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Key Messages

- Aerosol loading over India has substantially increased during the recent few decades. The annual mean 500 nm aerosol optical depth (AOD) from ground-based observations shows an overall increasing trend of $\sim 2\%$ year⁻¹ during the last 30 years (high confidence). This trend in AOD is subject to seasonal variability. The rate of increase in AOD is significantly high during the dry months of December–March.
- The aerosol radiative forcing over India shows wide spatiotemporal variability resulting from the non-uniform distribution of aerosol burden over the region. Estimates of aerosol radiative forcing from measurements range from -49 to -31 W m⁻² at the surface (high confidence) and from -15 to $+8$ W m⁻² at the top-of-atmosphere (low confidence). The estimates at the top of the atmosphere are highly sensitive to the single scattering albedo values.
- The understanding of the aerosol indirect effect and aerosol impacts on precipitation has low confidence and needs to be addressed with process studies in different cloud systems and their environments.
- There is substantial spatiotemporal variability in the concentration of ozone (O₃) and its precursors over the Indian region. In general, there is an increasing trend in the ozone mixing ratios in the troposphere ($+0.7$ to $+0.9\%$ year⁻¹ during 1979–2005, medium confidence) and a decreasing trend in the stratosphere (-0.05 to -0.4% year⁻¹ during 1993–2015, medium confidence). Trends are driven by precursor gases emitted by anthropogenic activities.
- Over the Indian region, the estimates of radiative forcing (at the tropopause) due to tropospheric ozone increase since pre-industrial times vary between ~ 0.2 and 0.4 W m⁻².

5.1 Introduction

Aerosols and trace gases are essential drivers of climate change. They influence Earth's energy budget leading to climate change through various pathways. Their climatic impacts are eventually manifested as precipitation changes, increased evaporation, elevated temperatures, etc. Hence, information on their 'sources and sink,' 'physical and chemical processes,' and 'distribution' is important for an accurate prediction of the climate.

The Indian subcontinent is directly influenced by different aerosol species via changes in the insolation, atmospheric temperature structure, and alteration of the regional hydrological cycle. Along with absorption and scattering of

incoming solar radiation, aerosols interact with clouds modifying its radiative properties and precipitation efficiency (Box 5.1). The aerosol concentrations over the subcontinent are dominated by wind-driven desert dust, biomass burning, industrialization, agricultural activities, etc. Rapid growth in population, industrialization, and urbanization—over South, East, and Southeast Asia—has contributed to the significant rise in emissions producing different types of aerosol over the region. The associated increase in anthropogenic aerosol loading in recent decades (Satheesh et al. 2017) has led to increased reduction of surface insolation, contributing to solar dimming over the Indian landmass, affecting the energy balance at the surface (Ramanathan et al. 2005; Soni et al. 2012). The high aerosol burden has also been linked to changes in the hydrological cycle of the region (Box 5.3). The long-term decline in southwest monsoon precipitation has been associated with anthropogenic aerosol forcing over South Asia [(Krishnan et al. 2016), Box 5.3].

Box 5.1: How Aerosols Affect Regional Climate?

Atmospheric aerosols are tiny solid/liquid/mixed particles suspended in the air originating from natural or anthropogenic sources. With a typical lifetime of days to weeks in the troposphere and about a year in the stratosphere, aerosol size ranges from a few nanometers to several tens of micrometers. Aerosol particles influence the climate in different ways (Box 5.1, Fig. 5.1).

Fundamentally, aerosol particles absorb and scatter incoming solar radiation modifying the global and regional radiative budget. Non-absorbing aerosols like sulfate, nitrate, and sea spray scatter shortwave radiation back to space leading to a net cooling of the climate system while absorbing aerosols produce the opposite effect. Carbonaceous aerosols (black carbon, organic carbon) and mineral dust can absorb and scatter sunlight producing either warming or cooling effects determined by aerosol properties and environmental conditions. Absorbing aerosols also affect climate when present in surface snow by lowering surface albedo, yielding a positive radiative forcing, directly changing the melting of snow and ice. Although depending on the local emissions and transport processes, regionally, the anthropogenic aerosol radiative forcing can be either negative or positive; it is well established that globally, the radiative effect of anthropogenic aerosols produces cooling of the planet (IPCC 2013).

Additionally, aerosol particles act as cloud condensation nuclei (CCN) and ice nuclei (IN) and therefore have a significant impact on cloud properties

Fig. 5.1 A schematic showing the different sources of aerosols in the atmosphere and their effect on the radiative budget. Adapted from the figure provided by Brookhaven National Laboratory

and precipitation. An increase in CCN forming aerosols in a cloudy region produces more, but smaller, cloud droplets reflecting more solar radiation to space leading to a cooling of the Earth's surface, known as the first indirect effect (cloud-albedo effect). Also, smaller droplets suppress collision coalescence requiring longer growth time to reach raindrop size, increasing the cloud albedo and enhancing the cooling effect, known as the cloud-lifetime effect or the second indirect effect. In contrast, in a saturated and buoyant environment, an increase in CCN forming aerosols can invigorate the cloud through microphysical processes; however, there is significant uncertainty in such impacts.

Furthermore, absorbing aerosols alter the air temperature causing an increase in lower level static stability inhibiting convection leading to a decrease in cloud cover, known as the semi-direct effect. However, the net effect of absorbing aerosols on precipitation depends on the vertical variation of the particles and background conditions.

Ozone, photo-chemically active trace gas, plays a crucial role in the climate system due to its implications on radiative processes and resulting dynamical changes. The majority of (ninety percent) the ozone in the atmosphere occurs in the stratosphere, where it is formed naturally by chemical reactions involving solar ultraviolet radiation (sunlight) and oxygen molecules (WMO 2019a). Remaining 10% of ozone

occurring in the troposphere is mainly produced from precursor gases (e.g., methane (CH₄), nitrogen oxides (NO_x), volatile organic compounds (VOCs), carbon monoxide (CO), methane (CH₄)). The industrialization, vehicular emission, and other anthropogenic activities have accelerated the emission growth of ozone precursors leading to a continued rise in tropospheric ozone concentrations (Sinha et al. 2014). On the contrary, addition of these gases and ozone-depleting substances (ODSs) produces significant stratospheric ozone loss leading to chemical and dynamical changes in the troposphere and stratosphere. The measurements over India during the past two–three decades show rapid changes in ozone (O₃) mixing ratios in the troposphere and stratosphere. Considering the critical role of atmospheric ozone in the climate system, in this chapter, we document an assessment of emissions of ozone precursors, trends in ozone and related gases, the influence of transport processes on their distribution over the Indian region.

It is difficult to detangle the cause and effect of climate change. The temperature changes have significantly been affected by the atmospheric burden of aerosols and trace gases. The transport processes produce a significant impact on the redistribution of aerosols and trace gases (e.g., dust transport from West Asia in pre-monsoon season; monsoon convection-based lifting of aerosols and trace gases into the lower stratosphere, tropopause folding events in the winter/pre-monsoon season). The changes in their concentrations at the receptor region affect the temperature, radiative forcing, clouds, and aerosol-cloud interactions.

Box 5.2: What are Trace Gases?

The Earth's atmosphere consists of large amounts of nitrogen (78% by volume) and oxygen (21% by volume). The remaining 1% of the atmospheric gases are known as trace gases because they are present in small concentrations, mostly one part per billion (ppb) or lower. The sources of trace gases can be natural or anthropogenic. The natural sources are biogenic, volcanoes, lightning and forest fires, and emission from the Oceans. The global ocean is a source of several trace gases, including sulfur-containing gases. The trace gases are also formed in the atmosphere through chemical reactions in the gas phase. The anthropogenic sources of trace gases are fossil fuel combustion, fossil fuel mining, biomass burning, and industrial activity, etc. (Brasseur et al. 1999).

The most important trace gases found in the atmosphere are greenhouse gases. These trace gases are called greenhouse gases because they help to keep Earth warm by absorbing sunlight. In the troposphere, water vapor, ozone (O_3), carbon dioxide (CO_2), methane (CH_4), sulfur dioxide (SO_2), and nitrous oxide (N_2O) are the important greenhouse (trace) gases. The two most abundant greenhouse gases by volume are water vapor and CO_2 (Brasseur et al. 1999).

Ozone acts as a greenhouse gas in the troposphere, while in the stratosphere, filter out the incoming ultraviolet radiation coming from the Sun. Thus, it helps in protecting life on the Earth. The human-made processes have injected new trace gases into the atmosphere, for example, chlorofluorocarbons (CFCs), which damage the ozone layer in the stratosphere.

The increased burden of trace gases in the atmosphere leads to global warming and climate change. The focus of the current chapter is ozone and related trace gases, while other trace gases, e.g., CO_2 , CH_4 , and, N_2O , etc., are discussed in Chap. 4.

salt, dust, and primary biological aerosols are naturally produced in the atmosphere. Over the Indian subcontinent, the aerosol emission rates are 8.9 Tg year^{-1} for NMVOCs, 0.7 Tg year^{-1} for BC, 1.9 Tg year^{-1} for primary organic aerosol, 2.9 Tg year^{-1} for SO_2 , 5.8 Tg year^{-1} for NH_3 , and, 0.5 Tg year^{-1} for biomass burning aerosols (IPCC 2013).

The major source of BC emissions in India is the combustion of solid biofuel ($172\text{--}340 \text{ Gg year}^{-1}$), while other sources include wood fuel (143 Gg year^{-1}), dried cattle manure (8 Gg year^{-1}), and crop waste (21 Gg year^{-1}). The relative contributions of fossil fuel, open burning, and biofuel consumption to total BC emissions over the Indian subcontinent are 25%, 35%, and 42%, respectively. Biofuel consumption also results in the emission of OC ($583\text{--}1683 \text{ Gg year}^{-1}$). The relative contributions from fossil fuel, open burning, and biofuel consumption to the total OC emissions are estimated to be 13%, 43%, and 44%, respectively (Venkataraman et al. 2005).

