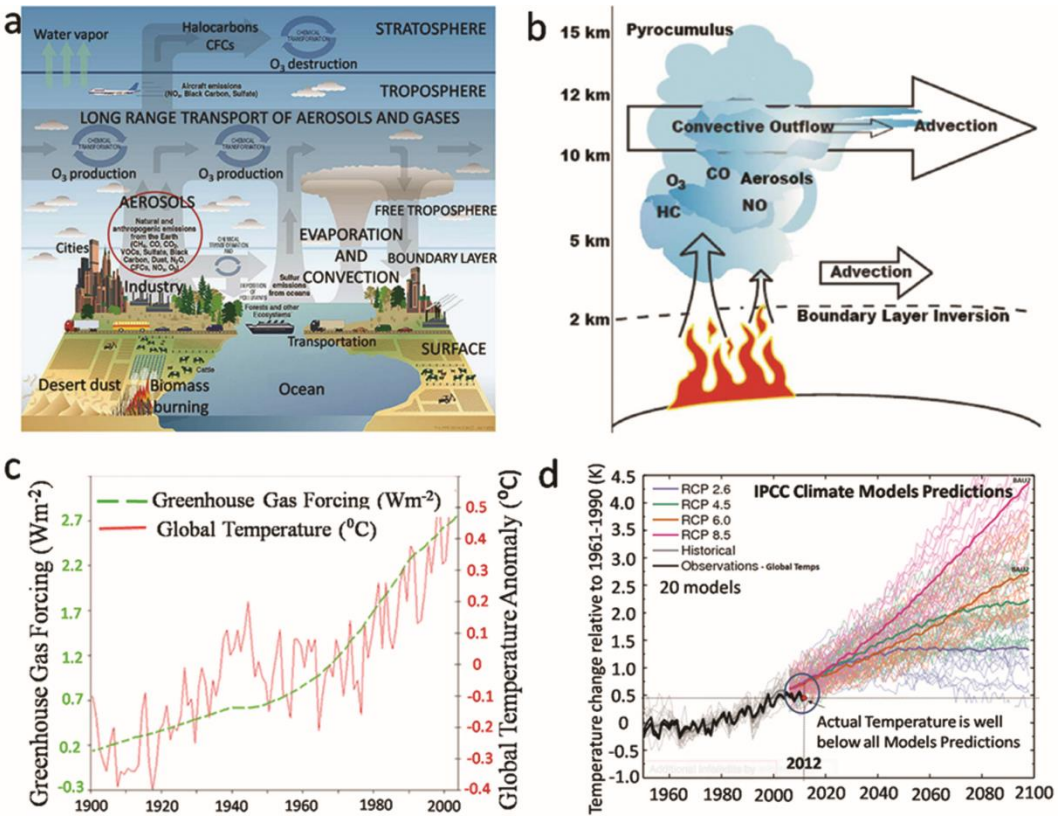

Fig. 1 — (a) Schematic representation of chemical and transport processes of aerosols and gases (source: Rozaini[27]), (b) advection of biomass burning emissions into atmosphere and their subsequent long-range transport in the upper atmosphere (source: Keywood *et al.*[71]), (c) simultaneous variation between greenhouse gas forcing and global temperature anomaly (source: NASA GISS), and (d) IPCC CIMP5 climate model failure global temperature prediction.

determining the radiative effects[11,12,28,29]. These parameters are spatially and temporally highly variable due to varied source origins[1]. In this concern, the study of atmospheric aerosol particles is of central importance, because they play important role in the atmospheric chemical and physical processes and their consequent possible effects on local and global climate[28]. Thus, the informations about the physical and chemical properties of aerosols are important particularly for precise radiative transfer calculations[30]. So, we could predict climate more accurately. It is hoped that the present study will provide an outlook into the current status of aerosols and their potential radiative effect on climate.

2 Aerosol Research Programmes: Current Status

Atmospheric aerosol is a topic of research interest since the Roman Empire times[31]. But, first systematic studies on aerosols were initiated by Aitken in 1881[32]. Over the years, a great deal of progress was made in aerosol research[33,34]. Nowadays, aerosol measurements are being conducted using ground-based sensors, satellite-based sensors and by conducting field-base campaigns. Worldwide, there are numerous aerosol networks, regional or global in scope. In 1993, NASA launched a world-wide aerosol measurement programme called AERONET (Aerosol Robotic NETwork) programme. At present, there are more than 450 AERONET network stations world-wide. The AERONET programme is a federation of ground-based remote sensing aerosol networks (https://aeronet.gsfc.nasa.gov/). Observing aerosols from space, more than three decades have been passed since the launch of the first satellite instrument. Since then, various satellite remote sensing technologies have been developed for monitoring atmospheric aerosols[35]. In 1999 and 2002, NASA launched Moderate Resolution Imaging Spectrometer (MODIS) sensors on board of Terra satellite and Aqua satellite, respectively[36]. These satellite-based sensors are continuously providing data nearly at global coverage[36]. In 2006, NASA launched the Clouds and Aerosol Lidar for Pathfinder Spaceborne Observations (CALIPSO) mission. Satellite instruments such as

