Dust Induced Changes in Ice Cloud and Cloud Radiative Forcing over a High Altitude Site

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ABSTRACT

Aerosol-cloud interaction is the subject of considerable scientific research, due to the importance of clouds in controlling climate. In the present study, three years (2011–2013) satellite observations are used to investigate the aerosol indirect effect (AIE) over Dehradun. The low values of Angstrom exponent ($\alpha$) during March–July are attributed to the loading of dust-like coarse particles in the atmosphere, whereas the analysis of aerosol type and Spectral Radiation Transport Model for Aerosol species (SPRINTARS) simulated aerosol species variation supports the fact. Moderate Resolution Imaging Spectroradiometer (MODIS) derived AOD data are associated to the cloud product to examine the dust impact on properties of liquid cloud and ice cloud. The positive values of aerosol cloud interaction effect (ACI) for ice cloud during pre-monsoon (March–May) and monsoon (June–August) seasons reveal the significant impact of dust on ice clouds over Dehradun, which is maximum during May (~0.24 ± 0.05). The present study shows that ice cloud effective radius (ICER) decrease with AOD during dust period. The increase in ice water path (IWP) and ice cloud optical depth (ICOD) reveals the impact of dust on heterogeneous ice generation in low level clouds. However, there is no relation between dust and liquid water cloud during dust period. It is difficult to provide definite conclusions that the dust and cloud changes are driven by the same meteorological conditions. Cloud and the Earth’s Radiant Energy System (CERES) derived flux data are used to examine the associated changes in TOA cloud radiative forcing. The diminution in effective size of ice crystal due to aerosol first indirect effect traps more longwave radiation and reflects more solar radiation. Both first and second indirect effects enhance cloud cooling, whereas the dust induced cloud warming is mainly the result of the semi-direct effect.

Keywords: Dust; Ice cloud; Cloud radiative forcing; Aerosol indirect effect.

INTRODUCTION

Aerosols have a crucial role in the life cycle of clouds by acting as cloud condensation nuclei (CCN) or ice nuclei (IN), and can modulate the distribution of radiative heating indirectly. Concerning the four major terrestrial sources of aerosols are biomass burning, desert dust, anthropogenic aerosol and biogenic aerosol, dust aerosols are responsible for significant climate forcing through their direct effect on radiation (Sokolik and Toon, 1996; Patel and Kumar, 2015) and their indirect/semi-direct effect on clouds and precipitation (Huang et al., 2010; Min and Li, 2010). An indirect effect from dust aerosols over both concerning warm and cold clouds have been studied by Li et al. (2010) and Min et al. (2009) in different studies. Rosenfeld et al. (2001) found reduced precipitation in shallow convective clouds near the source of Saharan dust. This was hypothesized to be due to lowering of the coalescence efficiency in clouds formed by smaller cloud droplets. The correlation between the presence of dust and ice formation in modestly supercooled altocumulus clouds was demonstrated by Sassen (2002). Levi and Rosenfeld, (1996) have reported the increase in concentration of ice nuclei during dust storm events in Israel. In addition, dust is also responsible for semi-direct effects, where the mixing of dust and absorbing aerosol changes the cloud properties. The loading of absorbing aerosols can produce a local heating that in turn changes the relative humidity and stability of the atmosphere and thereby modulate cloud formation and lifetime. This induces a change in cloud cover and cloud albedo, depending on the vertical distribution of aerosols (Koch and Del Genio, 2010). It has been observed that the semi-direct effect of dust aerosols suppresses the thickness of the clouds (Hung et al., 2006a, b; Yorks et al., 2009).

The Indian subcontinent is highly influenced by aerosols, where the tropical and subtropical climate in turn modulate the aerosol characteristics (Ramachandran and Cherian, 2008). India is an ideal region to study aerosol-cloud interaction.
due to significant spatiotemporal variations in aerosol and cloud characteristics. Desert dust, mainly transported from Africa, Arabian Peninsula and adjacent Thar Desert, is the most frequent aerosols present over the Indian sub-continent during pre-monsoon and monsoon seasons (Prijith et al., 2013; Kaskaoutis et al., 2014). Generation and advection of dust aerosols depends on the weather conditions and the strength of the monsoon circulation (Manoj et al., 2011). Dust aerosols can modify the radiative budget both at the surface and top of the atmosphere (TOA) significantly (Patel and Kumar, 2015). According to climate model simulations, dust induced heating of the atmosphere over West Asia and North Africa rapidly modulate monsoon rainfall over central India (Vinoj et al., 2014). Dust induced heating may cause significant changes in regional scale processes in the absence of large-scale processes during monsoon breaks. The attention on dust aerosol properties over India is found to be increasing but very few studies are reported the impact of dust on cloud and climate over India. In this, the present study helps to understand the impact of dust on cloud and radiation.