The transport sector, the second-largest contributor to organic aerosols over India, shows an emission rate of 0.14 ($0.1\text{--}0.3$) Tg year^{-1} for BC and 0.07 ($0.02\text{--}0.2$) Tg year^{-1} for OC. For both the emissions, diesel vehicles were found to be the primary cause (92% to BC and 78% to OC). However, the combined emission rate of BC and OC from both the industry and transport sectors is $0.23 \text{ Tg year}^{-1}$ and $0.15 \text{ Tg year}^{-1}$, respectively (Sadavarte and Venkataraman 2014). In the Indian rural sector, the emission estimate of organic and elemental carbon from biomass fuels over the IGP is $361.96 \pm 170.18 \text{ Gg}$ and $56.44 \pm 29.06 \text{ Gg}$, respectively (Saud et al. 2012). The estimates of aerosols emissions from open burning (forest and crop waste) are $102\text{--}409 \text{ Gg year}^{-1}$ for BC, $399\text{--}1529 \text{ Gg year}^{-1}$ for OC, and $663\text{--}2303 \text{ Gg year}^{-1}$ for organic matter. This overall contributes to about 25% of the total BC, OC/OM emissions (Venkataraman et al. 2006).

Residential sector SO_2 emissions have been estimated at 0.2 ($0.08\text{--}0.4$) Tg year^{-1} with major contribution from dung-based (56%) and coal-based stoves (19%). Emissions of SO_2 from agriculture were estimated at 0.09 ($0.02\text{--}0.2$) Tg year^{-1} . From the industry sector and transport sector, emission estimates of SO_2 were reported to be 7 ($6.0\text{--}9.6$) Tg year^{-1} and 0.08 ($0.04\text{--}0.3$) Tg year^{-1} , respectively, for the year 2015 (Pandey et al. 2014).

From 1996 to 2015, there has been a 30% increase in BC emissions due to increased emissions from informal industries, while OC emissions increased by only 4% (Pandey et al. 2014). Vehicular emissions of BC have increased by 112% during 1991–2001. From all the sources, the estimated BC emission for India is around 835.50 Gg for 1991 and 1343.78 Gg in 2001, indicating a growth of about 61% during the 1990s. During the same period, trends in SO_2 emissions increased by 32% (Pandey et al. 2014).

5.2 Aerosols

5.2.1 Emissions of Different Aerosol Species

Atmospheric aerosols originate from two distinct pathways—either by direct emission of primary aerosols into the atmosphere (e.g., dust, sea salt, OC, BC) or by the formation of secondary aerosols via atmospheric chemical reactions (e.g., sulfate, nitrate, ammonium, and SOA). While BC, along with sulfate, nitrate, and ammonium, has anthropogenic sources like incomplete combustion of biomass and fossil fuel, sea

5.2.2 Long-Term Change in Aerosol Optical Depth

Long-term changes in aerosol loading over the Indian subcontinent have been estimated using ground-based single station measurements (Dani et al. 2012; Kaskaoutis et al. 2012) as well as multi-station long-term (>25 years for some stations) ground-based observational network database (Babu et al. 2013; Krishna Moorthy et al. 2013). Observations from ARFINET observatories spread over 35 locations across India reported an increase in annual mean AOD ($0.008\text{--}0.02\text{ year}^{-1}$) with a rate of $2.3\%\text{ year}^{-1}$ (of its value in 1985), while AOD trend in the last decade (2001–2011) alone showed a rapid increase of $\sim 4\%\text{ year}^{-1}$ (Fig. 5.2) (Babu et al. 2013; Krishna Moorthy et al. 2013). However, the estimates of AOD trends vary from one location to another, with rural locations showing weak negative trends while industrial stations over peninsular India show high positive trends (Babu et al. 2013). AOD increasing trend of 2% per year is reported over Pune during the period 1998–2007 (Dani et al. 2012). Recent AERONET data over Pune from 2008 to 2017 also shows a positive trend in the range of 2.4–4% per year. The rate of increase of aerosol loading over the country is considerably high ($0.0005\text{--}0.04\text{ year}^{-1}$) during the dry winter months (December–March), while due to the contending effects of dust transportation and wet scavenging of aerosols by the monsoon precipitation, trends are weak or insignificant during the pre-monsoon and summer monsoon seasons (Babu et al. 2013). Furthermore, the long-term change in Angstrom wavelength exponent over India shows an increasing trend implying a relative buildup

of fine anthropogenic aerosols compared to coarser natural aerosols over the region (Dani et al. 2012; Satheesh et al. 2017). In contrast, recent ground observations have revealed a decreasing trend in BC concentrations over various locations in India (Ravi Kiran et al. 2018; Manoj et al. 2019; Sarkar et al. 2019). ARFINET observations (2007–2016) recorded a decreasing trend in BC mass concentrations over India, at a rate of $\sim 242 \pm 53\text{ ng m}^{-3}\text{ year}^{-1}$ (Manoj et al. 2019). The negative trend in BC concentrations in the backdrop of

rising BC emission trends (Sahu et al. 2008; Pandey et al. 2014) brings forth the uncertainty in BC estimates due to scattered observations over the source region (IGP) (Rana et al. 2019).

The trends in AOD over the Indian subcontinent have also been reported from satellite observations. The annual mean trend of AOD from MODIS observations for the period 2000–2014 shows an increase of $\sim 40\%$ over the Indian landmass (Srivastava 2017). MODIS regional trends show that the annual mean AODs have increased by $>40\%$ in major urban cities like Jaipur, Hyderabad, and Bengaluru during 2000–2009, while it has decreased ($\sim 10\%$) over high-altitude sites of Dehradun and Shimla (Ramachandran et al. 2012). Strong seasonal variability in AOD trends is also observed from satellite observations. During winter (DJF), a significantly increasing trend ($1\text{--}2\%\text{ year}^{-1}$, $0.02\%\text{ year}^{-1}$) is observed over the subcontinent, especially IGP (Srivastava 2017). SeaWiFS (Sea-Viewing Wide Field-of-View Sensor) AOD shows an increasing trend of $0.0053 \pm 0.0011\text{ year}^{-1}$ during DJF over 1998–2010 (Hsu et al. 2012). The post-monsoon/winter decadal trend from

Fig. 5.2 **a** Long-term 500 nm AOD trends derived from ARFINET station data. Different colors and symbols differentiate the stations and **b** long-term trend in regional mean aerosol optical depth at 500 nm. Adopted from Krishna Moorthy et al. (2013). © American Geophysical Union. Used with permission

MISR (Multi-angle Imaging SpectroRadiometer) for the period (March 2000–February 2010) also shows an increasing trend in the range $0.01\text{--}0.04\text{ year}^{-1}$ over urban centers and densely populated rural areas (Dey and Di Girolamo 2011). On the contrary, a significant and widespread decreasing trend in AOD is observed in the pre-monsoon (MAM) period over certain parts of northern India from satellite measurements (MODIS, MISR, and OMI) (Pandey et al. 2017). However, the decrease in AOD is more prominent over northwest India (-0.01 to -0.02 year^{-1}), while a clear increasing trend ($0.01\text{--}0.02\text{ year}^{-1}$) is observed in the eastern part of the IGP. During the monsoon season, AOD trends do not exhibit any statistically significant signal but show slight (significant at 90%) positive values $+0.003$ to $+0.0017$ during the post-monsoon season (SON) over the IGP. During this period, strong positive trends (2–8%) are observed over northeast India. Over the oceanic regions, significant positive trends ($2\text{--}6\%\text{ year}^{-1}$) have been reported during DJF and MAM. A positive trend in AOD ($>6\%\text{ year}^{-1}$) was also observed during JJAS over the Northern Bay of Bengal.

Box 5.3: How Aerosols Impact Monsoon Precipitation?

The non-uniform distribution of aerosols in the atmosphere creates uneven atmospheric heating and surface cooling patterns, which drive changes in atmospheric circulation and regional rainfall. On longer timescales, the declining trend in ISM precipitation post-1950s has been linked to the rising anthropogenic aerosol burden over various regions

(Ramanathan et al. 2005; Bollasina et al. 2011; Ganguly et al. 2012; Sanap and Pandithurai 2015; Sanap et al. 2015; Krishnan et al. 2016; Undorf et al. 2018). Local and remote aerosols alter the land–sea temperature contrast as well as the tropospheric temperature structure, both of which have a profound influence on the onset and sustenance of south Asian monsoon. An associated weakening of the monsoon overturning circulation (Fig. 5.3) due to anthropogenic aerosol-induced surface radiative changes results in suppression of ISM rainfall.

On shorter timescales, aerosols can either enhance or suppress monsoon convection depending on its properties and spatiotemporal variations. During pre-monsoon and early monsoon months, absorbing aerosols like locally emitted BC and soot from domestic and industrial sources, as well as transported dust from West Asia, accumulate over IGP and Tibetan plateau and contribute to invigoration of precipitation through the ‘elevated heat pump’ hypothesis (Lau and Kim 2006; Lau et al. 2006). Increased aerosol loading over the IGP and TP during pre-monsoon season due to transport associated with El Niño causes precipitation enhancements of $\sim 0.5\text{--}1.5\text{ mm day}^{-1}$ through an anomalous aerosol-induced warm core in the atmospheric column (Fadnavis et al. 2017). The heating induced relative strengthening of the cross-equatorial moisture flow reduces the severity of drought in El Niño years. On weekly timescale, atmospheric heating from accumulated dust aerosols over the Arabian Sea strengthens monsoon westerlies and helps in the

Fig. 5.3 Latitude–pressure section showing meridional overturning anomaly during the summer monsoon season. Adapted from Sanap et al. (2015). © Springer. Used with permission

intensification of monsoon precipitation over central India (Vinoj et al. 2014). Additionally, aerosols can cause suppression of rainfall during monsoon breaks via atmospheric stabilization and increased moisture divergence (Dave et al. 2017).