Cloud-Aerosol LIdar with Orthogonal Polarization (CALIOP) on board of CALIPSO can measure the distribution of aerosols in a vertical slice of the atmosphere[37]. Indian Ocean experiment (INDOEX), Arabian Sea monsoon experiments (ARMEX), smoke, clouds, and radiation-Brazil (SCAR-B) experiment are few examples of field-based campaigns[38-40]. NASA has been a leader in providing global aerosol characterizations through observations from satellites, ground networks, and field campaigns, as well as from global and regional modeling[41]. In future, i.e., by 2023 NASA planning to execute an aerosol-cloud ecosystem mission, as part of the NASA Earth Science Tier-2 Decadal Survey missions[41]. AeroCenter is an interdisciplinary union of researchers at NASA's Goddard Space Flight Center, which was formed in 2002. AeroCenter is regularly conducting seminars, meetings to present up-to-date aerosol research, including measurement techniques; remote sensing algorithms; modeling development; field campaigns; and aerosol interactions with radiation, clouds, precipitation, climate, biosphere, atmospheric chemistry, air quality, and human health (http://aerocenter.gsfc.nasa.gov/). At regional level there are also numerous aerosol networks, for example the systematic characterization of aerosols in India was started in 1988 during Indian Middle Atmosphere Programme (IMAP)[42]. In 1991, India started another programme called Aerosol Climatology and Effects (ACE) under the umbrella of Indian Space Research Organization-Geosphere Biosphere Pragramme (ISRO-GBP). Presently is being carried out under Aerosol Radiative Forcing over India (ARFI) network programme. This is one of the largest programme carried out in the country with more than 42 ARFI network stations[42].

3 A Global View of Aerosols Distributions

Figure 2a provides an overview of the horizontal distribution of aerosols averaged over 2000-2013, using the application of MODIS -an aerosol remote sensor on board of Terra satellite. MODIS aerosol optical depth (AOD) at 550 nm confirms that the horizontal distribution of aerosols is highly inhomogeneous. The continental aerosol plumes can be seen widely distributed over oceanic regions. Satellite observations captured the Sahara dust plume -a most widely distributed aerosol plumes that can be seen to extend several thousand kilometers over the Atlantic Ocean. Satellite observations also identified five regional aerosol hotspots around the world namely: eastern China, Taklamakan region, Indo-Gangetic Plains (stretches over eastern Pakistan, northwestern part of India and Bangladesh), tropical regions of Africa, and the Arabian region (extending from Iraq to Yemen). Over these aerosol hotspots, the monthly mean (period: 2000-2013) AODs exceed 0.6. The AOD for the March to May (average period: 2000-2013) (Fig. 2b) found clearly different from that for the September to November (average period: 2000-2013) (Fig. 2c). This feature reflects seasonal dependent behaviors of aerosols. The strong seasonality can be seen over Amazone basin in South America and South Africa. The seasonal biomass burning is most prominent over these regions[43,44]. Strong seasonality can be observed over Mediterranean region and Arabian Sea. The aerosol-induced atmospheric feedback effects are most effective in these aerosol hotspots regions[45].

At global scale, natural aerosols are about three to four times larger than the anthropogenic aerosols, but at regional level the anthropogenic aerosols are significant[15,46,47]. Sulfate, black carbon (BC) and organic carbon (OC) are emitted into the atmosphere from anthropogenic as well as natural sources, whereas, the mineral dust and sea salt are emitted by wind-induced formation from natural sources. Pie charts show the contribution of sulfate, BC, OC, sea salt, Nitrate, and mineral dust aerosol in building AOD over seven regions of the globe (Fig. 3)[48]. From pie charts, we revealed that there is a large spatial and temporal variability in global aerosol composition. It is noticed that the concentrations of anthropogenic aerosols e.g., sulfate, BC and OC are higher over East North America followed by eastern China, Cango and democratic republic of the Cango, and Europe. Over eastern China, East North America and Europe the major contributor is sulfate, whereas over Cango and democratic republic of the Cango is OC. The main sources for large fraction of sulfate and BC in eastern China are coal combustion in power plants and domestic fuel combustion, respectively[49]. Sea salt concentrations are noticed higher over South Pacific Ocean and Arabian Sea (see Fig. 3). The contribution of mineral dust in total composite AOD is as high as 57% over Mauritania and Sahel regions of Africa followed by Arabian Sea. The higher concentration of mineral dust over these regions is due to transport of mineral dust from the Sahara Desert and the Thar Desert, respectively[26]. Despite substantial

