Atmospheric aerosols and their effects on radiation, clouds, and precipitation in different meteorological scenarios

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ABSTRACT

Over the last few years, aerosol-related studies have been getting the utmost attention from weather and climate scientists as these particles can influence Earth's energy balance, hydrological cycle, major circulation patterns, severe weather events, glacier melting, air quality, and public health. This paper presents a comprehensive review of the current state of knowledge on the interactions of aerosols with radiation, clouds, and precipitation. The two main pathways through which aerosols impact weather and climate are aerosol-radiation interaction (ARI) and aerosol-cloud interaction (ACI). While ARI involves changes in the radiation balance in the atmosphere and on surfaces due to aerosol scattering and absorption, ACI is associated with the effects of aerosols on cloud properties and precipitation efficiency by acting as cloud condensation and ice nuclei. A chain of complicated interactions involving various meteorological factors occurs when ARI and ACI are combined with atmospheric dynamics, impacting weather and climate. Aerosol particles can also play a substantial role in influencing extreme weather events (e.g., thunderstorms, lightning, dust storms, tropical cyclones, heatwaves, etc.) as they may modify the circulation patterns. Besides, aerosol-induced ACI and ARI may be linked with large-scale circulations through their impact on the radiation budget and generation of localized convection. The main challenges currently impeding progress in understanding the interactions between aerosols and clouds are also highlighted. One of the major issues is the lack of simultaneous measurements of cloud dynamics, microphysics, and aerosols across a broad geographical area, which hinders the ability to draw accurate conclusions from observational data. On the modeling side, there is considerable variability in cloud microphysics parameterizations, including ice nucleation, mixed-phase properties, and hydrometeor size and fall speed, leading to a significant spread in modeling outcomes. Therefore, collaborative efforts are required to advance the understanding of aerosol-cloud-climate interactions to reduce the uncertainty in measurements, providing more accurate future projections in the context of climate change.

Keywords: *Aerosols; Aerosol-Radiation Interaction; Aerosol-Cloud-Interaction; Extreme Weather Events; Large-scale circulations.*

1. Introduction

Aerosols have been at the forefront of atmospheric, environmental, and climate sciences due to their interaction with the atmosphere, cryosphere, biosphere, and ocean. By definition, aerosols are collections of tiny solid or liquid airborne particles, with the exclusion of all hydrometeors (e.g., water droplets, ice particles, graupels, snowflakes, etc.). These particles have diameters of size ranging from nanometers to micrometers (Pöschl, 2005). Their concentrations and characteristics vary significantly throughout time and space due to the wide variety of particle sources and their relatively brief atmospheric residence times of a few hours to many weeks (Kuniyal and Guleria, 2019). The types and sources of the aerosol particles are discussed in section 1.1. Also, the shape, size distribution, and chemical composition of the atmospheric aerosols are the three most important properties, which are discussed in the subsequent section 1.2.

1.1 Types and sources of aerosols

In general, atmospheric aerosols are categorized into two main types, i.e., primary and secondary. Primary types designate aerosols released directly into the air in the solid or liquid phase. These can be emitted by incomplete combustion of fossil fuels, the influence of wind friction on land or ocean surface, biomass burning, volcanic eruptions, etc. (Pöschl, 2005; Schuster et al., 2006; Pósfai & Buseck, 2010; Guleria and Kumar, 2018). Secondary aerosols are instead produced from

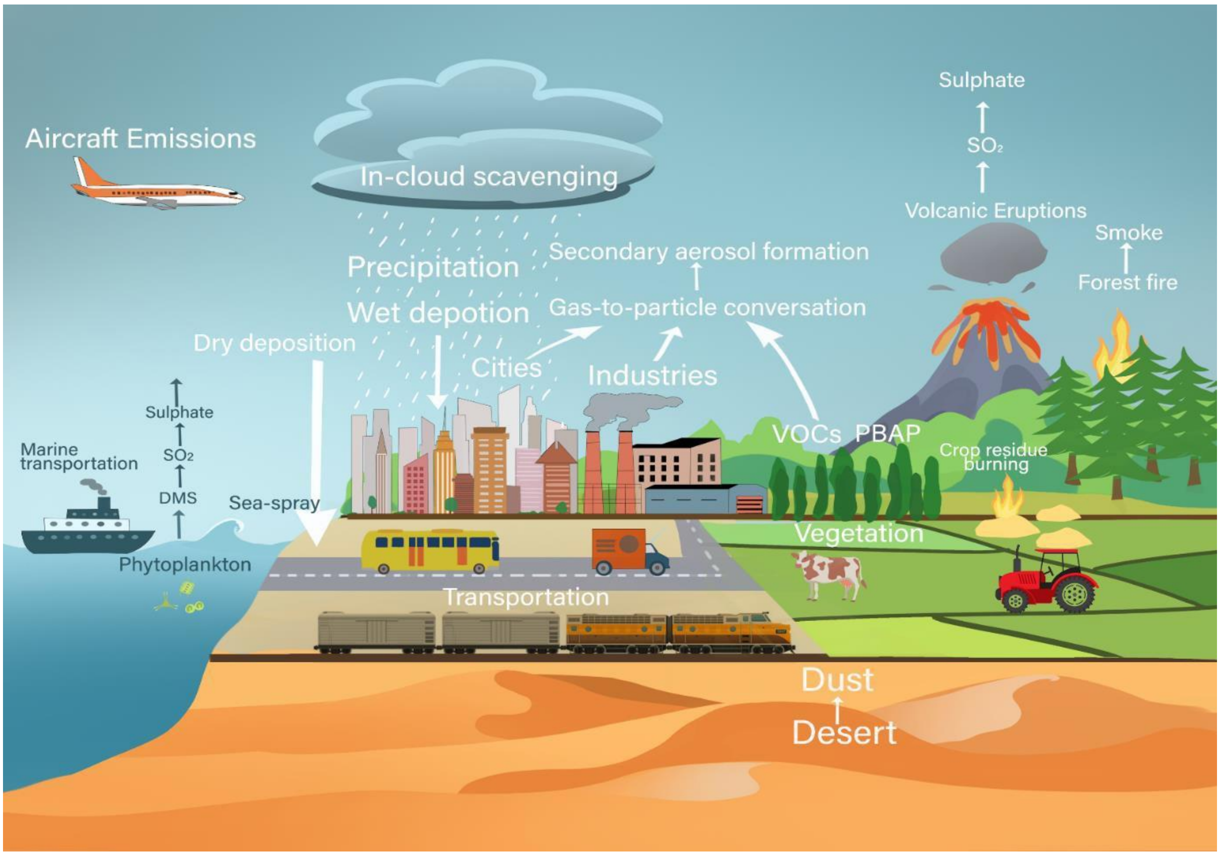

Figure 1: Schematic representation of various sources and sinks of aerosols.

various gaseous precursors via gas-to-particle conversion processes (Junge, 1963; Pósfai and Buseck, 2010; Kuniyal and Guleria, 2019). Mixtures of substances like sulfate, nitrate, or organic particles mainly comprise these types. Certain low-volatile organic compounds, like some of the hydrocarbons in automobile exhaust or condensable from biomass burning, condense onto aerosols adjacent to emitting sources but not immediately within them, creating a "gray zone" between primary and secondary emissions (Levin and Cotton, 2008). Overall, the primary aerosols (> 1 μm) are relatively larger than secondary aerosols (< 1 μm) (Eck et al., 1999; Kuniyal and Guleria, 2019). Aerosol particles can be injected into the air from various natural and anthropogenic sources. Natural sources include emissions from the desert, ocean, plants, fires, and volcanoes, whereas anthropogenic aerosols primarily come from the burning of fossil fuels (e.g., coal and oil), biofuels (e.g., wood, vegetable oils, animal waste, etc.), alternative fuels (e.g., peat), human-caused forest fires, agricultural crop-residue burning, construction activities, etc. (Chin and Kahn, 2009). The schematic diagram in Figure 1 illustrates various sources and sinks of atmospheric aerosols.

Among all aerosol types, soil or mineral dust is the most noticeable one due to its enormous mass emission flux (Kaufman et al., 2005). These aerosols come from both natural and anthropogenic sources. Natural source regions include places with a severe soil moisture deficit and relatively scarce or no vegetation (e.g., deserts, semi-arid deserts, ephemeral channels, dry lake beds, etc.). Due to the lack of soil dryness and vegetation cover, wind friction can separate soil particles and make them airborne (Zhao et al., 2006). Together with natural sources, human-induced changes in land use/land cover in the context of climate change can also dramatically worsen dust emissions (Tegen et al., 2004). In areas with relatively higher wind speeds, sea salt aerosols account for most of the primary marine aerosol concentration (Bates et al., 2005; Gantt and Meskhidze, 2013). The fundamental mechanisms of their formation are the ripping of droplets from wave crests, and the popping of rising bubbles that breaking waves inject beneath the water's surface (Schulz et al., 2004). Certain types of phytoplanktons create dimethylsulfide (DMS), a gaseous compound oxidized in the atmosphere to produce aerosols, including sulfates (Figure 1).

Volcanoes can spew bits of crushed rock and minerals (often known as volcanic ash) during violent eruptions. Volcanic ash may travel hundreds to thousands of kilometers but tends to sink fast since it comprises microscopic particles (Cole-Dai, 2010). Volcanoes also release sulfur-rich gases, such as sulfur dioxide (SO2) and hydrogen sulfide (H2S), which are oxidized in the atmosphere and turn into submicronic sulfate aerosols (Figure 1; Textor et al., 2004). These sulfur-rich gases are more effectively converted to sulfate than most anthropogenic SO2 because they are released mainly from mountain summits in the upper troposphere, which is less susceptible to dry deposition (Penner et al., 2001). In general, volcanic aerosols only stay in the atmosphere for a few weeks. However, depending on the area and injection height, volcanic aerosols have a substantially extended residence period if the eruption is strong enough to inject sulfur gases into the stratosphere (Oppenheimer et al., 2011; Dalal et al., 2023). This residence time can range from a few months to more than a year.