5.2.3 Climate Model Simulations and Future Projections of Aerosol Properties

Quantifying the effects of anthropogenic aerosols on regional climate cannot be done based on observations alone. Modeling studies are needed to enumerate the aerosols–climate interactions and prediction of future climate change. Model intercomparison projects like CMIP and AEROCOM provide a multi-model platform for evaluating the capability of various climate models to simulate the observed variability of atmospheric aerosols. Evaluation of the present-day 550 nm AOD from several models participating in CMIP, AEROCOM with satellite observations (MODIS, MISR) reveals considerable bias (in the range $\sim \pm 0.3$) in aerosol optical properties estimations over the Indian subcontinent (Sanap et al. 2014; Pan et al. 2015; Misra et al. 2016). Very few models (HADGEM2-ES, HADGEM2-CC, IPSL-CM5A-MR) can capture the observed spatiotemporal distribution of aerosol loading over the subcontinent (Sanap et al. 2014; Misra et al. 2016). In the majority of the models, a negative anomaly in aerosol loading mainly occurs over IGP, western India, and the Arabian Sea and is attributed to significant underestimations in BC emissions and wind-driven dust transport. Also, the comparison of model-simulated extinction profiles with CALIOP observations shows that the bias in the models generally occurs in the lower tropospheric levels (below 2 km) and can be attributed to low emissions from agricultural waste burning and biofuel usage in the emission inventories.

In the future, changing climate and changing emissions would result in changes in aerosol concentration and associated forcing. Future projections of aerosol emissions for

2015–2100 have been integrated into the nine different emission scenarios defined for CMIP6 based on new future pathways of societal growth, the Shared Socioeconomic Pathways (SSPs; Gidden et al. 2019). The yearly changes in aerosol optical properties for future scenarios derived using CMIP6 emission scenarios (Gidden et al. 2019) and MAC-SP parametrization show a pronounced decrease in global AOD by 2100 in all the scenarios excepting SSP3-70 and SSP4-60 (Fiedler et al. 2019). The time evolution of annual mean 550 nm AOD from nine different scenarios during the period 2015–2100 for the Indian landmass is plotted in Fig. 5.4. Most of the scenarios show increasing AOD during the initial period, with the maximum positive trend occurring in SSP5-85 and SSP3-34OS. Excepting SSP3-70, for the other scenarios, anthropogenic aerosol loading over India is projected to decline after 2030–2050 and reach levels much lower than the present aerosol level by the year 2100. The decrease in 550 nm AOD by 2100 for all the nine scenarios ranges between -66.5% and -0.63% with SSP1-19 producing the least aerosol forcing and SSP3-70 generating the maximum aerosol forcing over the Indian subcontinent. A projected decrease in dust aerosols over India by 2100 has also been reported causing changes in precipitation and soil moisture (Pu and Ginoux 2018). Climate projections of future emission scenarios show significant impacts on the north–south temperature gradient over India and Indian monsoon (Guo et al. 2015).

5.2.4 Aerosol Radiative Forcing

5.2.4.1 Radiative Forcing Due to All Aerosols

The annual global mean estimates of direct radiative forcing for aerosol speciation are (1) $-0.4 \pm 0.2 \text{ W m}^{-2}$ for sulfates; (2) $-0.09 \pm 0.06 \text{ W m}^{-2}$ for organic carbon; (3) $+0.40 \pm 0.40 \text{ W m}^{-2}$ for soot; (4) $+0.00 \pm 0.20 \text{ W m}^{-2}$, $-0.10 \pm 0.2 \text{ W m}^{-2}$, and $-0.11 \pm 0.2 \text{ W m}^{-2}$ for biomass burning, mineral dust, and nitrate, respectively (IPCC 2013). ACCMIP models show all-sky 1850 to 2000 global mean annual average total aerosol

Fig. 5.4 Time evolution of 550 nm AOD averaged over India for different scenarios

radiative forcing is -0.26 W m^{-2} (-0.06 to -0.49 W m^{-2}); however, screening based on model skill in capturing observed AOD yields the best estimate of -0.42 W m^{-2} (-0.33 to -0.50 W m^{-2}) (Shindell et al. 2013). The comparison of aerosol RF at the TOA (2005–1850) in CMIP5 models with the Modern Era Retrospective-analysis for Research and Analysis (MERRA) indicates that most of the CMIP5 models have underestimated aerosol RF over South Asia (Sanap et al. 2014). The ACCMIP model shows global mean pre-industrial to present-day aerosols direct RF (1850 and 2000) is $-0.26 \pm 0.14 \text{ W m}^{-2}$ (-0.06 to -0.49 W m^{-2}), and top-of-atmosphere (TOA) is $-1.2 \pm 0.5 \text{ W m}^{-2}$ although variability across models is large in many locations (Shindell et al. 2013).

Over the Indian landmass, there is a large spatiotemporal variation in both magnitude and sign ranging from -26 to $+14 \text{ W m}^{-2}$ at TOA and -63 to -2.8 W m^{-2} at the surface. A recent comprehensive analysis of aerosol direct radiative effect (DRE) by Nair et al. (2016) based on 27 ARFINET observatories, four AERONET stations, and four IMD stations estimated the seasonal variation of DRE over India. The regional mean DRE for TOA and surface is reported to be -8.6 ± 3 and -28.2 ± 12 , -26 to $+14 \text{ W m}^{-2}$, respectively, during winter and -6.8 ± 4 , -33.7 ± 12 – -26 to $+14 \text{ W m}^{-2}$ during spring. The effective radiative forcing calculated by models indicates that over India, as high as 580 mW m^{-2} is because of residential biofuels. However, this is offset due to sulfate from power plants, which contribute about -30 mW m^{-2} (Streets et al. 2013).

In western India, observations of aerosol surface forcing over Ahmedabad, Gujarat, showed that surface forcing was found to be highest during post-monsoon ($-63 \pm 10 \text{ W m}^{-2}$), followed by dry ($-54 \pm 6 \text{ W m}^{-2}$) and lower values during pre-monsoon ($-41.4 \pm 5 \text{ W m}^{-2}$) and monsoon ($-41 \pm 11 \text{ W m}^{-2}$) seasons (Ganguly et al. 2005).

In the northeast of India, atmospheric aerosol radiative forcing estimated from the ARFINET observations over India (2010–2014) shows the highest radiative forcing in the pre-monsoon season ranging from 48.6 W m^{-2} in Agartala to 25.1 W m^{-2} in Imphal. Wintertime radiative forcing follows the pre-monsoon season at these locations. The heating rate resultant from this forcing is high at 1.2 K day^{-1} and 1.0 K day^{-1} over Shillong and Dibrugarh, respectively, in this season. However, Agartala experiences higher surface forcing (-56.5 W m^{-2}) and, consequently, larger heating of the atmosphere (1.6 K day^{-1}) in winter (Pathak et al. 2016). In the case of the top-of-atmosphere (TOA), radiative forcing is found to be negative during dry ($-26 \pm 3 \text{ W m}^{-2}$) and post-monsoon (-22 W m^{-2}), while positive values are obtained during monsoon (14 W m^{-2}) and pre-monsoon (8 W m^{-2}). Large differences between TOA and surface forcing during monsoon and pre-monsoon indicate a large

absorption of radiant energy ($\sim 50 \text{ W m}^{-2}$) within the atmosphere during these seasons. It can lead to heating rates as high as 5.6 K day^{-1} (Ganguly and Jayaraman 2006).

In north India, observations at New Delhi show a consistent increase in surface forcing, ranging from -39 W m^{-2} (March) to -99 W m^{-2} (June) and an increase in heating of the atmosphere from 27 W m^{-2} (March) to 123 W m^{-2} (June). Heating rates in the lower atmosphere (up to 5 km) are 0.6, 1.3, 2.1, and 2.5 K day^{-1} from March, April, May, and June 2006, respectively (Pandithurai et al. 2008). Observations at a semi-urban location, Hisar, showed an increase in the shortwave atmospheric forcing 16 W m^{-2} during clear periods to 49 W m^{-2} for foggy days. Longwave cooling of the atmosphere increased from about -2 W m^{-2} for clear conditions to about -3 W m^{-2} during foggy periods (Ramachandran et al. 2006).

Observations at Hyderabad displayed diurnally averaged values of direct shortwave radiative forcing in the range of -15 to -40 W m^{-2} at the surface, about 15% lower compared to that over the Bay of Bengal region and 22% higher than over the Arabian Sea. TOA forcing observed was in the range of $+0.7$ to -11 W m^{-2} , about 50% lower compared to both these regions. This results in a heating rate of nearly 0.8 K day^{-1} for the first 2 km in the atmosphere (Ganguly et al. 2005). Additional observations have also been made on cruises, with the INDOEX campaign in the 1990s, and showed that concentrations were relatively high throughout much of the cruise, even when the ship was at considerable distances from land. The northeast monsoonal low-level flow can transport sulfates, mineral dust, and other aerosols from the Indian subcontinent to the ITCZ within 6–7 days. These transports result in an increase in AOD at the equator by as much as 0.2 and a decrease in the solar radiative forcing at the sea surface by about 10 – 20 W m^{-2} (Krishnamurti et al. 1998).