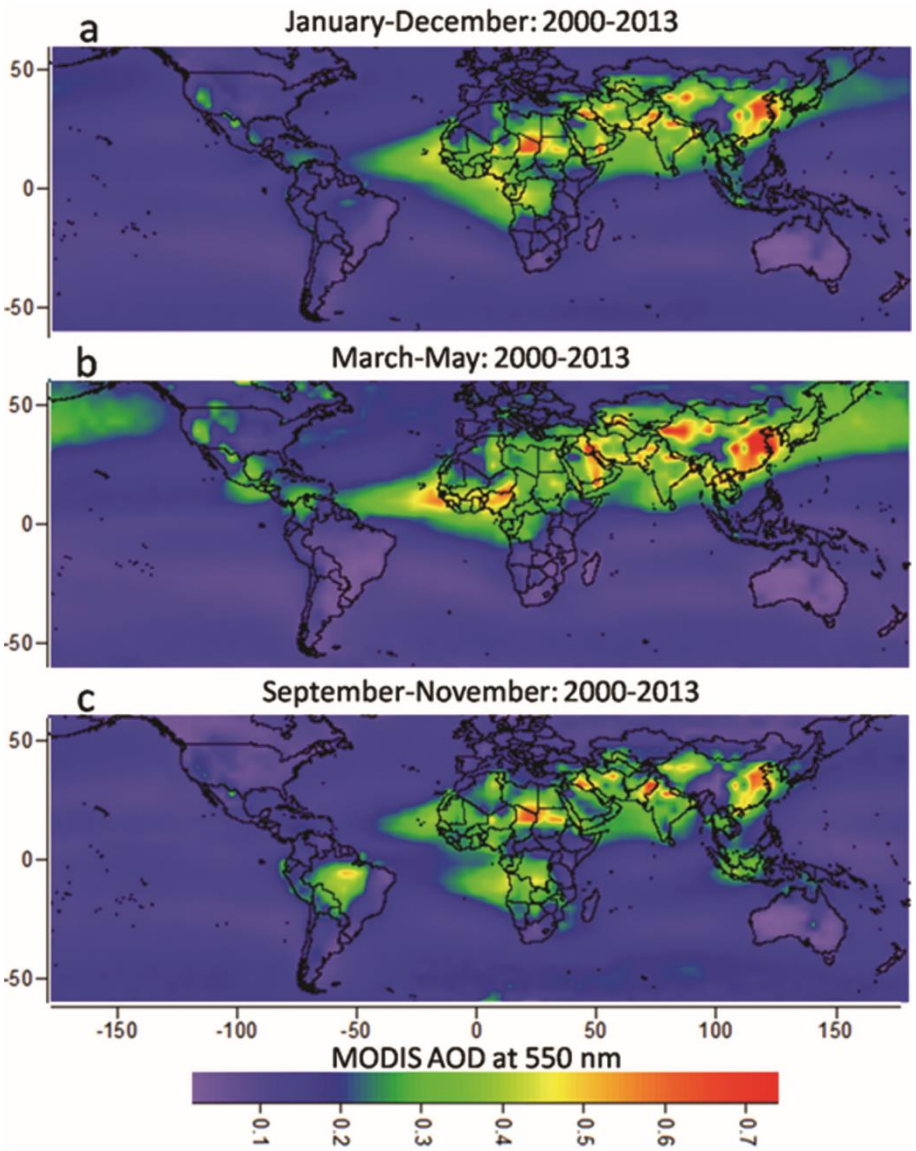

Fig. 2 — Total AOD at 550 nm derived from Terra satellite observations for January-December 2000-2013 (top panel), March-May 2000-2013 (middle panel) and September-November 2000-2013 (bottom panel), illustrating seasonal changes in aerosol emissions.

advancement of space and ground-based sensors there is a limited understanding of the greater complexity of horizontal distribution of atmospheric aerosols. This is due to large uncertainties in the chemical composition and the anthropogenic contribution to the AOD. Among aerosol types the contribution of sulfate aerosol in building AOD due to hygroscopic growth is significant. However, the contribution from OC in the existing aerosol loading is underestimated in most of the global aerosol models[50].

4 Radiative Effect of Aerosols on Climate

When solar radiation enters into the Earth's atmosphere, it interacts with aerosols and thereafter undergoes scattering and absorption process[12]. This effect of aerosol on the solar radiation is called direct radiative effect[12,51]. Aerosols scattering reflect back a considerable fraction of solar radiation back to space. This result less solar radiation reaches the earth's surface, which produces regional cooling effect. On the other hand aerosol absorption has the opposite effect, and tends to warm the climate system, for brief explanation see Fig. 4. Figures 4a and c explains the instantaneous radiative effects of aerosols, while Figures 4b and d illustrate their overall impact after the climate system has responded to their radiative effects. The direct radiative effect produces climate forcing by changing the planetary albedo[52]. The magnitude of

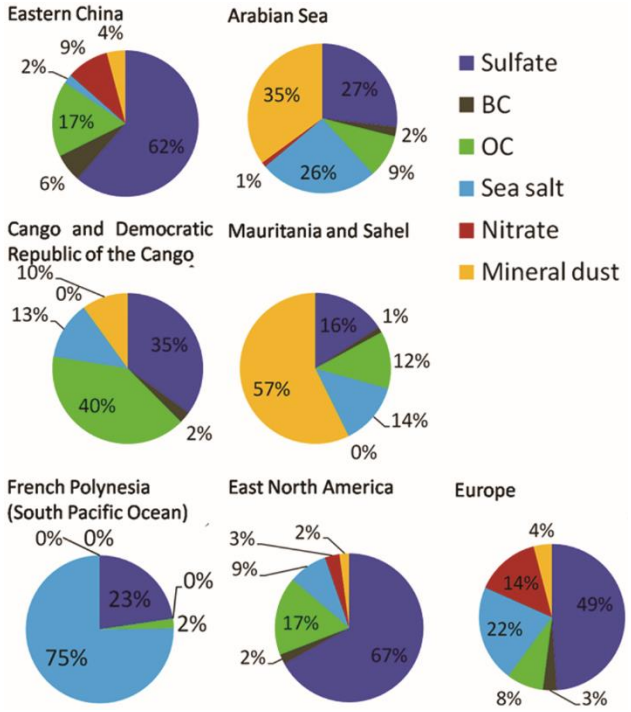

Fig. 3 — Pie charts shows the contribution of sulfate, BC, OC, sea salt, Nitrate and mineral dust aerosol in building AOD over different regions of the globe (source: Myhre *et al.*[48]).

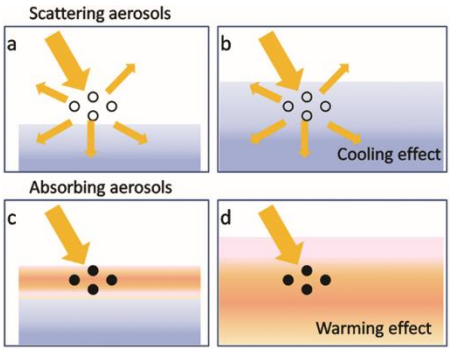

Fig. 4 — Aerosol-radiation interactions: (a,b) scattering aerosols reflecting considerable fraction of solar radiation back to space, resulting less solar radiation at the earth's surface, which leads to regional cooling, (c,d) absorbing aerosols absorbing considerable fraction of solar radiation, resulting heating of aerosol layer, but the earth's surface which receives less solar radiation, can cool regionally. At the larger scale there is a net warming of the surface and atmosphere because the atmospheric circulation and mixing processes redistribute the thermal energy (source: IPCC[25])