Primary biogenic aerosol particles (PBAP) such as spores, pollen, viruses, fungi, bacteria, plant, and insect waste mainly come from the terrestrial biosphere (Després et al., 2007). Depending on their size, the wind can carry these particles for varied distances once in the air. There are several ways that fungi expel their spores, including high-speed catapult-like discharge, and as this release frequently occurs at night, there is significant diurnal fluctuation (Gilbert, 2005; Elbert et al., 2007). Some insects create brochosomes, spherical particles of only a few hundred nm in size that are regularly seen in the atmosphere (Bigg, 2003). Moreover, certain pollen species are forcibly expelled, often during the day (Edwards et al., 2005). Some PBAPs contain a variety of particles made of microorganisms and exopolymer secretions in diverse combinations created at the sea surface (Leck and Bigg, 2005). Certain plants and algae release volatile organic compounds (VOCs) that undergo atmospheric oxidation, condense, and enrich the aerosols in the atmosphere with organic material (Figure 1). These are known as secondary biogenic aerosols.

Burning of biomass, which includes household biofuel usage and open vegetation (including savannas, woods, and agricultural waste) fires, is also one of the main contributors to the global upsurge in aerosol particles (Figure 1; Sarkar et al., 2022). And the particles injected from biomass burning are called pyrogenic aerosols (e.g., carbonaceous aerosols). The combustion of fossil fuels in both stationary and moving sources (e.g., automobiles, thermal power plants, industries, domestic cooking, etc.) also produces a variety of aerosols depending on the fuel burnt and the kind of combustion technology applied (Lighty et al., 2000). And the most common ultrafine primary aerosols from burning oil products are also carbonaceous aerosols (Bond et al., 2004). Carbonaceous aerosols from burning fossil fuels and burning biomass are projected to decrease over the 21st century throughout the globe, although regional increases are anticipated, notably in Asia (Streets et al., 2004). Also, coal burning creates many coarse mode ash particles primarily controlled by stack technology, particularly in industrialized nations (Andreae and Rosenfeld, 2008). Besides, secondary aerosols can also be formed from various gaseous pollutants (e.g., sulfur oxides, nitrogen oxides, hydrocarbons, etc.). Internally mixed particles with some soluble materials are produced by condensing primary and secondary aerosols, and H2SO4 from the oxidization species.

1.2. Size distribution and chemical compositions

Both observation and modeling studies suggest that the dust mass distribution around the sources, peaks at ten μm or greater diameters (Andreae and Rosenfeld, 2008). These particles may contain clays (e.g., kaolinite and illite), carbonates (e.g., calcite and dolomite), iron oxides (e.g., hematite and goethite), or quartz (Twohy et al., 2009). Whether dust aerosols could act as CCN (cloud condensation nuclei) or IN (ice nuclei) primarily depends upon their chemical composition and size distribution. They could hardly behave as CCN at commonly observed supersaturations if they are composed of insoluble silicate or clay minerals. However, many of these particles would persist as an interstitial aerosol in deep convective clouds and ascend to the middle and upper troposphere, where they may function as an effective IN (DeMott et al., 2003). Compared to the uncoated ones that prefer to stay interstitial, those coated with soluble chemicals (e.g., chloride, sulfate, nitrate, etc.) would be among the first to be activated and function as giant CCN (GCCN) due to their large size (Kelly et al., 2007). Nevertheless, there is a possibility that dust particles may already include soluble components at the moment of emission or that they may acquire soluble salt coatings during atmospheric processing, particularly in clouds (Yin et al., 2002; Kelly et al., 2007).

The term ‘sea spray’ may be preferable to ‘sea salt’ as these particles could also include biological matter and other organic contaminants (Andreae and Rosenfeld, 2008). Organic material includes both water-soluble and insoluble components such as biological waste, microorganisms, etc. (Middlebrooks et al., 1998; O’Dowd et al., 2004; Kaku et al., 2006). The proportion of organic components rises with the reduction in particle size and, in bioactive locations, may reach 90% in the size fraction of about 100 nm (Cavalli et al., 2004; Nilsson et al., 2007). The size of these aerosols usually ranges from 100 nm to several tens of µm. These particles may significantly increase the concentration of primary marine CCN above the amount that sea salt particles can account for (Lohmann and Leck, 2005). Since most of them are in the coarse mode, these particles might be crucial for GCCN, even though they are few in number (Andreae and Rosenfeld, 2008).

PBAP spans a wide range from a few tens of nanometers to tens of microns due to various sources (Andreae and Rosenfeld, 2008). Pollen, spores, and large bacteria typically lie in the range of 1–100 µm, whereas smaller bacteria and viruses are often less than 1 µm. Debris is typically greater than 100 µm. Particles in rainforest areas are made up mostly of organic material, although many lack or have very few identifiable morphological or bulk chemical fingerprints (Andreae and Crutzen, 1997). Their size, soluble matter content, and surface characteristics determine whether or not they may serve as an IN or a CCN (Bauer et al., 2003). Due to their vast surface area and low water contact angles, they can already be activated at extremely low supersaturations and function as GCCN (Sharma and Rao, 2002). Because of this, they can contribute considerably to the development of precipitation in situations when significant levels of the pollutant CCN would typically prevent the occurrence of warm rain (Rosenfeld et al., 2002).

Pyrogenic aerosols are primarily composed of carbonaceous materials like black carbon (or ‘soot’) and organic carbon, with a small amount of other inorganic components (Reid et al., 2005; Sarkar et al., 2022). The inorganic element consists of certain soluble salts (e.g., potassium, ammonium, sulfate, nitrate), some insoluble dust particles, and ashes. Around half of the organic fraction is water soluble, with dehydrated sugars or cellulose breakdown products such as levoglucosan (Graham et al., 2002). Due to their chemical composition and the predominant internal mixing, most pyrogenic particles contain a sizable portion of soluble (organic and inorganic) substances (Li et al., 2003). As a result, they are much easier to activate as CCN since they age, grow larger, and contain more soluble material (Andreae et al., 2004). These aerosols also frequently exhibit a separate coarse mass mode with a peak around 4 µm diameter (Radke et al., 1991). Nevertheless, it is unlikely that such a mode will substantially impact cloud microphysics. However, the function of soot as IN is likely restricted to cirrus generation by homogenous freezing in the presence of a few productive dust IN (Kärcher et al. 2007).

Ant emission flux from fossil fuel combustion, are the most significant ones. They contain a combination of hydrophobic, water-insoluble materials (such as gasoline, combustible petroleum compounds, and soot) and soluble inorganic and organic elements, especially H2SO4 produced from the oxidized sulfur in the fuels (Hudson, 1991; Zuberi et al., 2005). There is a fair amount of debate on the hydrophobicity of ‘fresh soot’ since the size and composition of these particles can have quite diverse CCN characteristics. While some studies reveal that pure soot is hydrophobic, some found that fresh gasoline soot particles can act as effective CCN (Hudson, 1991; Hudson et al., 2000; Zuberi et al., 2005). It seems that highly pure hydrocarbon soot emitted from burning CH_4, hexane, or aviation fuel, is initially somewhat hydrophobic (Zuberi et al., 2005; Mikhailov et al., 2006). Contrarily, the soot particles released by diesel engines and most of the other combustion sources already include a negligible amount (between 3 to 15%) of inorganic or organic soluble materials, and the soluble percentage increases if fuels contain more contaminants, like sulfur (Kittelson, 1998; Sakurai et al., 2003). This additional sulfate could enhance the CCN activity in the events like Kuwait oil smoke plume (Hudson and Clarke, 1992). Diesel engines may contribute more to CCN since their particles are often larger and more sulfur-rich than gasoline engine particles (Zielinska et al., 2004).

While some modern gasoline engines release particles predominantly hydrophobic organic products low in sulfur and other water-soluble components, the engines employing leaded gas generate highly soluble particles mainly consisting of chloride and lead bromide. However, the absence of lead compounds has significantly reduced the CCN capability of gasoline engine particles. Another factor in the initial weak CCN activity is the small size of these aerosols. For instance, the usual number modal diameters of the particles released by petrol engines under high-speed driving circumstances are 60–120 nm and 40–80 nm, respectively (Weingartner et al.,1997; Andreae and Rosenfeld, 2008). While these sulfuric acid-based particles are too small to contribute directly to the generation of CCN, they can add soluble content to other particles, such as soot, or grow large enough to function as CCN after a few coagulation stages (Giechaskiel et al., 2005). For example, Lammel and Novakov (1995) demonstrated that just 10% of soluble matter is needed to produce soot particles with equivalent CCN activity to ammonium sulfate particles of a similar size. Besides, their CCN activity is further influenced by additional factors, such as surface coating of soluble material and microcapillary gaps. And CCN efficiency improves even more as it ages due to the condensational absorption of soluble compounds, coagulation with soluble particles (such as sulfates), and chemical interactions (with OH, O_3, etc.) at the surface (Schneider et al., 2005).

2. Aerosol-Radiation Interactions

The aerosol radiative effect through absorption or scattering of solar radiation, referred to as aerosol-radiation interaction (ARI), is an important pathway to influence atmospheric stability, hydrological cycle, cloud development, deep convection, and sometimes even large-scale circulation (Ramanathan et al., 2005; Andrews et al., 2006; Bollasina et al. 2011; Stocker, 2014; Vinoj et al., 2014; Sanap and Pandithurai 2015). ARI can alter the planetary albedo, which further causes climate forcing (Twomey, 1977). The main factors that govern the radiative properties of aerosols are their concentration, size, shape, and chemical composition (Andrews et al., 2006; Satheesh, 2012). Due to various origins, these factors are quite variable in view of spatiotemporal scale (Pöschl, 2005). ARI might happen in several ways, and these interactions are one of the leading causes of uncertainty in climate modeling and prediction (Rosenfeld et al., 2014). The variety of pathways through which aerosols and radiation interact (Figure 2) are described in the following subsections.