The long-term historical surface temperature variations over the Indian subcontinent reveal an absorbing aerosol-induced statistically significant cooling of about $0.3 \text{ }^{\circ}\text{C}$ since the 1970s (Krishnan and Ramanathan 2002). In concurrence, the temporal and spatial variability in annually averaged global irradiance, diffuse irradiance, and bright sunshine duration over twelve stations of solar radiation network of India Meteorological Department (IMD) evaluated for the period 1971–2005. It showed a consistent decrease in the decadal mean all-sky global solar radiation at the surface for India, which was attributed to aerosols (Soni et al. 2012). The decadal mean of the global solar radiation for the decade was 221.5 W m^{-2} for 1976–1985. From 1986 to 1995, the observed global radiation decreased by 3.6 W m^{-2} and further by 9.5 W m^{-2} during the decade of 1996–2005 (Soni et al. 2012). The declining trend of all-sky global irradiance over India as a whole was 0.6 W m^{-2}

Fig. 5.5 (Left panels) Linear, third-order polynomial, and 5-year moving average fits to annual and seasonal time series of all-sky global irradiance averaged over all the twelve solar radiation stations and to clear-sky global irradiance averaged over eight stations. (Right panels) Linear, third-order polynomial, and 5-year moving average fits to

annual and seasonal time series of all-sky diffuse irradiance averaged over all the twelve solar radiation stations and to clear-sky diffuse irradiance averaged over eight stations, adopted from (Soni et al. 2016). © Elsevier publications. Used with permission

year^{-1} during 1971–2000 and $0.2 \text{ W m}^{-2} \text{ year}^{-1}$ during 2001–2010 (Soni et al. 2016). This decrease in global irradiance is matched with an increase in the diffused radiation over the same period indicating an increase in the aerosol levels, as shown in Sect. 5.2.2 (Fig. 5.5).

5.2.4.2 Radiative Forcing Due to Different Species of Aerosols

Among the different species of aerosols, BC is the most important light-absorbing anthropogenic aerosol that contributes to atmospheric warming (Bond et al. 2013). During pre-monsoon season, high concentrations of BC are observed over northwest India and the IGP region. Radiative transfer calculations from observations suggest that from January to May, diurnal-averaged aerosol forcing at the surface is -33 W m^{-2} , and at the TOA above 100 km, it is observed to be $+9 \text{ W m}^{-2}$ (Badarinath and Madhavi Latha 2006). Similarly, large amounts of BC have been observed in Bangalore, both in absolute terms and fraction of total mass ($\sim 11\%$) and submicron mass ($\sim 23\%$). Estimated surface forcing is as high as -23 W m^{-2} , and TOA forcing is $+5 \text{ W m}^{-2}$ during relatively cleaner periods. The net atmospheric absorption translates to atmospheric heating of

0.8 K day^{-1} for cleaner periods and 1.5 K day^{-1} for less clean periods (Babu et al. 2002). It should be noted that a recent study indicates the reduction in BC over India, suggesting a decrease in BC caused heating of the atmosphere (Manoj et al. 2019).

Over the Indian region, the contribution to net cooling by sulfate aerosols is much larger than over other parts of the world (Verma et al. 2012). Estimates of monthly mean direct radiative forcing from sulfate aerosols using a coarse resolution model over India is high in December and January (-3.5 and -2.3 W m^{-2}), is moderate from February to April and November (-1.3 to -1.5 W m^{-2}) and low during May–October (-0.4 to -0.6 W m^{-2}) (Venkataraman et al. 1999). The sulfate aerosol radiative forcing over INDOEX (Indian Ocean Experiment) domain was found to be -1.2 W m^{-2} during INDOEX-FPP 1998 and -1.85 W m^{-2} during INDOEX-IFP 1999 (Verma et al. 2013). Aerosols originating from India, Africa, and West Asia lead to the reduction of total surface radiation by 40–60% (-3 to 8 W m^{-2}) over the Indian subcontinent and adjoining ocean (Verma et al. 2011). During the northeast winter monsoon, natural and anthropogenic aerosols reduce the solar flux reaching the surface by 25 W m^{-2} , leading to 10–15% less insolation at

the surface. South Asia is the dominant contributor to sulfate aerosols over the INDOEX region and accounts for 60–70% of the AOD by sulfate (Reddy et al. 2004).

5.2.5 Impact of Absorbing Aerosols on Himalayan Snow/Ice Cover

Aerosols also impact the snow albedo in the high mountains in the north of India. During the pre-monsoon season, long-range transport and advection of desert dust over the Himalaya (Duchi et al. 2014) leads to its deposition on Himalayan snow, particularly over western Himalaya, and results in long-term reduction in snow albedo (Gautam et al. 2013). This snow darkening leads to accelerated snowmelt (Lau and Kim 2010). Similarly, dust above clean snow can also lead to the absorption of solar radiation at shorter wavelengths, but the warming is instantaneous and reduces when the dust layer is advected away or deposited (Gautam et al. 2013). Along with dust, light-absorbing BC and organic carbon aerosols from biomass burning get transported and lifted to the third pole from neighboring areas. The impact of BC on snow is similar to that of dust aerosols. On the deposition of BC on snow, it reduces surface albedo leading to increased absorption of shortwave radiation affecting the surface snowmelt.

Kopacz et al. (2011) found that emissions from northern India and central China contribute to the majority of BC in the Himalayas, although the precise source location varies with season. The Tibetan Plateau receives most BC from western and central China, as well as from India, Nepal, the Middle East, Pakistan, and other countries. The magnitude of contribution from each region also varies with season. The effect on this part of the cryosphere is highest in pre-monsoon and lowest during monsoon season (Gertler et al. 2016). Some studies (Xu et al. 2009) have analyzed ice cores for long-term trends of BC deposition on Himalayan glaciers. Xu et al. (2009) reported results from five ice core samples at various locations across Himalaya. Most of the locations show high concentrations in the 1950s–1960s and lower values in 1970–1980s. The authors attributed this temporal variation to the differential long-range transport of European emissions. The cryospheric soot concentrations over southern Himalaya show an increasing trend from the 1990s, which reflects an increase in present-day emissions (Xu et al. 2009). Jacobi et al. (2015) found a 4–5% reduction of albedo from BC over Himalaya. Kaspari et al. (2014) computed the change in albedo due to BC and dust over the Mera glacier and reported a 6–10% albedo reduction during the winter–spring season. The annual snow albedo surface forcing has been found in the range of 3–6 W m⁻² (Jacobi et al. 2015). The radiative forcing in the snow-covered regions due to the BC-induced snow albedo effect can vary

from 5 to 15 W m⁻², an order of magnitude larger than radiative forcing due to the direct effect, and with significant seasonal variation in the northern Tibetan Plateau (Kopacz et al. 2011).

5.2.6 Aerosol-Cloud Interaction: Indirect Effect of Aerosols

Aerosols alter clouds changing the precipitation patterns and intensity over India. Initial attempts in the AIE in the monsoon environment were largely based on the satellite data. Panicker et al. (2010) reported positive (negative) AIE values during drought (excess monsoon) years for four consecutive years from 2001 to 2004 using satellite data demonstrating an inverse relationship between AIE and ISMR. Quantitative estimates of AIE using two different methodologies have given two close values of 0.13 and 0.07 over India, and the difference is attributed to the aerosol effect on the dispersion of cloud drop size distribution (Pandithurai et al. 2012). During monsoon season over India, AIE increases from 0.01 to 0.23, with an increase in liquid water path (Harikishan et al. 2016). Further, AIE derived from cloud drop number concentration and effective radius at different liquid water contents recorded at a high-altitude cloud physical laboratory in the Western Ghats provides a better understanding of the role of aerosol effect on cloud drop dispersion (Anil Kumar et al. 2016).

The Cloud-Aerosol Interaction and Precipitation Enhancement Experiment [CAIPEEX; (Kulkarni et al. 2012)] over India, mainly focusing on continental clouds, documented the precipitation process and associated aerosol-cloud interaction over the region. CAIPEEX in situ observations showed a significant increase in the cloud droplet number concentration with aerosol, as compared to the INDOEX study and several other reported results from around the world (Fig. 5.6). High aerosol loading into the atmosphere enhances the CCN, increasing the number concentration and decreasing the size of cloud droplets and a narrow droplet spectrum. This may further suppress collision coalescence and warm rain may form at an elevated layer (Konwar et al. 2012).

CAIPEEX observations revealed dilution in cumulus clouds over India (Nair et al. 2012) with pre-monsoon clouds in the high aerosol environment having more adiabatic cores than the monsoon clouds (Bera et al. 2019). Also, the clouds have a large amount of supercooled liquid water (>3 gm⁻³) with mixed phase (Prabha et al. 2012). The cloud drop effective radius increases with the height from the cloud base (Prabha et al. 2012) and, the depth of warm rain was having a direct relationship with the subcloud aerosol. A decrease in the droplet spectral width of pre-monsoon cloud droplet sizes due to aerosol was reported by Prabha et al. (2012), and the

Fig. 5.6 Observations of cloud droplet number concentrations and associated aerosol number concentrations with data from INDOEX and CAIPEEX over the Indian continental region, error bars indicate spatial variability. Adapted from Prabha and Khain (2020). © John Wiley and Sons. Used with permission

variation in spectral width was also largely variable with airmass characteristics (Bera et al. 2019). CAIPEEX observations have also documented both higher ice mass and number concentration in monsoon clouds, compared to pre-monsoon clouds with warm microphysics in the monsoon clouds determining the ice and mixed-phase microphysical properties whereas boundary layer moisture plays a key role in the initial developmental stages (Patade et al. 2014).

Documenting the aerosol and associated activation characteristics as cloud condensation nuclei or ice nuclei particles (INP) is essential for better characterization in numerical models. Observational data from Nainital shows that enhanced CCN concentrations coincide more with periods of aerosol absorption as compared to periods of aerosol scattering (Gogoi et al. 2015). Aerosol chemical composition, aerosol number size distribution, and CCN data show that the predictability of CCN improves when SOA component is considered in hygroscopicity estimates (Singla et al. 2017). Precipitation susceptibility estimates showed that clouds having medium liquid water content (0.6–0.8 mm) were highly affected due to aerosols (Leena et al. 2018). The vertical variation of aerosol as CCN is important, as the CCN spectral characteristics are significantly different near the surface and the cloud base (Varghese et al. 2016). Vertical distribution of aerosol types reveals a mixture of both biomass burning and dust aerosols (Padmakumari et al. 2013). The role of aged BC particle or bioaerosol acting as CCN or INP is yet to be investigated over the Indian region. Physical and chemical characterization of aerosol along with CCN and INP activity is required in future studies.