scattering and absorption of solar radiation depends upon physico-chemical properties of atmospheric aerosol. BC aerosol absorbs a large fraction of solar radiation and is considered as an important component to climate forcing[25]. The sulfate aerosol scatters a large fraction of solar radiation and is also an important part of climate research[12]. Aerosol that mainly scatter solar radiation produces cooling effect, whereas that absorbs solar radiation produces net warming effect. In general, in the atmosphere the mixing state of aerosol particles is dominant over scattering and absorbing aerosol particles. Therefore, net effect of mixing state of aerosol particles on Earth's energy budget is determined by surface and cloud characteristics. Scattering aerosols above a dark surface are most efficient, whereas absorbing aerosols are less efficient[53]. A dark surface already absorbs a large fraction of the solar radiation. Thus, absorbing aerosols have a small effect. On the other hand absorbing aerosols above a bright surface are most efficient particularly when positioned above the clouds, whereas scattering aerosols are less efficient. A bright surface amplifies the total reflectance of solar radiation. Thus, scattering aerosols produce small effect. Atmospheric processes like as condensation and coagulation can alter the mixing state of BC. The resulting BC-containing particles can become hygroscopic, which in turn modify clouds microphysical properties, amount and lifetime of clouds. This effect of aerosols on the solar radiation is called indirect radiative effect. A subset of aerosol particles is also act as cloud condensation nuclei (CCN) and ice nuclei (IN), on which cloud droplets and ice particles can form and here is shown in Table 1. Common CCN in the atmosphere are composed of sea salt, sulfate, nitrate, and some organics. Therefore, effectiveness of an aerosol particle to act as CCN is a key parameter for determining the indirect effect, which is a function of the size, chemical composition, mixing state and ambient environment[25]. The clean cloud contains larger cloud drops. The large concentrations of natural and anthropogenic aerosols increase cloud droplet number concentration and hence form polluted cloud. Distribution of the same cloud liquid water content over more, hence smaller, cloud droplets results to higher cloud reflectivity. This microphysical induced effect of aerosol with same low water content (LWC) is called as 'first indirect effect'[54], 'cloud albedo effect'[55], or Twomey effect[52]. The albedo effect cannot be easily separated from the other effects; in fact, the processes that decreases the cloud droplet size per given liquid water content also decrease precipitation formation, increase cloud height, and cloud lifetime. This microphysical induced effect of aerosol is called as 'second indirect effect'[54], the 'cloud lifetime effect'[55] or the 'Albrecht effect'[9] (Fig. 5). The absorbing aerosol leads to heating of the lower

Table 1 — The characteristics and role of the main aerosol species (after IPCC[25])

Aerosol species	Size distribution	Main sources	Key climate properties
Sulfate	Primary: Aitken, accumulation and coarse modes Secondary: nucleation, Aitken, and accumulation modes	Primary: marine and volcanic emissions Secondary: oxidation of SO_2 and other S gases from natural and anthropogenic sources	Light scattering. Very hygroscopic. Enhances absorption when deposited as a coating on black carbon. CCN active
Nitrate	Accumulation and coarse modes	Oxidation of NO_x	Light scattering. Hygroscopic. CCN active
Black carbon	Freshly emitted: <100 nm Aged: accumulation mode	Combustion of fossil fuels, biofuels and biomass	Large mass absorption efficiency in the shortwave. CCN active when coated. May be IN active
Organic aerosol	Primary organic aerosols : Aitken and accumulation modes. Secondary organic aerosol: nucleation, Aitken and mostly accumulation modes. Aged organic aerosols: accumulation mode	Combustion of fossil fuel, biofuel and biomass. Continental and marine ecosystems. Some anthropogenic and biogenic non-combustion sources	Light scattering. Enhances absorption when deposited as a coating on black carbon. CCN active (depending on aging time and size)
Brown carbon (Type of organic aerosols)	Freshly emitted: 100-400 nm Aged: accumulation mode	Combustion of biofuels and biomass. Natural humic-like substances from the biosphere	Medium mass absorption efficiency in the UV and visible. Light scattering
Mineral dust	Coarse and super-coarse modes, with a small accumulation mode	Wind erosion, soil resuspension. Some agricultural practices and industrial activities (cement)	IN active. Light scattering and absorption. Greenhouse effect
Sea spray	Coarse and accumulation modes	Breaking of air bubbles induced e.g., by wave breaking. Wind erosion	Light scattering. Very hygroscopic. CCN active. Can include primary organic compounds in smaller size range
marine primary organic aerosols	Preferentially Aitken and accumulation modes	Emitted with sea spray in biologically active oceanic regions	CCN active