2.1. Direct effect

The primary energy source that controls the Earth's climate is the sun, and the solar energy entering the atmosphere at the top, travels through the atmosphere before arriving at the surface. Thus, not all the incoming shortwave radiation reaching the top of the atmosphere makes it to the surface. A substantial portion is scattered back to space or absorbed by atmospheric aerosols (Kant et al., 2017; Sarkar et al., 2022). As these interactions directly influence the Earth's radiative budget, this process is termed as the 'direct effect (DE)' of aerosols (Figure 2). The amount of scattering and absorption of sunlight is determined by the physicochemical properties of aerosols (Kuniyal and Guleria, 2019). BC is thought to be the second-most significant contributor to global warming after CO_2 due to its highly absorbing properties

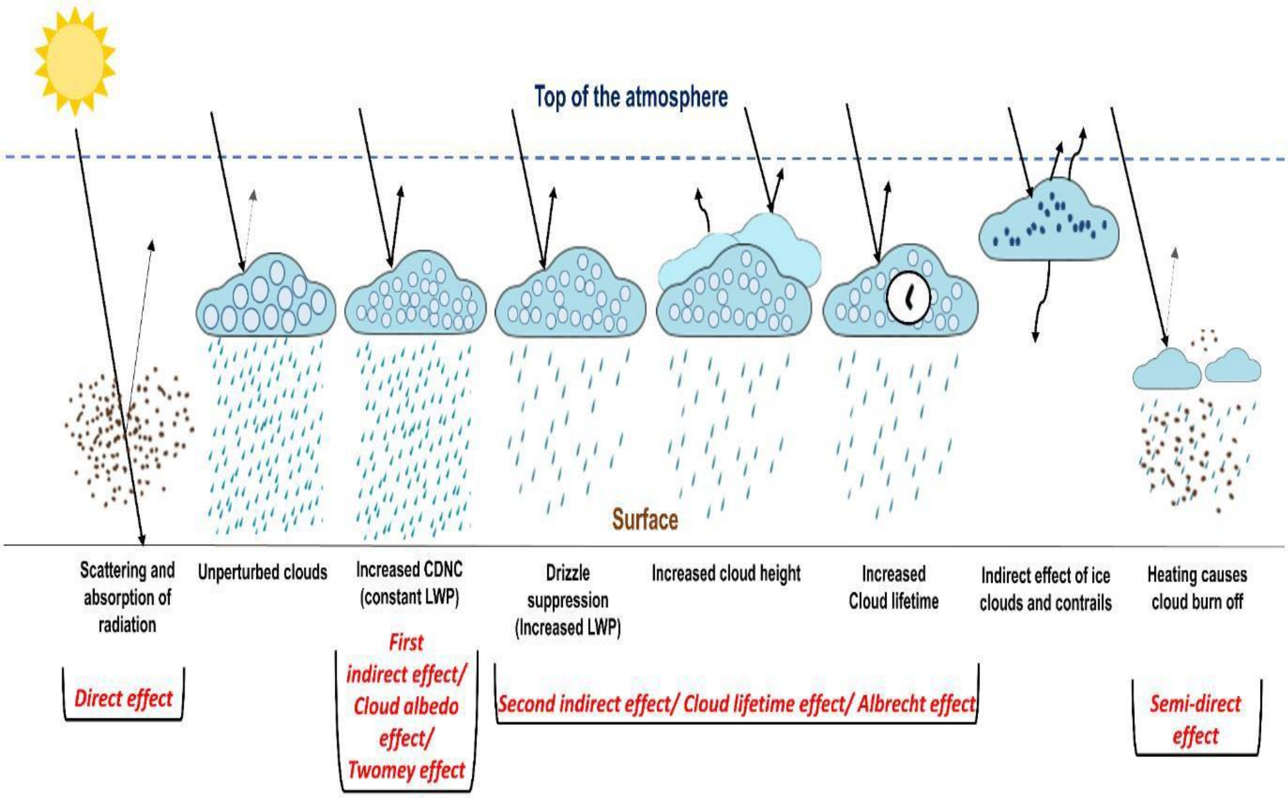

Figure 2: Schematic diagram of various effects of aerosols on radiation, clouds, and precipitation. This figure is redrawn after adopting from IPCC (2013).

(Jacobson, 2000). While BC plays a dominant role in absorbing solar radiation, sulfate is most efficient in scattering among all other aerosol types (Charlson et al., 1992). When an aerosol mostly scatters solar radiation, it has a cooling impact and a warming effect when it absorbs (Sarkar et al., 2022).

Often, aerosol particles in the mixing state predominate in the atmosphere compared to those in the scattering and absorbing states. As a result, the net impact of aerosol mixing state on the Earth's radiation budget depends on the surface and cloud properties (Pöschl, 2005). While the same aerosols over a dark surface can have a cooling effect, their influence on high-albedo surfaces at high latitudes may have a net warming effect (Benedetti and Vitart, 2018). This could be especially true for absorbing aerosol types like BC (Satheesh et al., 2008). In cloud-free areas, there may be significant direct effects on factors like surface temperature, precipitation, and circulation. On the climate change time scale, both absorbing and non-absorbing aerosols prevent solar radiation from reaching the surface, resulting in global dimming, which causes the cooling of the Earth's surface (Ramanathan et al. 2005). This further slows down the hydrological cycle in the tropics and eventually weakens the Asian monsoon. However, atmospheric heating due to strong absorption by aerosols like BC and dust can produce a water cycle feedback through the 'Elevated Heat Pump (EHP) effect', strengthening the Asian monsoon against the solar dimming effect (Lau et al., 2006).

2.2. Semi-direct effect

When absorbing aerosols are embedded in or around a cloud layer, they can reduce the relative humidity of the layer and promote the evaporation of cloud droplets due to their strong absorption of solar radiation. This eventually decreases cloud cover and liquid water path (LWP), resulting in the positive radiative forcing on the climate. This process is called the 'semi-direct effect' or SDE (Figure 2; Hansen et al., 1997; Kaufman and Koren, 2006). Some general-circulation model-based studies have shown that the semi-direct effect is a positive feedback that may compete with the indirect effect by significantly increasing the total direct effect of absorbing aerosols (Hansen et al., 1997; Cook and Highwood, 2004). However, Penner et al. (2003) suggested that the semi-direct effect may be positive or negative depending on the vertical position of the absorbing aerosols. For instance, if absorbing aerosols reside above the low-level cloud deck, they may reinforce the temperature inversion, stabilize the free troposphere, and reduce the entrainment of dry air to the cloud top, leading to the thickening of low-level clouds and a corresponding cooling of the climate (Wilcox, 2010; Koch and Del Genio, 2010; Sarkar et al., 2022). Besides, absorbing aerosols-induced surface cooling can prevent PBL (planetary boundary layer) from further development, which causes the trapping of particles near the surface, resulting in a haze-like scenario (e.g. Panda et al. 2009) through an aerosol-radiation-PBL feedback loop. This is often called the 'dome effect' as it resembles a dome from a distance (Ding et al., 2016). In addition, if a simultaneous rise in low-level cloud fraction happens, it can further cause a decline in radiation reaching the surface and create another positive feedback termed the 'aerosol-cloud-PBL' loop (Qu et al., 2018; Sarkar et al., 2022).

While the stabilizing impact of absorbing aerosols above can increase boundary-layer clouds (such as stratocumulus clouds), they may also decrease surface evaporation and moisture availability, restricting the development of cumulus clouds (Koren et al., 2004; Fan et al., 2008). However, in some land regions, lofted absorbing aerosols may strengthen upper-level convection and encourage low-level convergence that can draw moisture from nearby oceans to the cloud base, causing an increase in cloud cover (Koch and Del Genio, 2010). In addition, some modeling studies reported a decline in upper-level clouds, primarily due to lower upper-level humidity or cloud burnoff by the lofted biomass-burning aerosols, resulting in negative cloud feedback (Penner et al., 2003). Contrarily, if absorbing aerosols are suspended below cloud layers, the additional heating can accelerate upward motions, causing an increase in cloud cover and LWP (Feingold et al., 2005). Overall, the semi-direct effect may be important on local scales since the local climatic circumstances SDE.

2.3. Snow darkening effect

Another way aerosols can influence the Earth's climate is through the snow darkening effect (SNDE), which results from the reduction of albedo of the snow surfaces due to the strong absorption of solar energy caused by the deposited absorbing aerosols, such as BC and dust (Nair et al., 2013; Qian et al., 2015; Das et al., 2022). This mechanism is the outcome of a series of positive feedback processes (Qian et al., 2015). In the beginning, when snow begins to melt, the total number of absorbing aerosols (AAs) in the snow rises as some are collected at the snowpack's surface rather than being carried away by melt water (Flanner et al., 2007). As a result of the increased heating caused by the rise in AAs, the snowpack's effective grain size also increases, reducing snow albedo further (Hadley and Kirchstetter, 2012). However, at greater number concentrations, grain sizes can decrease because of the mass loss from the rapid melting of the topmost layers (Painter et al., 2013). As this process continues, enough snow melts to reveal the darker surface beneath, causing more warmth and snow removal from the glacier, known as 'snow-albedo feedback' (Flanner et al., 2007; Qian et al., 2015). Consequently, this early snow cover removal causes surface warming, further affecting hydrological cycles, regional circulations, and ecosystems (Qian et al., 2011). IPCC (2013) reported that this aerosol-induced adjustment of the snow-albedo feedback, which is highly uncertain, is one of the main forcing mechanisms impacting climate change. Modeling studies also indicated that this mechanism has even higher potency in warming and melting snow than greenhouse gases (Flanner et al., 2007; Skiles et al., 2012).

Previous studies on the radiative impacts of aerosol-induced SNDE primarily focused on the effects of BC (Rahimi et al., 2019; Usha et al., 2021). However, dust particles can play a substantial role in SNDE too. For instance, Sarangi et al. (2019) illustrated that the effect of dust-induced snow darkening is stronger than that of black carbon at elevations of 4000 m in mountainous areas of Asia.

During springtime, prevalent westerlies are linked to the high dust loading over Tibetan Plateau, favoring the long-range transport of dust from dry and semi-arid areas of North Africa, Central and East Asia, and the Middle East to the Himalayas (Nair et al., 2013; Mao et al., 2019). The deposition of AAs on snow throughout the Himalayan-Tibetan Plateau (HTP) area can cause a significant surface warming of 1-4 °K, reducing snow cover by 8% and the number of snow cover days by 20 days across the area (Usha et al., 2022). Consequently, this faster snow melting can cause a considerable rise in river runoff over the HTP region (Usha et al., 2020). Besides, increased deposition of BC on the Greenland ice sheet can be expected in future with the significant increase in wildfires over the northern hemisphere due to rising temperatures (Soja et al., 2007; Thomas et al., 2017).