Numerical investigation of aerosol effect over the monsoon region shows that cloud microphysics processes are important for a break to active transition during monsoon season with higher concentrations of absorbing aerosols producing invigoration of convection strong moisture convergence and increased upper level heating (Hazra et al. 2013). Within the deep convective clouds during monsoon, an increase in soluble aerosol led to a marginal increase in precipitation attributing to enhanced updrafts in the warm phase and invigoration of mixed-phase cloud processes (Gayatri et al. 2017). Mixed-phase clouds contribute a significant part of monsoon clouds, which are least understood and need further focused process studies. A systematic approach aerosol impact on the cloud system effects needs to be investigated, and the regional impacts on precipitation through redistribution of clouds need to be understood.

5.2.7 Impact of Convective Transport of Aerosols

During the monsoon season, deep convection transports boundary layer aerosols from Asia to the UTLS (Fadnavis et al. 2013, 2017). These aerosols form a layer near the tropopause (13–18 km) known as the Asian Tropopause Aerosol Layer 'ATAL' (Vernier et al. 2009). Development of the ATAL is associated with convective transport of aerosols from the lower atmosphere to the UTLS (Fadnavis et al. 2013; Vernier et al. 2015). Observations from satellites

(CALIPSO, SAGE-II), balloonsonde, and the CAREBIC aircraft reveal that the ATAL is composed of nitrates, sulfate, BC, organic aerosols, and dust particles (Vernier et al. 2018). Studies indicate that these aerosols are transported into the lower stratosphere and produce a significant impact on stratospheric temperature and circulations (Fadnavis et al. 2017). MERRA-2 data also shows abundant quantities of carbonaceous aerosols and dust in the mid and upper troposphere over India, arising from enhanced biomass burning emissions as well as westerly transport from the Middle East deserts during May–June (Lau et al. 2018). Model simulations indicate that carbonaceous aerosol transport into the UTLS enhances heating rates by ~ 0.03 – 0.08 K per day in the upper troposphere (300–100 hPa). These carbonaceous aerosols induce a seasonal mean anomaly aerosol radiative forcing of $\sim +0.37 \pm 0.26$ W m $^{-2}$ at the TOA and -4.74 ± 1.42 W m $^{-2}$ at the surface (Fadnavis et al. 2017). Asian summer monsoon anticyclone region contributes an increase of $\sim 15\%$ to the Northern Hemisphere column stratospheric aerosol. This elevated aerosol layer also aids in aggravating monsoon droughts during an El Niño episode (Fadnavis et al. 2019).

5.3 Trace Gases

Ozone variations in the troposphere and stratosphere play a key role in maintaining the Earth's radiative budget and climate change (Logan et al. 2012); it is important to know its assessment. In this section, we provide an assessment of its trends from past literature. Trend estimates in total ozone column, tropospheric ozone column, and surface measurements are reported from in situ observations, satellites remote sensing, and model simulations.

5.3.1 Trends in Ozone

5.3.1.1 Total Ozone Column

The past studies report estimates of trends in the total ozone column at various stations widespread over the Indian region. Although, there are limited ozone monitoring stations over India, trend estimates from ground-based measurements like Dobson spectrophotometer (DU year $^{-1}$) and satellite remote sensing (% year $^{-1}$) are consistent. Measurements over north India (20–35° N) show statistically significant (σ uncertainty level) negative (declining) trends while they are positive (increasing) over south India (8–20° N). For example, Multi-Sensor Reanalysis (MSR/MSR-2) and TOMS observations over north India show a decreasing trend of -0.08 to -0.15% year $^{-1}$ during 1979–2008 (Tandon and Attri 2011) and -0.03 to -0.11% year $^{-1}$ during January 1979–December 2012 (Sahu et al. 2014). The observations in south India show a positive trend of 0.01 – 0.03% year $^{-1}$ during January 1979–December 2012. Dobson spectro-photometer tropospheric column ozone measurements also show a decreasing trend -0.01 DU year $^{-1}$ at Varanasi, (in north India), and positive trend $+0.14$ DU year $^{-1}$ at Kodaikanal, (in south India) during 1957 and 2015 (Pathakoti et al. 2018). The above studies indicate that amplitude of trend varies with location and time period of measurement, and the trend estimates have medium confidence.

In the global context, the ozone assessment report shows that total ozone has been stable since about 1996 in response to emission control of ozone-depleting substances (ODSs) (Chehade et al. 2014; Zvyagintsev et al. 2015). Future trends in total column ozone over the globe and tropics (25° S–25° N) are tabulated in Table 5.1 (Cionni et al. 2011; Eyring et al. 2013).

Table 5.1 Ozone trend over the Indian region and CMIP5 annual mean future trends over tropics

Ozone trends	Indian region	Tropics 25° S–25° N from CMIP5 annual mean future trends (2090s–2100s) in the RCPs (Cionni et al. 2011)
Total column ozone	North India: -0.03 to -0.11% year $^{-1}$ South India: $+0.01$ to $+0.03\%$ year $^{-1}$ (1979–2012) (Sahu et al. 2017)	RCP 2.6: -2 DU (-1%) RCP 4.5: 0 DU (0%) RCP 6.0: 0 DU (0%) RCP 8.5: 7 DU (4%)
Troposphere	Tropospheric column: 0.3 ± 2.6 to $2.7 \pm 2.3\%$, year $^{-1}$ (Saraf and Beig 2004)	RCP 2.6: -4 DU (-17%) RCP 4.5: -2 DU (-10%) RCP 6.0: -2 DU (-10%) RCP 8.5: $+5$ DU (18%)
Stratosphere	0.27 ± 0.67 to $1.3 \pm 0.65\%$ year $^{-1}$ (~ 200 – 50 mb) (1993–2005) -0.45 ± 0.8 to $-0.57 \pm 0.62\%$ year $^{-1}$ (30 – 10 mb) (1993–2005) (Fadnavis et al. 2014a)	RCP 2.6: 2 DU (1%) RCP 4.5: 2 DU (1%) RCP 6.0: 2 DU (1%) RCP 8.5: 2 DU (1%)

5.3.1.2 Tropospheric Ozone

Although a majority of the ozone is concentrated in the stratosphere, tropospheric ozone plays a vital role in atmospheric chemistry, determining the oxidative capacity of the atmosphere through the production of the hydroxyl radical (OH), and can also act as a pollutant affecting human health and crop productivity. Efforts have been taken toward estimating trends in surface and tropospheric column ozone. The estimated amplitude of trend varies with season and location (urban and rural). The annual mean trends in tropospheric column ozone derived by integrating the vertical profiles of ozonesonde data (in situ observations on balloon platforms) over Delhi, Pune, and Trivandrum (1972–2001) show an increasing trend of $2.7 \pm 2.3\%$, $0.9 \pm 1.8\%$, and $0.3 \pm 2.6\%$ year⁻¹, respectively. These values of trends are in close agreement with that obtained from TOMS data (Saraf and Beig 2004). Nimbus-7 and Earth Probe satellite—Total Ozone Mapping Spectrometer (TOMS) data for the period of 1979–2005 show positive trends 0.7 – 0.9% year⁻¹ over South Asia. Also, the trends estimated from the MOZART model are in agreement with observations over the Bay of Bengal region (0.4 ± 0.29 – $0.6 \pm 0.43\%$ year⁻¹, Beig and Singh 2007). The regressed tropospheric ozone residual (TOR) data shows an annual trend of $\sim 0.4 \pm 0.25$ 1 $\sigma\%$ per year over the northeastern Gangetic region (Lal et al. 2012). Similar estimates of trends in ozone at various stations in India are reported (Lal et al. 2013, 2014, 2017).

The multi-model ensemble-mean from CMIP5 historical simulations (2009–2000) shows an increase in tropospheric column ozone ~ 25 – 35 DU (decadal mean) over the Indian region. Future projections (2090–2100) are tabulated in Table 5.1. ACCMIP multi-model simulations for relative changes of tropospheric ozone between 2000 and 2030 (2100) for the different RCPs show decrease of -5% (-22%) in RCP 2.6, 3% (-8%) in RCP 4.5, 0% (-9%) and increase in 15 RCP 6.0, and 5% (15%) in RCP 8.5 (Young et al. 2013). However, there are large uncertainties in model simulations over the Indian region due to uncertainties in emission inventories, model parameterization, chemistry representation, etc. (Fadnavis et al. 2015).

Surface ozone observations have shown an increasing trend at various locations around India, which are attributed to increasing anthropogenic activity. Naja and Lal (1996) reported increasing ozone by 14.7 ppbv during 1954–1955 and 25.3 ppbv during 1991–1993, which results in linearly increasing trends of 1.45% year⁻¹. The seasonal trends also show a significant increase, e.g., winter $\sim 1.91\%$ year⁻¹ and summer 0.86% year⁻¹ at Ahmedabad (23° N, 75.6° E). At southern peninsula station, Thiruvananthapuram (8.542° N, 76.858° E), surface ozone measurements were obtained during 1973–1975, 1983–1985, 1997–1998, and 2004–2014 (Nair et al. 2018). These measurements show a slow increase

of ~ 0.1 ppb year⁻¹ during 1973 to 1997 and faster growth of 0.4 ppb year⁻¹ afterward till 2009 after which it showed a steady-state till 2012 followed by a minor decrease. The above studies show that trends in surface and tropospheric column ozone are positive but have low confidence.