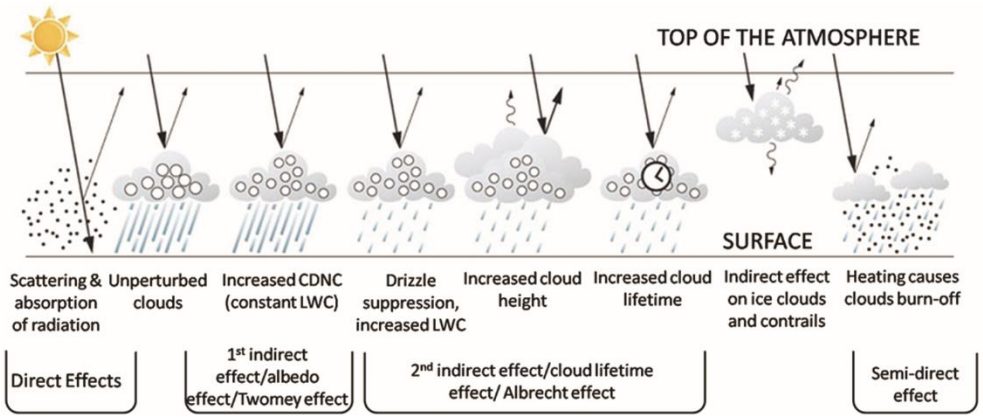

Fig. 5 — Schematic diagram showing the various radiative mechanisms associated with cloud effects that have been identified as significant in relation to aerosols. The small black dots represent aerosol particles; the larger open circles cloud droplets. Straight lines represent the incident and reflected solar radiation, and wavy lines represent terrestrial radiation. The filled white circles indicate cloud droplet number concentration (CDNC). The unperturbed cloud contains larger cloud drops as only natural aerosols are available as cloud condensation nuclei, while the perturbed cloud contains a greater number of smaller cloud drops as both natural and anthropogenic aerosols are available as cloud condensation nuclei (CCN). The vertical grey dashes represent rainfall, and LWC refers to the liquid water content (source: IPCC[14])

atmosphere that in turn changes the relative humidity and the stability of the lower atmosphere and thereby influences cloud formation and lifetime. This microphysical induced effect of aerosol is called as 'semi-direct effect'[56] (Fig. 5). However, the net effect of aerosol is uncertain and highly depends on the horizontal as well as vertical profile of absorbing aerosols such as BC. When BC is deposited on the snow surface, it darkens snow surface. This mechanism reduces the snow albedo, increases the absorption

efficiency of solar radiation, which in turn warms the surface and therefore contributes to glacier retreat[57]. The Tibetan Plateau is one of the largest sources of BC soot particles in the world[58]. The BC deposition on the surface of Tibetan glaciers has been a significant contributing factor to accelerate the melting rate of glaciers[58]. BC deposited on the surface of glaciers in China has been found to darken them by up to 5% that accelerate their melting rate. The large warming of the atmospheric layers from 2 to 6 degrees observed at several locations of India is due to mixing state of BC aerosols[53]. Recent investigations have shown that when BC exists in mixing state, its radiative impact significantly become larger[59,60]. In such a condition, the aerosol-induced warming could be much more than our expectations[53]. Recently, the aerosol-induced elevated warming observed below 2 km over northern Indian Ocean, whereas over central India it observed around 4 km. This exhibits strong meridional gradients ~4 K at atmospheric levels above 2 km, which is sufficient cause to alter the hydrological cycle[15] and hence Indian monsoon *via* "*Elevated Heat Pump Mechanism*"[19,61]. The significant concentration of mixing state of BC aerosols above the bright clouds is observed along the elevated sites of Indian Himalayas. This has enhanced the absorption efficiency of solar radiation, and plays an important role in drastic shrinking the Himalayan glaciers[21,62]. Kulkarni *et al*.[62] reported that the glacier area retreat by 21% from 1962 to 2001. The fractional contribution of BC aerosols in total composite AOD is however small (see Fig. 3), but its continuous deposition on snow or ice surfaces may accelerate the melting rate of polar ice or/ the Himalayan glaciers and in near future may raise several issues like; What are the climate implications of large warming by the elevated absorbing aerosol over most of the regions of the world particularly Himalayan regions? Yet, BC aerosol-induced reduction of snow albedo and its contribution to glacier retreat have only begun to receive attention[63-65].

5 Understanding of the Radiative Effect of Aerosols

There are numerous methods to examine how various drivers contribute to climate change. The climate response to a single factor could directly show its impact, but in general it is difficult to determine the impact of any single factor[25]. In estimation of impact of single factor we follow various metrics intermediate between cause and effect. The radiative forcing is one of the most widely used metrics and often used to quantify the potential effects of aerosol on climate. Over the period, atmospheric models have improved significantly, which provide radiative forcing estimates for a large set of aerosol components. The radiative effect of aerosol particularly of sulfate aerosol is earlier reported by Charlson *et al*.[11]. Later Haywood and Shine[66] had successfully estimated the radiative forcing from BC. To minimize the uncertainties in estimates of direct and indirect aerosol effect; a multi-model studies are performed[67]. Earlier estimates have shown that the observational based radiative forcing were larger than the model based calculations[68]. Later, the subsequently improvement in model based calculations have proved good agreement with observational based estimates[69]. This is due to the advancements in the observations system of aerosols and credit to improved ground-based and satellite-based techniques. This has contributed to constraining the current radiative forcing using aerosol observations. Direct aerosol radiative forcing is now considerably better quantified than previously and represents a major advance in understanding since the time of the Intergovernmental Panel on Climate Change (IPCC) third assessment report, when our understanding about various components of aerosol was considerably poor[14]. In IPCC 2007 report, first time direct aerosol radiative forcing due to combined effect of aerosols is reported -0.5 ± 0.4 Wm^{-2}, with a medium-low level of scientific understanding. The estimation of aerosol radiative forcing with minimum uncertainty is an example of recent progress.