3. Aerosol-Cloud Interactions

Aerosols can also alter the thermodynamics and macro/microstructure of clouds by serving as a CCN or IN, primarily determined by particle size distribution and chemical composition, as discussed in section 1.2. The influence of aerosols further alters cloud radiative response and precipitation efficiency, significantly impacting the global climate. IPCC (2021) termed these indirect effects (Figure 2), as a whole, 'aerosol-cloud interaction (ACI)'. ACI is considered one of the most important aspects of the hydrological cycle, circulation patterns, and global climate (Fan et al., 2016). Moreover, ACI can be further complicated by several dynamic and thermodynamic factors like chemical and physical aerosol characteristics, cloud microphysics, moisture content, updraft motion, atmospheric stability, etc. (Zhang et al., 2015). Thus, how aerosols alter cloud characteristics and precipitation through ACI significantly varies in the different cloud types primarily governed by these dynamics and thermodynamic factors. For instance, aerosol effects on deep convective clouds are far more intricate and poorly understood than shallow clouds due to the inherent complexity in terms of dynamics, thermodynamics, and microphysics (Fan et al., 2016). Thus, the indirect effect of aerosols on clouds is probably the highest of all uncertainties regarding global climate forcing (Tao et al., 2012; Rosenfeld et al., 2014). The total radiative forcing of ACI is reported as -0.45 Wm- 2 with a margin of error of -1.3 to 0 Wm-2 (IPCC, 2021). The mechanisms involved discussed in the following subsections.

3.1. First aerosol indirect effect

An increase in aerosol concentration causes a rise in the number of CCN, which in turn promotes an increase in cloud droplet number concentration. For fixed cloud liquid water content, this leads to a decrease in the size of cloud droplets and an increase in cloud reflectivity. Overall, this process contributes to the cooling of the Earth's climate. This aerosol effect is the 'first aerosol indirect effect' (FAIE) or 'cloud albedo effect', and is sometimes referred as the 'Twomey effect' (Figure 2; Twomey, 1977; Seinfeld et al., 2016; Kant et al., 2017, 2019b; Sarkar et al., 2021). According to Yuan et al. (2008), cloud droplet size and distribution are smaller and narrower in the presence of high CCN concentration (dirty environment) and larger and broader in the case of low CCN (clean environment). However, some studies also reported increased cloud droplet effective radius with the rising aerosol concentrations (Feingold et al., 2001; Liu et al., 2017; Sarkar et al., 2022). This growth of droplet size only happens when aerosol concentration approaches a specific threshold value or 'tipping point', suggesting that ACI may subsequently achieve saturation. This could further decrease cloud albedo, resulting in a negative relationship between aerosol concentration and cloud optical depth (Alam et al., 2014). This contrasting aerosol influence is often known as 'anti-Twomey effect'. This may be attributed to the excessive amount of absorbing aerosols in the atmosphere, which causes cloud droplets to vaporize (Tiwari et al., 2022).

The anti-Twomey effect appears to be more pronounced with larger particles or giant CCN (Kant et al., 2019a; Liu et al., 2020; Jose et al., 2021; Sarkar et al., 2022). According to Feingold et al. (2001), when numerous larger particles are present in the atmosphere, it causes massive competition between the smaller and larger ones for the available moisture. While smaller droplets tend to evaporate, larger ones persist in continual growth after activation. Therefore, this process also inhibits water vapor from condensing over the tiny particles, moving the droplet size distribution to the larger ones. Recently, Khatri et al. (2022) reported both negative and positive correlations between cloud droplet effective radius and aerosol in polluted conditions. Several observation-based studies using various platforms like aircraft, ship-racks, satellites, and ground-based instruments estimated FAIE and found that the values can fluctuate significantly with respect to time, place, aerosol characteristics, and cloud types (Radke et al., 1989; Feingold, 2003; Lu et al., 2007; Quaas et al., 2009; Jung et al., 2013; Wang et al., 2014). Also, various meteorological parameters, such as precipitable water vapor, vertical wind shear, and lower tropospheric stability may contribute to the more significant variation of FAIE (Kim et al., 2003; Yuan et al., 2008; Wang et al., 2014; Qiu et al., 2017).

3.2. Second aerosol indirect effect

High aerosol loading can also impact the evolution of clouds by altering microphysical properties due to the reduction in particle sizes. The narrower droplet spectrum results in the weakening of the collision and coalescence processes, which lead to the suppression of warm-rain processes and a consequent increase in cloud lifetime, liquid water content, and cloud fraction. These longer-lived clouds may offset the greenhouse effect by providing an additional negative climate forcing, significantly impacting the global climate change rate. This process is collectively called the ‘second aerosol indirect effect’ (SAIE) or ‘cloud lifetime effect’ (Figure 2; Albrecht, 1989; Ackerman et al., 2000; Seinfeld et al., 2016). However, both observational and modelling studies sometimes agree and at times disagree with this hypothesis (Ackerman et al., 2004; Small et al., 2009). Some observation-based studies suggested that air pollution in urban and industrialized areas could lower cloud particle size and inhibit precipitation (Rosenfeld, 2000; Sekiguchi et al., 2003). However, increasing CCN concentrations can also result in thin cloud layers by reducing the sedimentation rate, which is most pronounced when cloud base heights exceed 400 m (Wood, 2007). The struggle between moistening cloud layers by suppressing precipitation and drying them by enhancing the entrainment of overlying air determines changes in warm cloud macro and microphysical characteristics caused by aerosols (Eastman and Wood, 2018). Gryspeerdt et al. (2019) showed that aerosol impact on cloud LWP alterations dramatically relies on the local relative humidity. In a nutshell, two main differing factors control how aerosols influence the LWP. The first is increased entrainment and evaporation, whose combined effects decrease the LWP; the second is coalescence rate inhibition, which helps enhancing the LWP and suppressing the precipitation (Lebsock et al., 2008).

Saleeby et al. (2015) further indicated that rising CCN can decrease the cloud fraction of shallow cumulus clouds. Also, smaller cloud droplets and higher droplet number concentrations can facilitate cloud transition from open to closed cells, which is mainly prominent in the ship tracks (Goren and Rosenfeld, 2012). Open-cell clouds, the partially covered clouds of little concentrations of large droplets, show a significant upsurge in LWP, cloud optical depth, and precipitation in response to a slight aerosol increase (Fan et al., 2016). If aerosols continue to rise, they can eventually suppress precipitation and enhance the formation of closed-cell clouds (e.g., stratocumulus clouds). These closed-cell clouds hardly induce heavy rainfall since they comprise numerous tiny droplets. Adding more particles to closed cells further inhibits precipitation by reducing droplet size and strengthening entrainment, owing to intense evaporation (Ackerman et al. 2004). Thus, cloud characteristics and the stratocumulus clouds may considerably influence the direction and amplitude of the precipitation response (Christensen and Stephens, 2012). Establishing causal linkages between aerosols and cloud albedo caused by precipitation suppression has been difficult because the sequence of events linking SAIE microphysical and dynamical mechanisms is still unclear. This uncertainty results, due to our inability to separate the effects of meteorology from the aerosol impact on cloud microphysics (Rosenfeld et al., 2019). Therefore, constraining the SAIE in climate models is more challenging than the FAIE.

3.3. Aerosol invigoration effect

The delay in the warm-rain processes due to SAIE can also contemporarily enhance droplet mobility under convective conditions, thereby lifting numerous smaller droplets higher in the atmosphere where the mixed and cold phase processes occur. Once the freezing level is attained, ice production activities start within the rising cloud. And during the freezing of these smaller droplets, additional latent heat is released into comparatively colder surroundings, further strengthening the updraft, and forming deeper and wider convective clouds. This phenomenon involving the microphysical-dynamical coupling is known as the aerosol invigoration effect or AIvE (Rosenfeld and Woodley, 2000; Williams et al., 2002; Koren et al., 2005; Fan et al., 2009; Altaratz et al., 2014; Sarangi et al., 2018). This further implies that the clouds in a polluted environment will likely grow higher and become stronger thunderstorms than they would under cleaner conditions (Tao et al., 2012). This aerosol effect is primarily evident when there is less wind shear and when clouds have warmer bases (Rosenfeld et al., 2014). Contrarily, thermodynamic invigoration is rare for clouds with cold cloud bases under strong wind shear or dry conditions (Khain et al., 2005; Fan et al., 2009; Lebo et al., 2012).

The major factors influencing the aerosol effects on convective clouds are relative humidity, convective available potential energy, and wind shear, since these variables control the main microphysical processes and the microphysics-dynamics feedback mechanisms involved in deep convection (Khain et al. 2008; Fan et al. 2009; Tao et al. 2012). Besides, different types of convective systems could be significantly influenced by aerosols through the convective clouds formed during those events. For instance, aerosols may alter the size and strength of gust fronts, which would change the organization of the cloud system and influence precipitation and macrophysical cloud characteristics (Lebo and Morrison, 2014). On the other hand, supercell systems heavily influenced by dynamic pressure perturbations are less susceptible to aerosols (Morrison, 2012). Nevertheless, under different circumstances, increased aerosols may cause larger hydrometeors to form, reduce evaporative cooling, and weaken gust fronts, which keeps updrafts unaltered and stops supercells and powerful convective storms from dissipating (Rosenfeld and Bell 2011). Because there are so many heterogeneous ice crystal nucleation processes, it is also difficult to understand how aerosol particles might alter the IN characteristics. Aerosols can serve as IN by contact freezing, immersion, condensation freezing, or deposition nuclei (Lohmann and Feichter, 2005). While immersion freezing may be more frequent at lower temperatures, contact nucleation is often the most efficient way for ice nucleation at modest supercooling. Borys et al. (2003) reported that the rimming and snowfall rates are lower in orographic clouds for a given supercooled liquid water content when the supercooled cloud includes more droplets, as would be in the case of polluted conditions. Sherwood (2002) indicated that biomass-burning aerosols might cause a reduction in ice crystal size in deep convective clouds. Fan et al. (2013) identified a microphysical invigoration caused by decreased ice particle size and fall velocity, which can be an alternative cloud invigoration process. This mechanism could explain the enhanced cloud-top height and cloud cover observed in other studies (Andreae et al., 2004; Li et al., 2011; Niu and Li, 2011). Moreover, AIvE is less understood than any other ACIs due to the complexity involved in thermodynamics, dynamics, and microphysics associated with deep convection.