Profiles of ozone show variations in the vertical structure of ozone. From ozonesonde observations, Saraf and Beig, (2004) reported long-term trends in ozone at Trivandrum, Pune, and Delhi. The observed trend at Delhi was increasing between 1.5% year⁻¹ and 7.3% year⁻¹ during 1972–2001, but negative trends with low confidence (statically insignificant) ~ -0.5 to -3% year⁻¹ were observed at Pune and Trivandrum. Seasonal trends in tropospheric ozone are positive, particularly around 500 hPa and 200–300 hPa during months of January–March. The role of biomass burning and stratosphere–troposphere exchange is evident in these two layers. There is also a prominent increasing trend in ozone near the tropopause (~ 100 hPa) during June and July (monsoon month) (Fadnavis et al. 2014a). The GEOS-Chem model simulations also show a positive trend of 0.19 ± 0.07 ppb year⁻¹ (p -value < 0.01) (an annual mean) in the lower troposphere between 1990 and 2010 (Lu et al. 2018).

5.3.1.3 Ozone Trends in the Upper Troposphere and Stratosphere

It is important to understand ozone variability and trends in the UTLS since a small amount of variation of ozone in the UTLS has a large impact on radiative forcing and climate change (Forster and Shine 1997). Fadnavis et al. (2014a) reported an increasing ozone trend between 0.6 ± 0.65 and 2.35 ± 1.3 year⁻¹ in the upper troposphere and in the lower stratosphere from multiple satellite data sets and model simulations. The estimated trends are slightly positive up to 30 hPa and then negative between 30 and 10 hPa. Seasonal mean trends vary between -0.04 ± 0.3 and $3.48 \pm 2\%$ year⁻¹ (low confidence). In the stratosphere (20–50 km), ozone shows a decreasing trend of (medium confidence) $\sim -0.4 \pm 0.1\%$ year⁻¹ near 16–20 km while trends values are positive near 24–30 km (0.05 ± 0.04 to $0.1 \pm 0.9\%$ year⁻¹) (low confidence) during 1993–2015 (Raj et al. 2018).

A decrease in abundance of ozone-depleting substances (ODSs) under the compliance of the Montreal Protocol was the start of the recovery of stratospheric ozone. Since the atmospheric burden of ozone-depleting substances is declining, changes in CO₂, N₂O, and CH₄ will have an increasing influence on the ozone layer (WMO 2019b). Ozone layer changes in the latter half of this century will be complicated, with projected increase or decrease in different regions. Eyring et al. (2013) reported the evolution of stratospheric ozone over the CMIP5 historical period (1960 to 2005) and the sensitivity of ozone to future GHGs (2006–

2100) for the four different RCPs (2.6, 4.5, 6.0, and 8.5). The simulations with the 1980 baseline-adjusted stratospheric column ozone (time series from 1960 to 2100) over the tropics (25° S–25° N) show a decrease in tropical lower stratospheric ozone (100–30 hPa) and increase in the upper stratosphere (Cionni et al. 2011). A summary of annual mean trends in ozone in the troposphere and stratosphere discussed above from observations, and CMIP5 multi-models future projections are listed in Table 5.1.

5.3.2 Emissions of Ozone Precursors

One of the largest uncertainties in modeling studies is emission inventories. Global and regional emission inventories carry large uncertainties, especially in regions where observational data are sparse. In this section, we provide a brief overview of emissions of ozone precursors, e.g., NO_x, CO, and NMVOCs, over India. Jena et al. (2015) reported total NO_x flux ~1.5, 2.1, 2.4, 1.9, 1.7, and 1.4 Tg N year⁻¹ over India from six different inventories. Thermal power plants contribute 30% of the total NO_x emissions in India (Garg et al. 2006). The total surface NO₂ emissions in India are ~3.5 Tg year⁻¹ in 1991 and ~4.3 Tg year⁻¹ in 2001 (Beig and Brasseur 2006). The total NO_x emissions in 2005 amount to ~1.9 Tg N year⁻¹ (Ghude et al. 2013b). The growth in oil and coal consumption resulted in a growth rate of 3.8% ± 2.2% year⁻¹ between 2003 and 2011 for anthropogenic NO_x (Ghude et al. 2013a). This growth rate is comparable with the estimate made by EDGAR (V4.2; 4.2% year⁻¹), GAINS (3.6% year⁻¹), or (Garg et al. 2006) (4.4% year⁻¹) emission inventories. Sadavarte and Venkataraman, (2014) reported estimates of NO_x emissions ~5.6 (1.7–15.9) Tg year⁻¹ in 2015.

CO emissions show annual growth rate of 1.1% during 1985–2005 (Garg et al. 2006). The annual growth rate of CO from the transport sector is ~8.8% during 2001–2013 (Singh et al. 2017). The total CO emissions from India were 59.3 Tg year⁻¹ in 1991 and 69.4 Tg year⁻¹ in 2001 (Beig and Brasseur 2006). The CO emission from wheat straw burning in 2000 was 541 ± 387 Gg year⁻¹ (Sahai et al. 2007). Venkataraman et al. (2006) estimated ~13–81 Gg year⁻¹ of CO from biomass burning during 1995–2000. In 2000, CO emissions in India (63.3 Tg) were ~23% of Asia (279 Tg) (Streets et al. 2003).

Biogenic emissions are the largest natural source (~90%) of volatile organic compounds (VOCs) in the atmosphere (Guenther et al. 2006). The annual emissions of VOCs in India from anthropogenic and biomass burning sources were ~10.8 Tg and 2.2 Tg, respectively, in 2000 (Streets et al. 2003). Total anthropogenic emissions of non-methane volatile organic compounds (NMVOCs) were

9.81 Tg in 2010 (Sharma et al. 2015). The majority of NMVOCs emissions (60%) originated from residential combustion of biomass for cooking. Venkataraman et al. (2006) estimated NMVOC emission ~2.04–7.41 Tg year⁻¹ from biomass burning during 1995–2000 over India.

5.3.3 Trends of Tropospheric NO_x, CO, NMVOCs, and PAN

Trends in some of the ozone precursors (NO_x, CO, VOCs) are reported over the Indian region. Satellite observations from GOME, GOME-2a, OMI, and SCIAMACHY during 2002–2011 show a trend of 2.20 ± 0.73% year⁻¹ in NO₂ volume mixing ratios over India (Mahajan et al. 2015). While NO₂ volume mixing ratios from 1996 to 2006 showed an increasing trend of 1.65 ± 0.52% year⁻¹ in over India. The industrial regions of Mumbai and Delhi show increasing trends of 2.1 ± 1.1 and 2.4 ± 1.2% year⁻¹, respectively (Ghude et al. 2008). CO observations from MOPITT (Measurements of Pollution in the Troposphere) satellite during 2000–2014 show contrasting trends in the lower and upper troposphere. Estimated trends in lower-troposphere and columnar CO are negative -2.0 to -3.4 ppb year⁻¹ (-1.1 to -2.0% year⁻¹) and positive 1.4–2.4 ppb year⁻¹ (1.8–3.2% year⁻¹) in the upper troposphere (Girach et al. 2017). AIRS/AMSU satellite (2003–2012) shows a 2% increase in tropospheric CO concentration over the Indian region (Ul-Haq et al. 2015) (Fig. 5.7a). Emission estimates based on technology also show increasing trends ~19 Tg year⁻¹, e.g. (Sadavarte and Venkataraman 2014). Peroxyacetyl nitrate (PAN) is formed in biomass burning plumes. It is a secondary pollutant produced through the oxidation of VOCs and NO_x released from anthropogenic and biogenic sources. Recent satellite observations show an increasing trend in PAN ~0.1 ± 0.05 to 2.7 ± 0.8 ppt year⁻¹ during 2005–2012 in the UTLS over Asia (Fadnavis et al. 2015) (Fig. 5.7a). A significant increase in amounts of VOCs and air pollutants is observed (May 2012) in the Indo-Gangetic Plain (IGP). These observations show extremely high levels of both VOCs and the primary air pollutants in the evening and early morning hours in May 2012 (Fig. 5.7b). These increasing levels of VOCs may be contributing to positive trends in PAN in the UTLS. The observed trends in NO_x and PAN have high confidence, while CO and VOCs have low confidence. Ozone and its precursor gases, PM_{2.5}, PM₁₀, are being monitored since 2010 at various Indian stations by System of Air Quality Forecasting and Research (SAFAR) which is developed by the Indian Institute of Tropical Meteorology. Long-term observations from SAFAR will be helpful in obtaining future trends in ozone and its precursors over the India region.

Fig. 5.7 **a** Trends in trace gases over the Indian region. These trends are adopted from Beig and Singh 2007; Fadnavis et al. 2014a; Mahajan et al. 2015; Girach et al. 2017; Sahu et al. 2017. **b** Time series of the one-minute data in May 2012 for the mixing ratios of ozone (top panel),

and NO₂ and NO (second panel), SO₂ (third panel), CO (fourth panel), and mass concentrations of PM_{2.5} and PM₁₀ (bottom panel) adopted from (Sinha et al. 2014), Fig 5.7b. © Copernicus publications. Used with permission

5.3.4 Variations in Ozone and NO_x Due to Lightning

The composition of trace gases in the troposphere is influenced by lightning in addition to anthropogenic emissions. It is estimated that lightning contributes to about 10% of the global annual NO source (Schumann and Huntrieser 2007). It can contribute up to 90% variation of NO_x at the altitudes of 5–15 km. Variation of ozone in the middle/upper troposphere due to lightning may change ozone heating rates and may have an impact on Asian monsoon circulation (Roy et al. 2017).