The presence of BC in the atmosphere above highly reflective surfaces such as snow and ice, or clouds, may cause a significant positive radiative forcing, which is sufficient cause to induce atmospheric warming. The vertical profile is therefore important, as absorbing aerosols containing a large fraction of BC will exert a positive radiative forcing when positioned above the clouds (Fig. 6a). The considerable fraction of absorbing aerosols over northwestern India particularly along the elevated sites of Himalaya may leads to notable surface cooling and simultaneous increase in atmospheric heating of the order of 0.41-0.73 K day^{-1}. Such a strong atmospheric heating may potentially affect the atmospheric dynamics. As in most of the studies the effect of surface reflectance in atmospheric heating is ignored. Earlier estimates have shown that the BC mass concentration over India is quite large[28]. After ISRO-GBP initiatives it realized that the Indian region is characterized by elevated

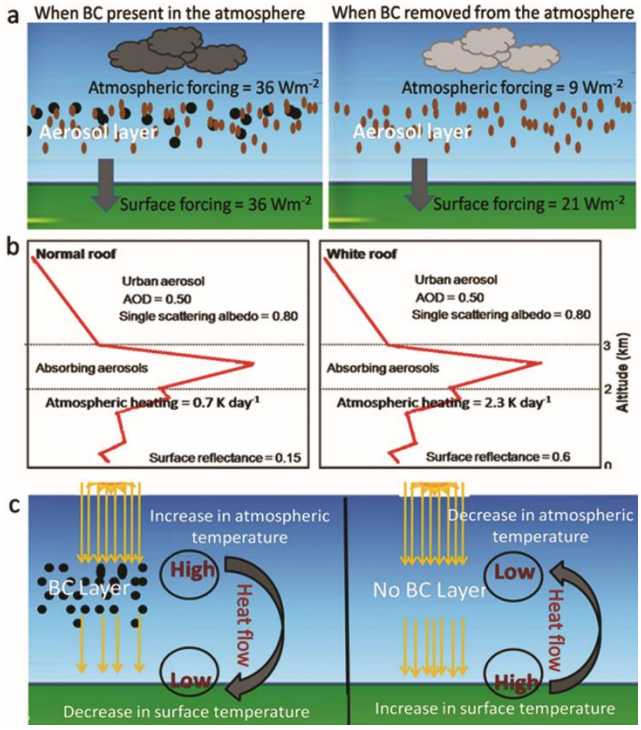

Fig. 6 — Climatic implications of removal of BC (taken/modified from Satheesh[28])

aerosol layers with significant concentration (i.e., 50-70%) around 2-3 km[28]. Satheesh[28] performed a sensitivity study taking into account the concept of whitening rooftops. The study was conducted to examine the effect of whitening rooftops with BC and without BC. Taking an example of urban aerosol, that study reveals that as a result of whitening rooftops the BC-induced atmospheric heating may increase from 0.7 to 2.3 K day^{-1} (Fig. 6b). This confirms that the consideration of white rooftops over regions which are accompanied with large BC loading, the BC-induced heating may increases to a large extent. From that sensitivity study, we conclude that if emission of BC is checked out the temperature of lower atmosphere will decrease, whereas on other hand the temperature of Earth's surface may suddenly increase, leads to sudden change in the warming/cooling patterns[70] (see Fig 6c). Therefore this study demands to investigate physical and chemical composition of aerosol components and their potential effects on climate, before attempting any BC mitigation strategies.

6 Conclusions

In summary, the understanding of potential effect of aerosol on climate is still a subject of further investigation. In estimation of radiative effect of aerosol, there is a need to conduct a comprehensive study of chemical composition of atmospheric aerosols at least from different climatic zones of the Earth. However, the initiatives have been taken by the atmospheric scientists in develop numerous radiative transfer models, but still there is a great uncertainty about the potential effect of aerosol on regional as well as global climate. Present study results report that the mitigation measure of absorbing aerosols may not be the possible solution, while addressing the climate related issues. This study also suggests that the 'semi-direct effect' of aerosols, wherein cloud cover is changed in response to aerosol heating. More extensive measurements of physical and chemical properties of atmospheric aerosol, along with the vertical distribution of the aerosol layer, are needed to reduce uncertainty of climate response to aerosol, particularly absorbing aerosol. Model based study confirms that the average aerosol radiative forcing over Indian region is significantly small in the atmosphere, in comparison to the large reduction of the net flux at the surface. Thus, absorbing aerosols redistribute radiative heating from the surface to the dust layer, unlike sulfate, which through reflection reduce the total radiative energy gained by the column. When the amount of absorbing aerosols are significant, the chemical composition of aerosol is not the only determinant of aerosol climate effects, but the altitude of the aerosol layer and types of clouds are also important. We conclude that Earth-atmosphere interactions are crucial for aerosol climate effects and can significantly influence the effects of absorbing aerosol emission controls, both on climate and air quality.

Acknowledgment

Authors are grateful to Dean (Research and Higher Studies) Career Point University Kota for their extended help. We acknowledge with thanks the different works and data sources (like MODIS, etc.) which have been, directly or indirectly, used and referred in the present work. Authors are thankful to worthy anonymous reviewers who have spared their invaluable time to go through our manuscript and to suggest very important constructive comments, which helped to improve the quality of paper.

References

1 Junge C E, *Air chemistry and radioactivity*, (Academic Press, New York), 1963.

2 Prospero J M, Ginoux P, Torres O, Nicholson S E & Gill T E, *Rev Geophys (UK)*, 40 (2002) 1002, doi:10.1029/2000RG000095.

3 Meszaros E, *Atmospheric chemistry fundamental aspects*, (Elsevier Scientific Publishing Company, New York), 1981.

4 Aloysius M, Mohan M, Parameswaran K, George S K & Nair P R, *Ann Geophys (Germany)*, 26 (2008) pp 431–440, doi:10.5194/angeo-26-431-2008.

5 Eck T F, Holben B N, Reid J S, Dubovik O, Smirnov A, O'Neill N T, Slutsker I & Kinne S, *J Geophys Res (USA)*, 104 (1999) 31333-31349, doi:10.1029/1999JD900923.