4. Impact of aerosols during extreme weather events

Extreme weather events are unusually severe weather conditions primarily governed by atmospheric thermodynamic and dynamic factors, which can cause substantial damage to agriculture, society, and natural ecosystems (Wang et al., 2022). Thunderstorms, lightning, dust storms, tropical cyclones, heat waves, etc., are just a few examples of these extreme events that frequently occur in a time frame of minutes to days. These disastrous events have received a lot of attention and have become an active topic for atmospheric research because of their catastrophic consequences (Duffy and Tebaldi, 2012; Mueller and Seneviratne, 2012). These extreme meteorological events are strongly influenced by regional and global circulation, and aerosols have been identified as one of the crucial modulators of these circulation patterns (Li et al., 2017). Due to extensive emissions, several places worldwide receive substantial aerosol loadings. Recently several studies further reported that human activities, mainly anthropogenic pollution, could be responsible for the recent increase in the frequency of these extreme weather events (Christidis et al., 2011; Doocy et al., 2013). Since storms are sensitive to even the slightest alteration in atmospheric thermodynamics and dynamics, other components must be fixed to distinguish the impacts of aerosols (Li et al., 2017). However, the role of atmospheric aerosols in developing severe weather events has not been thoroughly established due to the intricacy of atmospheric chemical and physical interactions. Nonetheless, a brief review is presented in this section to highlight the possible role of aerosols during the extreme weather events.

4.1. Thunderstorms and lightning

Thunderstorm events are powerful short-lived weather-related disturbances marked by cumulonimbus clouds that usually produce lightning, thunder, torrential downpours, hailstones, and highly turbulent surface winds (Araghi et al., 2016). Several modeling studies that investigated the aerosol effects on thunderstorm occurrences in polluted areas found evidence of inhibited collision and coalescence (Fan et al., 2007; Storer et al., 2010), delayed rainfall (Tao et al., 2007; Mansell and Ziegler, 2013), diminished cold pool size (Lerach and Cotton, 2012), and elevated updraft speeds owing to the increased latent heat release (Fan et al. 2007; Mansell and Ziegler 2013). Nevertheless, it is unclear whether the surface precipitation increases or decreases in polluted scenarios, with noticeable sensitivity to lower-level (0–3 km) relative humidity (Khain et al., 2008; Fan et al., 2009). Usually, higher concentrations of hygroscopic aerosol can intensify thunderstorms through ACI in a humid and convectively unstable atmosphere (Andreae et al., 2004; Khain et al.,2005; Zhang et al., 2007; Rosenfeld et al., 2008; Yang and Li, 2014). However, in a dry and polluted environment, absorbing aerosols through ARI may significantly decline atmospheric instability, suppressing thunderstorm activities (Fan et al., 2008; Li et al., 2016; Guo et al., 2016; Yang et al., 2016). Besides, depending on a particular storm's microphysical and dynamic processes, aerosol-induced change in CCN properties can also modify the hail amount and the size of the hailstones (Noppel et al., 2010).

Cloud electrification or lightning occurs when graupel and ice crystals collide with supercooled liquid water droplets in intense updrafts. Adding aerosols to clouds increases the quantity of supercooled water and invigorates the clouds, strengthening updrafts and increasing cloud electrification processes (Rosenfeld et al., 2008). Therefore, the effect of aerosols and the thermodynamic characteristics of clouds (e.g., increased instability, closer proximity of the cloud base to the freezing level, etc.) are equally important in understanding the variation in lightning frequency (Stolz et al., 2015). The difference in aerosol concentrations between land and sea may contribute to the increased frequency of lightning activity over land than the ocean (Seinfeld and Pandis, 2006). Many studies indicated a rise in lightning activities and intensified thunderstorms in highly polluted areas (Andreae et al., 2004; Sherwood et al., 2006; Wang et al., 2011; Yang and Li, 2014). However, the heat island effect may also be responsible for increased lightning activities above big cities due to altered thermodynamic conditions (Farias et al., 2009; Lal and Pawar, 2010). Yuan et al. (2011) found that the increased aerosol loading followed by volcanic eruptions boosted the lightning activity in the Philippines by modifying the cloud microphysics. Depending on the aerosol concentrations, the rate of lightning flashes strengthening can often reach as high as 30 times (Yuan et al., 2012). Besides, aerosols alter not only the frequency of lightning flashes but also the polarity of charge production processes within convective clouds. For instance, smoke aerosols from forest fires caused higher peak currents and enhanced cloud-to-ground flashes over various regions (Murray et al., 2000; Andreae et al., 2004; Rosenfeld et al., 2007). However, many studies also indicated that high aerosol concentrations could be unfavorable too for strengthening lightning activities (Williams et al., 2002; Smith et al., 2003; Altaratz et al., 2010). Since electrical properties are also connected with other meteorological factors, it is challenging to establish the link between the aerosols and lightning activities using only observations. Therefore, there is reason to question observational studies that claim aerosols increase lightning. Hence, more numerical modeling studies like Wang et al. (2011) and Mansell and Ziegler (2013) are urged, which could be able to separate well the non-linear relationship between aerosols and prevailing meteorological conditions.

4.2. Dust storms

Dust or sand storms in arid and semi-arid regions, are regarded as the occurrences when blowing dust causes visibility down to one kilometer or less, rendering some locations uninhabitable and endangering human health (Shao et al., 2011). Deep convection caused by intense surface heating can create powerful and turbulent winds that lift massive amounts of mineral dust particles into the free troposphere and carry them several thousand kilometers far from the source region (Engelstaedter et al., 2006; Zoljoodi et al., 2013). The Northern Hemisphere is home to most of the world's major dust storms, notably in the vast arid area known as the 'Dust Belt', stretching from the Sahara, the Middle East, and Central Asia to northwest India, Mongolia, and China (Middleton, 2019). Every year, 5–10 dust storms are observed across northwest India, and more than 50% occur in the pre-monsoon season (Goudie and Middleton, 2000). Two distinct dust storms are identified over this region, viz., 'Loo' caused by strong pressure gradients and 'Andhi' triggered by powerful downdrafts from thunderstorms (Middleton, 1989). They are mainly observed as advancing dust walls associated with cold pool outflows, marked by a sharp decrease in temperature and visibility, a potential change in wind direction, and a rise in moisture content and wind speed (Joseph et al., 1980). Like the 'Andhi' type, dust storms are called 'Haboobs' across the Middle East and Africa (Roberts and Knippertz, 2012). These are brought on by gust fronts that develop from thunderstorm downdrafts, which can produce localized dust accumulations by creating cold pools alongside divergent surface airflow (Anisimov et al., 2018). Even without surface precipitation, a desert's dry sub-cloud environment typically encourages downdrafts to reach the ground and produce gust fronts (Knippertz et al., 2009). Shepherd et al. (2016) demonstrated that there are three categories of factors that influence dust storm intensity, i.e., climate-related (viz., wind speed, turbulence, air temperature, pressure, etc.), particle properties (i.e., type, size, moisture contents, etc.), and land surface characteristics (i.e., vegetation density, the roughness of the terrain surface, etc.).

Each year, dry and semi-arid areas release 1000 and 3000 Tg of dust into the atmosphere (Huang et al., 2010). The most significant contribution to global dust emissions comes from the deserts of North Africa (50–70%) and Asia (10–25%), respectively (Tegen and Schepanski, 2009). These dust aerosols spend varying amounts of time in the atmosphere before being scavenged by wet deposition (mainly fine-mode aerosols from the long-range transport) and dry deposition (primarily coarse-mode aerosols) depending on their size, altitude, and prevailing weather conditions (Seinfeld and Pandis, 2016). The airborne sand and dust particles released during dust storms affect the atmospheric temperature profiles, the Earth's radiative balance, and cloud microphysical characteristics by acting as a CCN and IN (DeMott et al., 2010; Liu et al., 2011; Seinfeld et al., 2016). Besides, they can also function as a catalyst for reactive gas species, forming secondary aerosols. Their strong heating abilities may strengthen deep convective processes by accelerating moisture convergence when beneath a cloud layer (Huang et al., 2014). Also, due to their dual functionality as effective IN and GCCN, these particles compete for raising to the freezing level, where they might encourage ice formation, and rapid growth and deposition of these particles, as huge liquid droplets (van den Heever et al., 2006; Yuan et al., 2021). However, dust particles aloft the cumulus clouds can reduce the storm's strength by increasing the atmospheric stability, as discussed in section 2.2. Also, reduced shortwave radiation underneath the dust layers may decrease maximum surface temperature, eventually leading to declined surface wind speeds and dust emissions (Rémy et al., 2015). Thus, the short-term effect of dust on

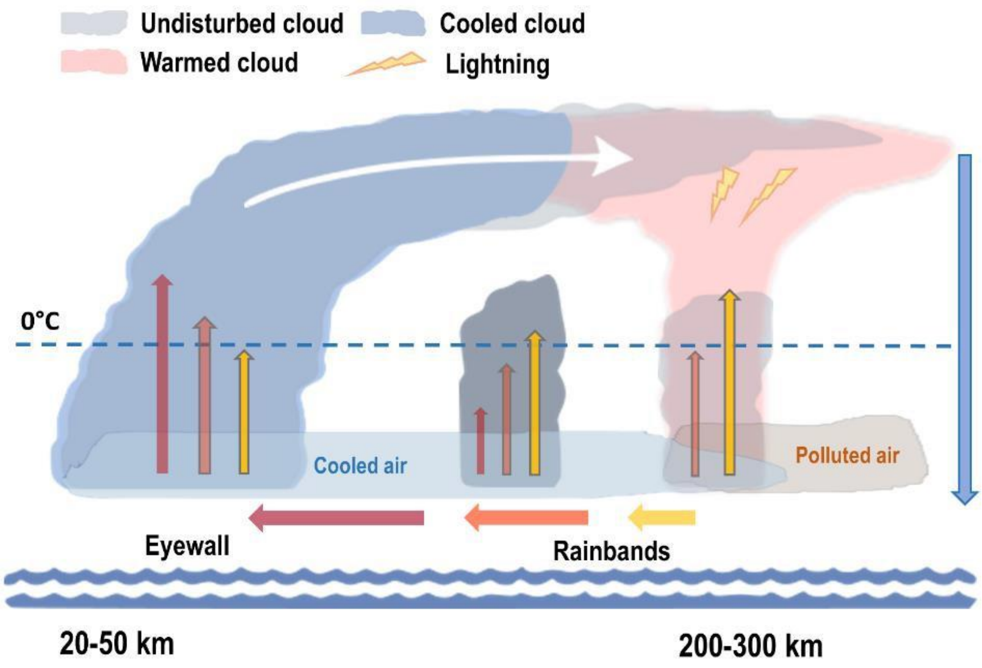

Figure 3: Schematic diagram of aerosol impacts on tropical cyclone (TC) for each of the three possible scenarios for aerosols: clean maritime condition (red), polluted condition (orange), and polluted condition with radiative forcing (yellow). Vertical velocities and low-level air flow are indicated by the upper and horizontal arrows, respectively. The length of the upper and horizontal arrows indicates the strength of the flow. Blue-colored down arrow represents the subsiding air farther away from the TC. This diagram is redrawn after adapting from Rosenfeld et al. (2012) and Wang et al. (2014).