Over the Asian region, lightning contributes ~40% to NO_x and 20% to ozone production in the middle and upper troposphere during the monsoon season (Fadnavis et al. 2014a). Previous studies (Bharali et al. 2015) have reported an increase in the O₃ mixing ratio ~18 ppbv during pre-monsoon and ~12 ppbv during summer associated with the lightning activity over Dibrugarh (27.4° N, 94.9° E) in northern India and over Hyderabad (17.44° N, 78.30° E) (a station in southern peninsular India) (Venkanna et al. 2016). Kavitha et al. (2018) reported an enhancement in NO_x (5.2–8.7 ppbv) and an associated reduction in surface O₃ mixing ratio (9.9–18.8 ppbv) during pre-monsoon and monsoon seasons due to lightning activity.

5.3.5 Radiative Forcing due to Ozone and Precursor Gases

The radiative forcing (RF) due to changes in tropospheric and stratospheric ozone is the third-largest GHGs contributor to RF since pre-industrial times. According to the (IPCC 2013), the total increase in global radiative forcing due to changes in ozone is +0.35 (0.15–0.55) W m⁻² (high confidence), with radiative forcing due to tropospheric ozone

+0.40 (0.20–0.60) W m⁻² (high confidence) and due to stratospheric ozone –0.05 (–0.15 to +0.05) W m⁻² (high confidence). ACCMIP tropospheric ozone future projections (2100–1850) show global mean annual average anthropogenic forcing $\sim 0.14 \pm 0.12$ W m⁻² in RCP 2.6, 0.23 ± 0.15 W m⁻² in RCP 4.5, 0.25 ± 0.09 W m⁻² in RCP 6.0, and 0.55 ± 0.30 W m⁻² in RCP 8.5.

According to the CMIP5 estimates, the tropospheric ozone radiative forcing from the 1850s to the 2000s is +0.23 W m⁻², lower than the IPCC estimate (IPCC 2013). The lower value is mainly due to (i) a smaller increase in biomass burning emissions; (ii) a larger influence of stratospheric ozone depletion on upper tropospheric ozone at high southern latitudes; and possibly (iii) a larger influence of clouds (which act to reduce the net forcing). Over the same period, decreases in stratospheric ozone, mainly at high latitudes, produce an RF of –0.08 W m⁻², which is more negative than the IPCC but is within the stated range of –0.15 to +0.05 W m⁻² (Cionni et al. 2011; Eyring et al. 2013).

Estimates over India suggest that the radiative forcing has changed in the range between 0.2 and 0.4 W m⁻² since pre-industrial times (Chalita et al. 1996). The radiative forcing effect from tropospheric ozone is regional due to its short lifetime. The model simulations with 10% reductions in the precursor's emission over India resulted in a decrease of ~ 0.59 W m⁻² (Naik et al. 2005).

For the other trace gases mentioned in this chapter, the resultant effect on radiative forcing is not direct. Gases such as CO, NO_x, and VOCs are precursors of ozone and hence have an indirect impact on radiative forcing. Additionally, gases such as sulfur dioxide (SO₂) and NO_x also contribute to the formation of sulfate and nitrate aerosols, which can have a net cooling effect on the atmosphere. Globally, the contribution from CO and NMVOCs toward ozone radiative forcing is estimated to be about +0.2 (–0.18 to +0.9) W m⁻²

and 0.1 (−0.06 to +0.14) W m^{-2} . For NO_x , due to its role in nitrate aerosol formation, the best estimate is a resultant negative forcing of −0.15 (−0.34 to +0.02) W m^{-2} . For sulfur dioxide, the estimate is −0.41 (−0.62 to −0.21) W m^{-2} (IPCC 2013).

5.4 Influence of Transport Processes

Monsoon sustains a remarkably efficient cleansing mechanism in which contaminants are rapidly oxidized and deposited to Earth's surface. However, some pollutants are lifted above the monsoon clouds due to deep convection and are chemically processed in a reactive reservoir before being redistributed globally, including to the stratosphere. Numbers of studies based on satellite remote sensing indicate the transport of CO , H_2O , PAN, Hydrogen cyanide (HCN), CH_4 , NO_x , etc., from the Asian boundary layer to the UTLS (Fadnavis et al. 2013, 2015, 2017). Enhancement of trace gases in the UTLS during the monsoon season alters local heating rates and radiative balance. Transported NO_x and associated ozone variations in the UTLS enhances the ozone heating rates by $\sim 1\text{--}1.4 \text{ K day}^{-1}$ in the upper troposphere (400–200 hPa) and radiative forcing $\sim 16.3 \text{ m W m}^{-2}$ over the Indian region. There is a positive impact of ozone heating rates and radiative forcing on the Indian monsoon circulation (Roy et al. 2017). There is a convective injection of polluted water vapor from the Asian region into the UTLS, which is then dispersed into the global stratosphere by the large-scale upward motion (Fu et al. 2006). The H_2O feedback amplifies the radiative forcing of anthropogenic greenhouse gases by a factor of ~ 2 .

In the lower troposphere, the transport of trace gases occurs to and from India with seasonal variations in the wind. The seasonal variation in most trace gases shows a dip during the monsoon season due to efficient wet scavenging by precipitation and the transport of clean marine air. Integrated Campaign for Aerosols, gases, and Radiation Budget (ICARB) measurements during the pre-monsoon season show elevated levels of CO (~ 100 ppb) over the Bay of Bengal and the Arabian Sea. These studies reveal that high amounts of marine CO are attributed to transport from the Indian subcontinent (Aneesh et al. 2008). Satellite observations also show high values of CO (130–160 ppb) and ozone (120–130 ppb) at 825 hPa near the location of cyclones occurring in the Bay of Bengal and Arabian Sea (Fadnavis et al. 2011).

The in situ observations are unable to explain the different atmospheric processes accountable for high pollution events. Therefore, chemistry transport models are valuable for providing a large-scale view of the regional impact of these gases and are useful for the interpretation of observations on local to global scale (Yarragunta et al. 2017). The simulated ozone

concentrations from the MOZART4 model when evaluated against ground-based observations revealed that the model captures the seasonal cycle of ozone amounts but overestimates the values of ozone concentration. The magnitude of observed ozone is in the range of 7–60 ppbv, whereas the quantity of simulated ozone is $\sim 27\text{--}53$ ppbv (Yarragunta et al. 2018). Lower tropospheric ozone over India during 2006–2010 as observed from OMI showed the highest concentrations (54.1 ppbv) in the pre-summer monsoon season (May) and the lowest concentrations (40.5 ppbv) in the summer monsoon season (August). Analyses from the GEOS-Chem model showed that the onset of the summer monsoon brings ozone-unfavorable meteorological conditions which all lead to substantial decreases in the lower tropospheric ozone burden (Lu et al. 2018). The influence of springtime (MAM) biomass burning in central India, the Indo-Gangetic region and the Bay of Bengal, on regional ozone distribution has been evaluated using a regional chemical transport model (WRF-Chem), and the Fire Inventory from NCAR (FINNV1). These simulations demonstrated that the springtime fire emissions have a significant impact on the ozone in this region (Jena et al. 2015).

5.4.1 Influence of Stratosphere to Troposphere Transport

Transport associated with tropopause folding produces a significant variation in ozone, humidity, and temperature. MLS and AIRS satellites show intrusion events of ozone-rich dry stratospheric air over northern India and the Tibetan Plateau region occurring every winter and pre-monsoon season. It enhances ozone amounts by $\sim 100\text{--}200$ ppmv in the UTLS (300–100 hPa) (Fadnavis et al. 2010). Tropopause folding in the subtropical westerly jet during the monsoon seasons sheds eddies into the deep troposphere (~ 700 hPa) which are a carrier of ozone-rich cold and dry air. These eddies spread stratospheric air in the upper troposphere, increasing the static stability of the troposphere (Fig. 5.8). These stratospheric dry air intrusions are associated with monsoon breaks and are evident in observations during 1979–2007 (Fadnavis and Chattopadhyay 2017).

The stratospheric folding tends to occur on the north-western side of the upper-level anticyclone resulting in intensified subsidence and reduces extreme rainfall upstream of the fold, while it enhances the precipitation at downstream of the fold. A typical pattern of suppression of extreme rainfall upstream and promotion downstream of the fold persists for about 1–2 days. Rossby wave breaking over West Asia inhibits deep monsoonal convection and thereby leading to a dry spell over India (1998–2010) (Samanta et al. 2016).

Fig. 5.8 Time-pressure cross section of anomalies in **a** temperature (K) averaged over 30–50° N, 75–110° E, **b** RH (%) averaged over 25–40° N, 60–75° E, **c** square of Brunt–Väisälä frequency (per sec*1E-5)

averaged over 30–50° N, 75–110° E. Adapted from Fadnavis and Chattopadhyay (2017). © American Meteorological Society. Used with permission

Distribution of trace gases in the UTLS is also affected by the stratospheric Brewer–Dobson circulation (Brewer 1949; Dobson 1956). The Asian summer monsoon is an important pathway for the transport of Asian tropospheric constituents into the stratosphere (Fadnavis et al. 2013, 2017). The HALOE aircraft observations of N_2O , CO, and O_3 indicate a significant increase in the impact of the South Asian tropospheric pollutants on the extratropical lower stratosphere (Müller et al. 2016). Inter-annual variations of stratospheric N_2O , CFC-11 (CCl₃F), and CFC-12 (CCl₂F₂) are modulated by the BDC. Satellite observations show that the transport of water vapor and HCN from the South and Southeast Asia occurs into the lower stratosphere by the monsoon convection and is then re-circulated by the Brewer–Dobson circulations. Thus, Asian trace gases and aerosols affect the chemical composition of the extratropical stratosphere (Fadnavis et al. 2013).

5.4.2 Influence of Transport Associated with Quasi-biennial Oscillation

The phenomenon of the equatorial quasi-biennial oscillation (QBO) is known to produce a significant impact on dynamics and chemistry over the tropical region. Studies indicate that the secondary meridional circulation induced by QBO produces a double peak structure in the stratosphere at the equator with maximum amplitude in the temperature and ozone at two pressure levels 30 and 9 hPa and a node at 14 hPa. Phase structure reveals that the temperature QBO descends faster than the ozone QBO (Fadnavis et al. 2008).