6 Schuster G L, Dubovik O & Holben B N, *J Geophys Res (USA)*, 111 (2006) D07207, doi:10.1029/2005JD006328.

7 Pósfai M & Buseck P R, *Annu Rev Earth Planet Sci (USA)*, 38 (2010) pp 17–43, doi:10.1146/annurev.earth.031208.100032.

8 Pöschl U, *Angew Chem Int Ed (Germany)*, 44 (2005) pp 7520–7540, doi:10.1002/anie.200501122.

9 Albrecht B A, *Science (USA)*, 245 (1989) pp 1227–1230, doi:10.1126/science.245.4923.1227.

10 IPCC, *Climate Change 2001, The Physical Science Basis*, (Cambridge University Press, Cambridge), 2001.

11 Charlson R J, Langner J, Rodhe H, Leovy C B & Warren S G, *Tellus Series a-Dyn Meteorol Ocean (Stockholm, Sweden)*, 43 (1991) pp 152–163, doi:http://dx.doi.org/10.3402/tellusa.v43i4.11944.

12 Charlson R J, Schwartz S E, Hales J M, Cess R D, Coakley J A, Hansen J E & Hoffman D J, *Science (USA)*, 255 (1992) pp 423–430, doi:10.1126/science.255.5043.423.

13 Andrews E, Sheridan P J, Fiebig M, *et al*., *J Geophys Res (USA)*, 111 (2006) D05S04, http://dx.doi.org/10.1029/2004JD005734.

14 IPCC, *Climate change 2007, The Physical Science Basis*, (Cambridge University Press, Cambridge), 2007.

15 Ramanathan V, Crutzen P J, Kiehl J T & Rosenfeld D, *Science (USA)*, 294 (2001) 2119-2124, doi:10.1126/science.1064034.

16 Ramanathan V, Chung C, Kim D, Bettge T, Buja L, Kiehl J T, Washington W M, Fu Q, Sikka D R & Wild M, *Proc Natl Acad Sci (USA)*, 102 (2005) 5326-5333, doi:10.1073/pnas.0500656102.

17 Menon S, Hansen J, Nazarenko L & Luo Y, *Science (USA)*, 297 (2002) 2250-2253, doi:10.1126/science.1075159.

18 Wang C, Kim D, Ekman A M L, Barth M C & Rasch P J, *Geophys Res Lett (USA)*, 36 (2009) L21704, doi:10.1029/2009GL040114.

19 Lau K M, Kim M K & Kim K M, *Clim Dyn (Germany)*, 26 (2006) 855-864, doi:10.1007/s00382-006-0114-z.

20 Ramanathan V & Feng Y, *Atmos Environ (UK)*, 43 (2009) pp 37–50, doi:10.1016/j.atmosenv.2008.09.063.

21 Lau K M, Kim M K, Kim K M & Lee W S, *Environ Res Lett (UK)*, 5 (2010) 025204, doi:10.1088/1748-9326/5/2/025204

22 Xu B Q, Wang M, Joswiak D R, Cao J J, YaoT D, Wu G J, Yang W & Zhao H B, *J Geophys Res (USA)*, 114 (2009) D17209, doi:10.1029/2008JD011510.

23 Dockery D W, Pope C A, Xu X P, Spengler J D, Ware J H, Fay M E, Ferris B G & Speizer F E, *N Engl J Med (USA)*, 329 (1993) 1753-1759, doi:10.1056/NEJM199312093292401.

24 Hadjimitsis D G, *Air Qual Atmos Health (Netherlands)*, 2 (2009) 89-97, doi:10.1007/s11869-009-0036-0.

25 IPCC, *Climate Change 2013, The Physical Science Basis*, (Cambridge University Press, Cambridge), 2013.

26 Vinoj V, Rasch P J, Wang H, Yoon J H, Ma P L, Landu K & Singh B, *Nature Geoscience (UK)*, 7 (2014) pp 308–313.

27 Rozaini M Z H, *The chemistry of dicarboxylic acids in the atmospheric aerosols, atmospheric aerosols-regional characteristics-chemistry and physics*, edited by Hayder A R, In Tech (2012), doi:10.5772/50127.

28 Satheesh S K, *Curr Sci (India)*, 102 (2012) 426-439.

29 Hansen J E, Sato M & Ruedy R, *J Geophys Res (USA)*, 102 (1997) D6 6831-6864, doi:10.1029/96JD03436.

30 Yu H, *et al*., *Atmos Chem Phys (Germany)*, 6 (2006) 613-666, doi:10.5194/acp-6-613-2006.

31 Nriagu J O, *Science (USA)*, 272 (1996) 223-224.

32 Kajima H, *J Meteorol Soc (Japan)*, 56 (1978) 602-607.

33 Holben B N, *Remote Sen Environ (USA)*, 66 (1998) 1-16.

34 Holben B N, Eck T F, Slutsker I, Smirnov A, Sinyuk A, Schafer J, Giles D & Dubovik O, AERONET's version 2.0 quality assurance criteria. In Asia-Pacific Remote Sensing Symposium, *Int Soc Opt Photonics (USA)*, (2006) 64080Q-64080Q-14.

35 Lee K H, Li Z, Kim Y J & Kokhanovsky A, Atmospheric aerosol monitoring from satellite observations: a history of three decades, in *Atmospheric and Biological Environmental Monitoring (Berlin)*, edited by Kim Y J *et al*., (2009) 13-38, doi:10.1007/978-1-4020-9674-7 2.

36 Remer L A, Kaufman Y J, Tanre D, *et al*., *J Atmos Sci (USA)*, 62 (2005) 947-973, doi:10.1175/JAS3385.1.

37 Winker D M, Vaughan M A, Omar A, *et al*., *J Atmos Ocean Technol (USA)*, 26 (2009) 2310-2323.

38 Mitra A P, *Indian J Marine Scie (India)*, 33 (2004) 30-39.

39 Kaufman Y J, Hobbs P V, Kirchhoff V W J H, *et al*., *J Geophys Res (USA)*, 103 (1998) D24 31783-31808, doi:10.1029/98JD02281.