surface radiation fluxes through dust-boundary layer-meteorology feedback mechanisms can also influence dust production during some episodic scenarios. Recent years have seen an increase in dust storms worldwide due to expanding desertification, fuelled by dust events that worsen drought conditions across semi-arid regions (Wang et al., 2008; Goudie, 2020). Therefore, understanding the interactions between aerosols, clouds, radiation, and precipitation over these arid and semi-arid regions is crucial, maybe more so than for any other place on the Earth.

4.3. Tropical cyclones

Tropical cyclones (or TCs), are one of the most devastating catastrophes worldwide, and are fuelled by the enormous amount of latent heat generated during condensation process and subsequent downpours. Therefore, it is reasonable to assume that aerosol-induced changes in the precipitation-forming mechanisms through microphysical effect would alter or reorganize the precipitation distribution, which can further modify the latent heating, and consequently impact the structure and intensity of the TCs (Figure 3; Rosenfeld et al., 2012). Since TCs can typically draw air from a distance equivalent to three times their radius, continental aerosols in an area with high pollution levels have a considerable probability of reaching the exterior of TCs before starting landfall (Zhao et al., 2018). For example, Nair et al. (2020) reported that when Ockhi developed into its mature stage, aerosol particles from the heavily populated Indo-Gangetic Plain were dragged into the storm, further influencing precipitation formation over the Arabian Sea. This penetration of submicron aerosols to the periphery of a TC from the nearby continents can delay the warm-rain processes and induce dramatic intensification of convective processes through the AIvE (as discussed in section 3.3), competing with the eyewall's convection (Rosenfeld et al., 2007; Carrió and Cotton, 2011; Jiang et al., 2016). Because the outermost region of the storm has more vigorous clouds, more rising air can be drawn there, weakening low-level convergence toward the center, further increasing central pressure, and a consequent drop in maximum wind speed (Rosenfeld et al., 2012). For instance, Khain et al. (2010) found that the successful penetration of continental aerosols into the outer rainbands of a TC led to a rise of 16 hPa in the central pressure and a lowering of the storm's maximum wind speed by 10-15 m s-1.

By melting and evaporating at lower altitudes, the enhanced ice precipitation within the peripheral clouds cools the air that converges towards the storm's center. Substantial low-level cooling creates cold pools that help in intensifying storm cells in the peripheral rainbands (Rosenfeld et al., 2007). These cold pools transport more moisture vertically, strengthening convection and increasing latent heating through a positive feedback loop (Zhang et al., 2009). Also, the re-evaporation of cloud droplets that did not form precipitation, contributes to further low-level cooling. Due to this additional cooling of the low-level air rushing to the core, the storm can be further weakened (Cotton et al., 2007; Rosenfeld et al., 2011; Wang et al., 2014). However, as transport depends on the local flow in such areas, the aerosols are not always carried efficiently from the surroundings where the tropical cyclone is embedded into the outermost rainband convection. The studies of Krall (2010) and Krall and Cotton (2012) revealed that Typhoon Nuri, which spread pollution from the Asian mainland, intensified early while ingesting pollution but later on, it lost strength. While several studies suggested that increased aerosol concentration in the outer rainband region might weaken TCs, some studies also showed evidence of strengthening TC intensity due to the penetration of continental aerosols in the periphery (Herbener et al., 2014; Evan et al., 2011). For instance, Evan et al. (2011) reported that continental aerosols favored TC intensification over the Arabian Sea by reducing vertical wind shear. However, if light-absorbing continental aerosols are penetrated into the peripheral rainband regions, ARI-induced low-level warming can further invigorate convection, increasing precipitation over the rainband regions and preventing water vapor transport into the eyewall areas, weakening the TC intensity (Figure 3; Wang et al., 2014). Thus, sometimes under the polluted scenario, ARI can dominate over ACI when TC reaches the continent (Liang et al., 2021). These enlarged rainband regions due to the anthropogenic aerosols may also increase TC rainfall areas, potentially leading to more intense flooding (Zhao et al., 2018).

Another kind of aerosol that can change TC intensity is GCCN (e.g., dust and sea-salt), which may have distinct effects on TCs compared to anthropogenic aerosols. Since GCCN can produce bigger-sized droplet embryos, a higher concentration of these particles can accelerate the raindrop formations, even in clouds of numerous little droplets (Blyth et al., 2003). As sea-salt production flux is a function of surface wind speed, the extremely powerful winds in and around the eyewall region can lift enormous amounts of sea-salt particles that produces rain consisting of sea water in the innermost spiral cloud bands (Rosenfeld et al., 2012; Jiang et al., 2019). Therefore, the rain that is typically inhibited by anthropogenic pollutants in the peripheral region may be partially restored by these highly hygroscopic GCCNs. A study by Khain and Lynn (2011) indicated the striking contrast between the pristine and polluted clouds in a TC. They indicated that while the clean clouds generated early warm rain before glaciating swiftly beyond the 0°C isotherm threshold, the dirty clouds delayed the onset of rain from heights of around 3 to 7 km, resulting in glaciation at approximately -22°C. Moreover, the center of a TC is almost saturated at low levels, preventing the creation of cold pools. Thus, the submicron CCN impact would be most substantial on the TC periphery, where wind speeds might be far lower than in the eyewall region, implying the presence of convection competition between the eyewall and outer rainbands (Liang et al., 2021). Several studies also illustrated how the Saharan dust aerosols influence the emergence and growth of TCs over the Atlantic Ocean by modifying thermodynamic structure, vertical wind shear, and distribution of the diabatic heating (Dunion and Velden, 2004; Zhang et al., 2007; Bretl et al., 2015; Twohy et al.,2017; Pan et al., 2018). They illustrated that while higher dust loading over the northern Atlantic Ocean prevented cyclonic disturbances from developing into TCs, dust particles from the Sahara Desert reduce their activity across the Atlantic Ocean by adding stable, drier air into the TC system. Twohy et al. (2017)

further indicated that dust particles lifted by a convective system across the eastern Atlantic Ocean increased concentrations of ice-forming particles in the upper levels of the troposphere. However, TC activities in the West Atlantic and the Caribbean Sea might be inhibited by the aerosol-induced cooling effects over the tropical North Atlantic Ocean (Lau and Kim, 2007).

4.4. Heat wave

Heat waves, defined as extended episodes of exceptionally high temperature, are one of the deadliest naturally occurring hazards that cause significant harm to ecological systems and human society (Perkins, 2015). These are usually linked with intense high-pressure systems that prevent the movement of rain-bearing low-pressure systems (Beniston and Diaz, 2004). Due to the rise in global mean temperature, these extreme events have become more frequent in recent decades over different parts of the globe (Donat et al., 2013; Mishra et al., 2015; Freychet et al., 2018). Notable examples include the severe, prolonged heat wave that swept throughout Europe in the 2003 summer, causing roughly 66000 fatalities, and another heat wave in western Russia in July 2010 resulted in 11000 deaths and 30% grain harvest losses (García-Herrera et al., 2010). Also, the agriculture industry and public health have suffered significant losses due to the heat waves that hit Central and Eastern China in July-August 2013, Northeastern China in 2014, and Western China in the summer of 2015 (Zhou et al., 2014; Wilcox et al., 2015; Sun et al., 2016; Ma et al., 2017). Each year, various metropolitan areas in India (particularly in Uttar Pradesh, Punjab, Bihar, Odisha, Telangana, and Andhra Pradesh) also experience severe heat waves during the pre-monsoon season (Ratnam et al., 2016; Kant et al., 2019c). In the summers of 2015 and 2016, these areas experienced an extreme heatwave situation that cost the lives of many people, animals, and birds (Pattanaik et al., 2016; Mishra et al., 2017). Ratnam et al. (2016) discovered two distinct types of heatwaves over India, one across the northwest of the country associated with air obscuring over the North Atlantic and the other over the eastern coast associated with the Matsuno-Gill effect. Additionally, a strong local influence on air temperature may significantly increase the severity of heatwave conditions (Ghatak et al., 2017).

The probability is high that severe heat waves will become more often, intense, and long-lasting under anticipated future global warming (Lau and Nath, 2014; Jones et al., 2015; Mora et al., 2017). Even though there is general agreement that greenhouse gases play a significant role in enhancing the intensity and duration of heat waves by raising the maximum temperature at the surface, there remains uncertainty on how aerosols could modulate these events by affecting radiation, clouds, and rainfall (Dileepkumar et al., 2018). Numerous studies have demonstrated that due to strict mitigation regulations to maintain air quality, aerosol emissions are projected to decrease globally over the 21st century, resulting in more extreme heatwave conditions (Levy et al., 2013; Xu et al., 2015; Horton et al., 2016).