Past studies indicate that cyclones are modulated by the phases of the QBO (Fadnavis et al. 2011, 2014b). In post-monsoon season, during the east phase, cyclones move westward/northwestward while during the westerly phase, they move northward/northeastward. During pre-monsoon

season, cyclones move northward/northeastward irrespective of phases of QBO. The possible interaction between the stratospheric QBO and cyclone is explained from the variation of winds, geopotential height, tropopause pressure, OLR, and SST (Fadnavis et al. 2011). QBO shows an influence on Indian Summer Monsoon Rainfall (ISMR). The ISMR is stronger during the west phase of QBO and weak during the east phase (Rai and Dimri 2017). QBO also influences the stratospheric aerosol layer. Satellite observations show that QBO modulates the vertical extent of the stratospheric aerosol layer in the tropics by up to 6 km, or ~35% of its mean vertical extent between 100 and 7 hPa (about 16–33 km) (Hommel et al. 2015).

5.5 Impact of Volcanic Eruptions

Volcanoes inject huge amounts of aerosols and trace gases in the upper troposphere and stratosphere, thereby drive the natural mode of climate variability through alteration of radiative forcing (Robock 2015). A volcanic eruption in the vicinity of India, e.g., Mt. Nabro during 11–13 June 2011, injected a large amount of water vapor, and SO_2 (1.3–2.0 Tg) in the upper troposphere and lower stratosphere over India. The aerosols injected into the stratosphere traveled large distances and thickened the stratospheric aerosol layer. The global lidar networks (EARLINET, MPLNET, and NDACC) and satellite (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation, (CALIPSO)) show that Mt. Nabro has increased stratospheric volcanic AOD by 0.003–0.04 (global mean) and heating by $\sim 0.3 \text{ K day}^{-1}$ between 16 and 17 km altitude (Fairlie et al. 2014). The aerosol surge causes tropospheric cooling and stratospheric warming by scattering and reflecting incoming solar radiation (von Glasow et al. 2009). Large volcanoes modulate the Inter-Tropical Convergence Zone via changes in the

hemispheric temperature gradient. There is a southward shift in the ITCZ location and South Asian Monsoon (Sinha et al. 2011) after the volcanic eruptions occurred during the last millennium (Schneider et al. 2014). A host of modeling studies shows a consistent decrease in Asian summer monsoon rainfall following volcanic (Zambri et al. 2017), barring a few studies which report an increase in the precipitation response in the post-eruption period, due to change in the land–sea thermal gradient (Joseph and Zeng 2011). Volcanic eruptions also influence climate; that is, it triggers El Niños (Ohba et al. 2013).

One of the important impacts of volcanoes is the loss of stratospheric ozone. The ozone loss and associated changes in photolysis rates affect the tropospheric/stratospheric temperature (cooling/warming) (Santer et al. 2003). The stratospheric ozone loss is linked with chemical reactions occurring over aerosol surfaces. There is a reduction of nitrogen oxides and chlorine activation, which leads to an increase in Equivalent Effective Stratospheric Chlorine (EESC) (Tie and Brasseur 1995; Tabazadeh et al. 2002). A drastic decrease in the stratospheric ozone over Antarctica due to a series of volcanic eruptions has been proposed to lead to large-scale changes in atmospheric dynamics resulting in massive de-glaciations in the past (McConnell et al. 2017). Previous work also indicates that volcanic eruptions can serve as a source of potential predictability (Gaddis 2013) by having links with the tropical precipitation via modulations of stratospheric ozone.

5.6 Knowledge Gaps

Aerosol-cloud-precipitation-meteorology interaction is one of the most challenging scientific issues requiring intensive observational and modeling with focused research from the climate science community. The complexity in the aerosol-cloud interaction arises from variations in dominant phase changes and microphysical and dynamical processes associated with different types of clouds. Concurrent measurements of aerosol size distribution, composition, cloud properties, microphysical parameters as well as the development of physical process scale studies based on observations over a varying space and times-scales, and translating them to climate models are essential to gain a good understanding on the role of aerosols in modifying weather and climate over India.

In addition, accurate representation of the absorbing aerosol hotspots, particularly BC and dust, is crucial to comprehend its impact on regional climate. Uncertainty in the measurement of single scattering albedo, the parameter determining the absorptive nature of aerosols, limits the correct quantification of the sign of TOA radiative forcing at regional scales. More in situ measurements of vertical

profiles of absorbing aerosols are also needed for better evaluation of model-simulated BC profiles and understanding its effect on monsoon precipitation through interaction with clouds and radiation. Also, the aerosol observational data from field campaigns and long-term monitoring sites from various sources and reanalysis products need to be gathered to make a comprehensive quality-controlled gridded product. Future field campaigns may be planned to address missing links in this regard and to reduce the uncertainty in the regional estimates of the direct and indirect effect of aerosols in state of the art GCMs.

In the case of trace gases, there is a considerable variation among the emission inventories of ozone precursors and related trace gases. Dedicated modeling and observational efforts are needed to improve ozone emission inventories over the Indian region. Model simulations show seasonal transport of chemical species over the Indian Ocean which affects the air–sea interaction and convective processes. However, the models show significant biases over the Oceans. There is a need to improve chemical processes and parameterization in the model to reduce the biases. Finally, there are limited studies quantifying the radiative impact of trace gases and associated climate change over the Indian region, and further modeling and observational studies in this direction are required.

5.7 Summary

The regional assessment of long-term in situ and remotely sensed observations over India shows a significant increase in aerosol loading over the subcontinent accompanied by robust seasonal variations. The trend in AOD is $\sim 2\% \text{ year}^{-1}$ (high confidence) during the last 30. The temporal build-up of aerosols is significantly high in the dry winter months, while changes are smaller in the pre-monsoon and monsoon season. This change has been attributed to rise in fine mode particles due to rapid growth in anthropogenic activities over the region in recent decades. CMIP5 multi-model simulations also capture the large increase in AOD over the Indian region between 1980 and 2000 with considerable bias in the three-dimensional heterogeneous distribution of different aerosol species.

There is a large seasonal as well as spatiotemporal variability in the aerosol radiative forcing. In general, the estimates of aerosol radiative forcing from measurements range from -49 to -31 W m^{-2} at the surface (high confidence), and -15 to $+8 \text{ W m}^{-2}$ at top-of-atmosphere (low confidence). The positive forcing at TOA is linked with the absorptive nature of the aerosols over the Indian region. Aerosols produce a declining trend of all-sky global irradiance over India. During 1986–1995, the observed global radiation decreased by 3.6 W m^{-2} and further by 9.5 W m^{-2}

during the decade of 1996–2005 (Soni et al. 2012). The declining trend of all-sky global irradiance over India as a whole was $0.6 \text{ W m}^{-2} \text{ year}^{-1}$ during 1971–2000 and $0.2 \text{ W m}^{-2} \text{ year}^{-1}$ during 2001–2010 (Soni et al. 2016). This decrease in global irradiance is matched with an increase in the diffused radiation over the same period indicating an increase in the aerosol levels.

Efforts were taken to understand aerosol-cloud interaction over the Indian region. The Cloud-Aerosol Interaction and Precipitation Enhancement Experiment [CAIPEEX; (Kulkarni et al. 2012)] has documented important processes associated with aerosol-cloud interaction over the Indian region. There is a significant increase in the cloud droplet number concentration with an increase in aerosols (Kulkarni et al. 2012). Very high aerosol loading causes narrowing of the droplet spectrum, collision coalescence is suppressed, and warm rain forms at an elevated layer (Konwar et al. 2012). During high aerosol loading conditions, clouds have a large amount of super-cooled liquid water ($>3 \text{ gm}^{-3}$) with the dominant mixed-phase (Prabha et al. 2012). Mixed-phase clouds contribute a significant part of monsoon clouds, which are not understood completely and need further focused process studies. Aerosols acting as CCN, and INP and their variability over the Indian region need further observations and can be used for the models or fine-tune the parameterization schemes.

Long-term observations of ozone (total column, vertical profiles, and surface measurements) and its precursors (CO , NO_x , VOCs) have been studied to estimate linear trends over the Indian region. Tropospheric ozone trends show spatiotemporal variations. Trend estimates vary with time due to changes in the emission of precursors gases. In general, ozone observations show increasing trends in the troposphere ($0.7\text{--}0.9\% \text{ year}^{-1}$ during 1979–2005) (high confidence) and decreasing trends in the stratosphere (-0.05 ± 0.04 to $-0.4 \pm 0.1\% \text{ year}^{-1}$ during 1993–2015) (medium confidence). The reported ozone observations over the Indian regions are of different time periods. However, the System of Air Quality Forecasting and Research (SAFAR) developed by the Indian Institute of Tropical Meteorology is monitoring ozone and its precursors, since 2010. These long-term observations will be helpful in obtaining future ozone trends over the India region. The CMIP5 multi-model future projections (the 2090s–2010s) over the tropics ($25^\circ \text{ S--}25^\circ \text{ N}$) show that the annual mean ozone trend is decreasing in the troposphere (except RCP8.5) and increasing in the stratosphere (Cionni et al. 2011). Seasonal trends in the troposphere and stratosphere, both, are influenced by emissions and, seasonal stratospheric intrusions, etc. The reported ozone trends have low confidence. Long-range transport processes (e.g., seasonal variations, transport between extra-tropics and tropics, stratosphere and troposphere, etc.) produce a significant variation in the loading of

tropospheric ozone leading to large changes in radiative forcing and dynamics. The model simulations show that increased tropospheric ozone since pre-industrial times has imposed ozone radiative forcing (at the tropopause) $\sim 0.2\text{--}0.4 \text{ W m}^{-2}$ over the Indian region (Chalita et al. 1996).

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