40 Rao P S, *Mausam (India)*, 56 (2005) 1-6.

41 Matsui T, Ichoku C, Randles C, *et al*., *Bull Am Meteorol Soc (USA)*, ES 203 (2014), doi:10.1175/BAMS-D-13-00153.1.

42 Moorthy K K, Niranjan K, Narasimhamurthy B, Agashe V V & Murthy B V K, *Aerosol climatology over India. ISRO-GBP MWR network and data base*, Scientific Report (India), SR-03–99 (1999) Indian Space Research Organisation (ISRO), Bangalore.

43 Bevan S L, North P R J, Grey W M F, Los S O & Plummer S E, *J Geophys Res (USA)*, 114 (2009) D09204, doi:10.1029/2008JD011112.

44 Levinson D H & Lawrimore J H, in *State of the Climate in 2007*, edited by Arguez A, Diamond H J, Fetterer F, Horvitz A & Levy J M, (WMO Bulletin, Switzerland), 2009 .

45 Lau W K M, Kim K M, Hsu C N & Holben B N, *WMO Bull (Switzerland)*, 58 (2009) 22-30.

46 Andreae M O, Jones C D & Cox P M, *Nature (UK)*, 435 (2005) 1187-1190, doi:10.1038/nature03671.

47 Seinfeld J H & Pandis S N, *Atmospheric chemistry and physics: from air pollution to climate change*, (Wiley, New York), 2006.

48 Myhre G, Myhre C E L, Samset B H & Storelvmo T, *Nature Edu Knowledge (UK)*, 4 (2013) 7.

49 Lu Z, Streets D G, Zhang Q, *et al*., *Atmos Chem Phys (Germany)*, 10 (2010) 6311-6331, doi:10.5194/acp-10-6311-2010.

50 Zhang Q, et al., *Geophys Res Lett (USA)*, 34 (2007) L13801, doi:10.1029/2007GL029979.

51 Satheesh S K & Ramanathan V, *Nature (UK)*, 405 (2000) 60-63, doi:10.1038/35011039.

52 Twomey S A, *J Atmos Sci (USA)*, 34 (1977) 1149-1152, doi:http://dx.doi.org/10.1175/1520-0469(1977)034<1149:TIOPOT>2.0.CO;2.

53 Satheesh S K, Moorthy K K, Babu S S, Vinoj V & Dutt C B S, *Geophys Res Lett (USA)*, 35 (2008) L19809, doi:10.1029/2008GL034944.

54 Ramaswamy V, Boucher O, Haigh J, *et al*., *Radiative forcing of climate change, in Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change* [Houghton J T, *et al*. (Ed)], (Cambridge University Press, Cambridge), 2001.

55 Lohmann U & Feichter J, *Atmos Chem Phys (Germany)*, 5 (2005) 715-737, doi:10.5194/acp-5-715-2005.

56 Ackerman A S, Toon O B, Taylor J P, Johnson D W, Hobbs P V & Ferek R J, *J Atmos Sci (USA)*, 57 (2000) 2684-2695, doi: http://dx.doi.org/10.1175/1520- 0469 (2000)057 <2684: EOAOCA> 2.0.CO;2.

57 Hansen J & Nazarenko L, *Proc Natl Acad Sci (USA)*, 101 (2004) 423-428, doi:10.1073/pnas. 2237157100.

58 Ohara T, Akimoto H, Kurokawa J, Horii N, Yamaji K, Yan X & Hayasaka T, *Atmos Chem Phys (Germany)*, 7 (2007) 4419-4444, doi:10.5194/acp-7-4419-2007.

59 Chandra S , Satheesh S K & Srinivasan J, *Geophys Res Lett (USA)*, 31 (2004) L19109, doi:10.1029/ 2004GL020662.

60 Vinoj V, Babu S S, Satheesh S K, Moorthy K K & Kaufman Y J, *J Geophys Res (USA)*, 109 (2004) D5, doi:10.1029/2003JD004329.

61 Lau K M & Kim K M, *Geophys Res Lett (USA)*, 33 (2006) L21810, doi:10.1029/2006GL027546.

62 Kulkarni A V, Bahuguna I M, Rathore B P, *et al*., *Curr Sci (India)*, 92 (2007) 69-74.

63 Gautam R, Hsu N C, Lau K M, Tsay S C & Kafatos M, *Geophys Res Lett (USA)*, 36 (2009) L07704, doi:10.1029/ 2009GL037641.

64 Gautam R, Hsu N C, Lau K M & Kafatos M, *Ann Geophys (Germany)*, 27 (2009) 3691-3703.

65 Gautam R, Hsu N C & Lau K M, , *J Geophys Res (USA)*, 115 (2010) D17208, doi:10.1029/2010JD013819.

66 Haywood J M & Shine K P, *Geophys Res Lett (USA)*, 22 (1995) 603-606, doi:10.1029/95GL00075.

67 Schulz M, Textor C, Kinne S, *et al*., *Atmos Chem Phys (Germany)*, 6 (2006) 5225-5246, doi:10.5194/acp-6-5225-2006.

68 Forster P, Forster, P, Ramaswamy V, Artaxo P, *et al*., *Changes in atmospheric constituents and in radiative forcing, Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, (Cambridge University Press, Cambridge), 2007.

69 Myhre G, *Science (USA)*, 325 (2009) 187-190, doi:10.1126/science.1174461.

70 Novakov T, Kirchstetter T W, Menon S & Aguiar J, *Geophys Res Lett (USA)*, 35 (2008) 1-4, doi:10.1029/2008GL034894.

71 Keywood M, Kanakidou M & Stohl A, *Crit Rev Environ Sci Tech (USA)*, 43 (2013) 40-83.