Dong et al. (2016) suggested that anthropogenic aerosol affected extreme summertime temperatures by inducing precipitation-soil moisture-cloud-temperature feedbacks across China. Meywerk and Ramanathan (2002) found that the radiative cooling of the sea surface caused by the accumulation of the Indian pollution plume might reduce the water evaporated at the ocean surface by 5%, leading to a probable reduction in rainfall across the northern parts of the Indian Ocean. Several studies also indicated that a decrease in precipitation across the Sahelian region could arise from the reduced convective activity in the lower troposphere due to the radiative influence of dust particles (Konaré et al., 2008; Mallet et al., 2009). Local summertime warming coupled with less rainfall results in lower evaporation and reduced overall cloud cover due to the lack of precipitation, causing the soil to dry up (Su and Dong, 2019). The positive feedback to surface heating is produced by enhanced sunlight reaching the surface, decreased cloudiness, diminished vertical latent heat fluxes, and reduced evaporation. Krishnan and Ramanathan (2002) highlighted the potential for surface warming in areas far from the aerosol haze to counteract the local cooling. Dave et al. (2020) further showed that temperature maxima, both regionally and globally, are enhanced by absorbing aerosols. Recently, Sousa et al. (2019) also reported that heat waves were associated with the Saharan dust intrusions across central parts of Europe and the peninsula of Iberia. Thus, the association of aerosols with heat waves for modulating the local, and regional weather is a possibility, and should be explored further.

5. Significance of aerosols during large-scale circulations

Several studies employing the global climate model (GCM) have shown that aerosol modifies large-scale circulations by influencing the radiation budget and causing regional energy imbalances (Ming and Ramaswamy, 2011; Wang et al., 2015). Under the current aerosol conditions, reduced and enlarged tropical circulations can be expected due to the modified cloud radiative forcing brought on by redistributed aerosols, as indicated by reduced meridional stream function, zonal winds over the tropics, and a poleward shift of the jet stream (Rosenfeld et al., 2014). Also, the substantial radiative cooling, mainly through ACI in the Northern Hemisphere, conceivably hinders the northern branch of the Hadley circulation (Ming and Ramaswamy, 2011). Rosenfeld et al. (2014) proposed that sulfate aerosols can impact tropical sea surface temperature (SST) through ACI and aerosol-radiation interaction (ARI), altering circulation and precipitation in tropical regions. Wang et al. (2015) observed that large-scale circulations were influenced by the change in peak pollution during the 1970s. A significant association between aerosol and Madden-Julian oscillations can also be found, with the aerosol concentration being high (low) during the dry (wet) phase of this natural oscillation (Tian et al., 2008). There have also been reports of increased smoke aerosol loading because of enhanced forest fires over the northern region of South America during the El Nino year 2006 (Le Page et al., 2008). Increased carbonaceous aerosols on the maritime continent can boost atmospheric warming by shortwave absorption and SST cooling by reducing net surface radiation (Rosenfeld et al., 2014). The combined impacts result in more stable air and cooler SST, reducing rainfall and exacerbating drought conditions (Tosca et al., 2010). In addition to biomass burning, El Nino-induced drought conditions may also be linked to increased dust emissions across the desert regions of Australia and western parts of Africa (Prospero and Lamb, 2003; Mitchell et al., 2010).

Many GCM-based studies reveal that the absorbing BC aerosol through ARI can cause a northward rainfall shift in the Intertropical Convergence Zone (ITCZ), particularly over the Pacific Ocean (Wang, 2004; Chung and Seinfeld, 2005). By incorporating ACI in the models, some studies also found that the dynamic consequence of such a forcing might have a remote influence on ITCZ precipitation, i.e., a shift in the opposite direction from that generated by BC (Ramaswamy and Chen, 1997; Rotstayn and Lohmann, 2002). Rotstayn et al. (2007) further indicated that a high Asian aerosol plume could change the meridional SST, surface temperature, and atmospheric pressure gradients throughout the Indian Ocean, keeping ITCZ in the Southern Hemisphere and causing monsoonal winds more likely to flow toward Australia. Furthermore, increased convection along the northern part of the Atlantic ITCZ might be caused by atmospheric heating due to the higher concentrations of dust particles during Saharan dust episodes over the North Atlantic through the EHP effect (Lau et al., 2009). On an even deeper level, Booth et al. (2012) hypothesized that aerosols from burning fossil fuels, biomass burning, and volcanic eruptions could significantly contribute to the decadal shift of the North Atlantic ITCZ in the 20th century.

Aerosols can play a crucial role in weakening the low-level monsoon circulation by reducing the summertime land-sea thermal contrast (Song et al., 2014). Through dynamical feedback and localized surface-level processes in the aerosol-monsoon interaction, local pollution can cause the earlier arrival of the primary monsoon systems (Bollasina et al., 2013). Over the past few years, many studies focused on the impact of aerosols including absorbing type (dust and BC) on the south-west or summer monsoon. For instance, Manoj et al. (2011) suggested that the process by which the summer monsoon breaks into active phases may be influenced by heating caused by aerosol absorption and energized convection. Whether or not they coexist with scattering aerosols, absorbing aerosols can change the meridian gradients of moist static energy (MSE) within the sub-cloud layers over the Indian subcontinent, leading to the heating of the PBL air and affecting the large-scale atmospheric stability (Wang et al., 2009). Hazra et al. (2013) indicated that the associated dynamical feedback pathway, including aerosol-induced ARI and ACI, and large-scale dynamics might impact monsoon intra-seasonal variability and predictability. An observational study by Panda et al. (2023) investigated about the responses of clouds to aerosols over south Asia and its association with Indian Ocean Dipole (IOD) scenarios concerning summer monsoon. They found that the height of monsoonal cloud tops is getting lowered in polluted conditions during both phases of IOD. And increased aerosol concentration could be associated with reduced rainfall over some regions during the summer monsoon. However, Vinoj et al. (2014) proposed that unusual dust buildup over the Arabian Sea through non-local feedback processes involving ARI-induced atmospheric warming, the shift in circulation patterns, and moisture convergence, could increase monsoon rainfall across central and northern India. Aerosols may also influence the winter monsoon due to aerosol-induced solar dimming and reduced temperature gradient between high and low latitudes and between land and ocean surfaces (Niu et al., 2010). Findings of both the observational and modeling studies suggest that increased BC loading over Africa could be linked to a considerable decrease in surface precipitation, cloud top height, and cloud abundance during the boreal cold seasons (Huang et al., 2009; Wang, 2004).

6. Current challenges, limitations, and future scope

6.1. Observational perspective

Satellite platforms like Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation and CloudSat and global gound-based observational networks like Aerosol Robotic Network provide valuable information about the optical characteristics of aerosols or their loading in terms of total extinction. Still, they do not directly pass on information about aerosol number concentration, size distribution, or chemical composition, which are crucial for understanding CCN effects (Tao et al., 2012). Also, cloud contamination and other issues make satellite observations of aerosols unreliable (Li et al., 2009). Measures taken for determining cloud droplet number concentration using existing sensor retrievals, such as those from MODIS, benefit only a few specific cloud types (Bennartz, 2007). The mean droplet size distribution smooths out any discrepancies in size distribution at various cloud locations, making it a less relevant property when averaged across a broad cloud area. Due to mixing processes, the distributions of cloud drop sizes at cloud bases should range significantly from those at higher altitudes when raindrops first start falling and from those at cloud edges (Fan et al., 2016). Even realistic explanations of ice nucleation remain speculative in most places, as measuring the ice nucleation properties (concentration, diversity, and IN capability) has remained challenging. Observational difficulties are further exacerbated by the frequent masking of ice nucleation by secondary ice production (Fan et al., 2016). Since nearly hydrometeor size distribution serves as the basis for all microphysical processes, the lack of measures associated with droplet size distributions is a significant barrier to improving the model depiction of cloud microphysics (Khain et al., 2015).

Too few or inadequate essential measurements exist to correctly characterize aerosols and cloud characteristics (especially for deep convection). AIvE may be tested directly with observations using valid CCN and vertical velocity datasets. However, accurately measuring vertical velocity for convective clouds has been challenging. Also, one cannot completely comprehend the feedback between dynamics and microphysics due to the lack of measurements of in-cloud microphysical parameters, such as the number and mass mixing ratios of hydrometeors (Fan et al., 2016). Measurements of aerosol characteristics, CCN, the size distribution of cloud droplets, and prevailing updraft at the cloud base are essential for distinguishing aerosol influences from meteorological factors, which can be possible through aircraft measurements. However, most of these measurements have been carried out for weakly convective clouds, which is not a statistically plausible representation of all cloud types (Fan et al., 2016). The main issue is the non-availability of vertical profiles of aerosols across large areas for mesoscale convective storms fueled by deep layers of air. Besides, penetration of aerosols into clouds cannot be detected by aircraft measurements, because these measurements are not that frequent and continuous. Therefore, our understanding of severe storm dynamics and its association with microphysics has been limited by the shortage of reliable data for measuring the vertical profile of aerosols across a large area.

6.2. Modeling perspective

With similar initial conditions, clouds and convection simulations in a modeling framework can differ significantly due to the different parameterizations, which range from a single moment to bin approaches (Khain et al., 2009; Fan et al., 2012). Khain et al. (2015) illustrated that the majority of bulk systems are poorly suited for investigating ACI because they usually (1) do not account for CCN budgets, which can lead to inaccurate conclusions about the aerosol effect; (2) use the saturation adjustments method, which excludes supersaturation and lessens the sensitivities of the bulk schemes to aerosols, rather than explicitly calculating diffusional growth according to droplet sizes and supersaturation; (3) use parameterizations for auto conversion typically developed under a limited set of circumstances and do not account for the auto conversion's temporal evolution; (4) use mean fall velocities for hydrometeor collision processes, which is a big issue for self-collections.; (5) employ two distinct sets of averaged fall velocity values, mass-mean and number-mean fall speed, in two-moment schemes, which would produce cloud areas of substantial mass but the insignificant amount or those having a considerable amount but with negligible mass; and (6) do not take into consideration of smaller particles that fall more slowly and larger ones that fall much faster. Mainly two- or three-moment bulk schemes may work well for cloud simulations in clean scenarios, especially with substantial dynamic forcing like huge squall lines (Morrison et al., 2015). Nevertheless, they frequently respond to the rise in CCN differently than bin models (Khain et al., 2009; Fan et al., 2012). Also, the bulk scheme does not accurately simulate the smaller size of ice particles and their terminal velocity in stratiform or cumulonimbus clouds under polluted conditions (Fan et al., 2016).

Bin parameterizations, on the other hand, are somewhat computationally expensive to perform. Significant uncertainties also exist for several physical parameters in bin models, including the condensation coefficient, collision efficiency, riming rate, falling speeds of various hydrometeors, and interstitial aerosol scavenging efficiency (Fan et al., 2016). One more problem with bin parameterizations is related to spectrum broadening. Because of these unresolved issues, using bin models may not be advantageous, especially when the size distribution and composition of the aerosol are unknown. However, bin parameterizations are more physically accurate. The uncertainty related to the model approach of microphysics can be reduced using strong constraints derived from aerosol and cloud measurements. How to effectively parameterize subgrid clouds at different resolutions is an important challenge in today's global and regional climate models. Most parameterizations in these models do not consider how aerosol interacts with those subgrid clouds. Nonetheless, the performance in modeling cloud characteristics and precipitation has been dramatically improved for subgrid parameterizations, considering various aerosol influences (Song et al., 2012; Grell and Freitas, 2014).

6.3. Future prospects

To date, only a few aerosol impacts may be quantified using the current satellite observations of CCN, cloud-based updrafts, and the structure of clouds (Rosenfeld et al., 2016). The extraction of CCN and aerosol optical characteristics should be a goal for next-generation satellites and worldwide networks. Continuous observations of CCN and cloud dynamics and microphysical features at different spatiotemporal scales gathered over numerous locations would offer plenty of information necessary to create a breakthrough in our understanding of ACI. Ground-based radar, lidar observations, and airborne measurements would be very beneficial to learn more about the processes governing mixed-phase and ice clouds (Fan et al., 2016). Recently, box closure experiments have provided a method to collect concurrent details in various climate zones via incorporating satellite, aircraft, and surface-based remote sensing measurements (Rosenfeld et al., 2014). The box closure technique offers new information for evaluating and developing models and parameterizations. It is also an excellent way to determine the changes in matter and energy in all its vital components in the climate system due to aerosol change. However, carrying out these experiments requires a significant efforts and sufficient funds and the cooperation among several closely coordinated groups. Besides field observations, laboratory studies on the ice nucleation processes, secondary ice production, collision efficiency, aerosol scavenging, and riming can significantly increase our understanding of these processes (Hallett and Mossop, 1974; Mitra et al., 1992; Niemand et al., 2012). Laboratory studies are always encouraged since findings from these experiments often assist us in developing appropriate model representations. For instance, the laboratory results of Garca-Garca and List (1992) served as the foundation for the current hypothesis of wet hail development in the spectral bin microphysics model proposed by Phillips et al. (2015). As diabatic processes play a dominant role in the climate system, the most effective approach to minimize ACI uncertainties in climate projection is to increase the accuracy of detailing in cloud microphysics parameterization. Lack of knowledge about size distributions, shapes, phases, and conversions of particles during deep convection demands the ongoing advancement of bin microphysics in cloud-resolving models. The aerosol wet scavenging topic needs further research and should be appropriately incorporated into the model (Seinfeld and Pendis, 2016). Also, the size distribution of different types of hydrometeors must be precisely simulated to explain cloud microphysics effectively (Fan et al., 2016). Integrating data gathered from polarimetric radars with bin microphysics could be a convenient approach to advance our understanding of hydrometeor behaviors and conversions (Kumjian et al., 2014). Till now, the depiction of cloud-borne aerosols is absent from almost all bin models, and some studies employed a straightforward method for CCN regeneration (Fan et al., 2009). The primary issue with bin microphysics for large-scale models is the computational cost. Hence, the scientific and technological advancements done till date, should be implemented in new algorithms to minimize the current computational expenses. Besides, cautionary measures should be taken when extrapolating an empirical parameterization for different scenarios. Such parameterizations should only be used when appropriate measurements can be performed to validate them. Given decreased particle size distributions and smaller bin sizes, a hybrid moment and bin method can reduce computational costs at the expense of some accuracy loss. Moreover, a global climate model must have parameterizations applicable to any region, climatic condition, and high temporal and spatial resolution. One general approach to reduce model uncertainty is to substitute empirical parameterizations with physics-based schemes, as these can be more easily adjusted to various cloud and climatic regimes (Fan et al., 2016). As an alternative, the reduced format of a specific global aerosol-climate model might be created by running a small number of global model simulations using a specific empirical method like the one employed by Tatang et al. (1997). In a full-scale parametric uncertainty analysis, one can utilize this reduced format model to imitate the behaviors of the global aerosol-climate model because it is significantly more computationally efficient with certain restrictions (Tao et al., 2012). If the modeling framework is able to consider the interactions including ACI and ARI appropriately through a physics-based parameterization, it may help in simulating the microphysical and hydrometeorological processes effectively and efficiently over most parts of the world.

7. Concluding remarks

This paper systematically reviews the current

Research on atmospheric aerosols and their influences on radiation, cloud, and precipitation over different parts of the globe. Also, their role in influencing extreme weather events and large-scale circulations is emphasized. Through radiative effect, i.e., ARI (including DE, SDIE, SDE), aerosols can alter planetary albedo, atmospheric stability, circulation pattern, and cloud distributions, influencing the Earth's climate. While scattering aerosols (e.g., sulfate) induce a cooling effect, absorbing aerosols (e.g., BC) causes warming in the climate system. Whether absorbing aerosols can increase or decrease the cloud distribution depends on their concentration, chemical structure, size distribution, altitude position relating to the clouds, and cloud types.

Aerosol can influence cloud properties and precipitation efficiency through ACI by acting as CCN or IN, which further depends upon their chemical composition and size distribution. While alteration in the droplet size and albedo of clouds with rising aerosol concentration for a given LWP falls under FAIE, the subsequent suppression of warm-rain processes and enhancement in cloud lifetime and cloud fraction is considered SAIE. Besides, the presence of GCCN activated on the larger particles like dust, sea salt, and some PBAPs can induce the 'anti-Twomey effect' and switches the droplet size distribution to the larger ones. Moreover, the prolonged cloud lifetime due to SAIE promotes more water to be reached beyond the freezing level, resulting in the additional release of latent heat in the upper troposphere, which further strengthens updrafts and helps in forming more taller convective clouds. This aerosol effect is designated as AIvE in section 3.3. Due to more complicated dynamics, thermodynamics, and microphysics, aerosol impacts on deep convective clouds are even more complex than those on warm clouds and, thus, somewhat less well understood. It has been challenging to comprehend how aerosol particles might modify the IN characteristics, given the diversity of heterogeneous nucleation processes of IN.

Since aerosol particles can modulate regional and global circulation patterns, they can also significantly influence extreme weather events. Under favorable meteorological conditions, submicron aerosols can enhance thunderstorms and associated lightning activities in polluted areas. However, absorbing aerosols via ARI may considerably reduce atmospheric instability in a dry, dirty environment, reducing thunderstorm activities. In addition to the increase in lightning flash frequency, aerosols also modify the polarity of the processes that produce a charge in convective clouds. Due to strong absorption capabilities, sand and dust particles can accelerate the convective processes by advancing moisture convergence when below a cloud layer. Also, these particles may compete to be lifted to the freezing level because of their dual functioning as effective IN and GCCN. However, dust particles above cumulus clouds might decrease the dust storm's strength by enhancing atmospheric stability.

Moreover, aerosol-induced modifications to the precipitation-forming processes through microphysical effects might change or reorganize the distribution of precipitation in the TC, which may further affect the latent heating and ultimately impact the storm's structure and intensity. In most cases, the penetration of small particles to the storm's periphery from the nearby land areas might invigorate convection around the rainband regions, weakening the TC intensity. If light-absorbing particles are introduced into the outer rainband regions, ARI-induced low-level warming can further energize convection and weaken TC. The submicron CCN influence would be most significant on the TC perimeter, where wind speeds may be much lower than in the eyewall area, indicating convection rivalry between the eyewall and outer rainbands. Also, by creating feedback between rainfall, soil moisture, clouds, and temperature over land areas, anthropogenic aerosols could impact extreme summer temperatures, increasing the occurrences of heatwaves. Besides, the maximum surface temperature can be increased by absorbing aerosols like desert dust aerosols at both regional and global scales. Aerosols alter large-scale circulations by affecting the radiation budget and causing regional energy imbalances. The northern portion of the Hadley circulation is assumed to be hampered by significant radiative cooling, primarily via ACI in the Northern

Hemisphere. There is a strong correlation between aerosols and Madden-Julian oscillations too. Increased emissions of carbonaceous aerosols caused by widespread forest fires and dust particles generated from the desert regions may also be related to El Nino-driven drought conditions. It is also evident that the decadal shift of the North Atlantic ITCZ in the recent century have been influenced by aerosols from burning fossil fuels, burning biomass, and volcanic eruptions. Local pollution can result in an earlier arrival of the major monsoon systems via dynamical feedback and localized surface-level processes in the aerosol-monsoon interaction. Absorbing aerosols-induced atmospheric heating through ARI could modulate large-scale atmospheric stability, circulation patterns, and monsoon rainfall over widespread areas.

Systematic measurements of CCN, cloud dynamics, and microphysical characteristics at various spatiotemporal scales gathered across several sites could be helpful in advancing our understanding of ACI in different weather and climate scenarios. To further understand the mechanisms governing mixed-phase and ice clouds, surface-based observations using active sensors like radar, lidar, and aircraft measurements would be more beneficial. The box closure experiment is an excellent example of gathering simultaneous information in several climate zones via incorporating aircraft, satellite, and surface-based measurements. However, performing these experiments is difficult due to the requirement of more resources. The laboratory studies are therefore encouraged to investigate the fundamental mechanisms of ice nucleation, secondary ice production, collision efficiency, aerosol scavenging, and riming to help improve the numerical modeling of concerned processes. The most efficient way to reduce the uncertainties in climate projections is to improve the accuracy of cloud microphysics details in the cloud-resolving models. Integrating data from polarimetric radars with microphysics could be a practical solution to improve our understanding of hydrometeor behaviors and conversions. A global climate model needs physical-based parameterizations applicable to any climatic zone and large spatiotemporal scale. Alternatively, the reduced format model of a particular global aerosol-climate model could be used to increase computational efficiency. The research direction outlined above must be continued for better understanding of the interaction of aerosols with radiation, clouds, and precipitation under different meteorological conditions.

Acknowledgments

The fellowship support for Ankan Sarkar through INSPIRE (IF180959) provided by the Department of Science and Technology, Government of India, is highly acknowledged.

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