In the present study, we make use of Moderate Resolution Imaging Spectroradiometer (MODIS) and Cloud and the Earth’s Radiant Energy System (CERES) derived aerosol, cloud and radiation data during 2011–2013, to investigate the dust induced change in cloud dynamic and radiative properties over a high altitude site (Dehradun and surroundings). The aim of this research is to investigate changes in both phase of clouds (liquid cloud and ice cloud) and associated change in cloud radiative forcing at the top of the atmosphere (TOA).

**STUDY AREA**

The Dehradun region (30.31°N and 78.36°E) is the capital of the Uttarakhand state (Fig. 1). It is located at a mean altitude of 700 meter above mean sea level, extends 80 km in length and 20 km in width in the Shivalik range of Himalayas. The region experiences the low-level calm north/northeasterly synoptic winds during winter (December–February) are from the polluted regions; temperature is colder and the atmosphere is dry (Fig. 1). In pre-monsoon (March–May) winds originate and travel through an arid/marine region from west (Fig. 1). In this season westerly winds carries the mineral dust from adjacent Thar Desert and Arabian Peninsula (Sikka, 1997; Patel and Kumar, 2015). The strong winds are from marine and western region carries moisture (RH is high (> 70%) during monsoon (June–August) (Fig. 1). There is shift in the wind patterns from

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**Fig. 1.** ECMWF derived seasonal variation of synoptic wind pattern and relative humidity over India during winter, pre-monsoon, monsoon and post-monsoon. The labeled contour describes the wind speed (m s⁻¹), whereas the color bar shows the variation of relative humidity (%). The black dot shows the study region (Dehradun).
southwest to northeast during post-monsoon (September–November). In post-monsoon RH is low (< 50%) over north and west India, while RH is high (> 70%) over east and south India (Fig. 1). In this season, the atmosphere is loaded with black carbon and other fine mode organic particles due to large-scale biomass-burning from the Indo-Gangetic plan (IGP) (Kant et al., 2012). In addition, the mean annual rainfall is recorded ~2000 mm, mostly in the monsoon season (70%–80% of the total annual rainfall).

**DATA AND METHODOLOGY**

Three years (January, 2011–December, 2013) of satellite observations are used to investigate aerosol-cloud-radiation interaction over Dehradun and surrounding regions.

**MODIS Derived Aerosol and Cloud Data**

The Moderate Resolution Imaging Spectroradiometer (MODIS) instruments allow to study many of the Earth’s terrestrial and oceanic characteristics with a single instrument. In the present study, data from MODIS remote sensor on board the Aqua satellite are used, which provide an opportunity to study aerosols and cloud properties from space with high accuracy. In the present study, MODIS AOD (at 550 nm) level 2 products (10 km × 10 km; MOD04 L2, version 6) along with MODIS level-2 cloud product (1 km × 1 km, MOD06 L2, version 6) are used to study aerosol cloud interaction. MODIS derived AOD at 550 nm (Kauffman et al., 1997) is used as a proxy of CCN for this study for further AIE analysis. The accuracy of the MODIS derived AOD is 0.05τa ± 0.15 (τa means AOD) over land (Levy et al., 2010), which is validated against ground-based remote sensing. MODIS level-2 cloud properties are retrieved from visible and near infrared wavelengths (King et al., 2003) and proved in good agreement with ground-based cloud measurements of warm clouds (Platnick et al., 2003). The MODIS derived cloud parameters used in this study include cloud fraction, cloud effective radius (CER), cloud optical depth (COD) for both liquid and ice clouds along with liquid water path (LWP) and ice water path (IWP). However, the primary focus of this study is to examine the dust induced change in cloud properties, so only data from lower cloud layer are considered, which may have direct impact of dust. To match the resolution, we aggregated MODIS derived aerosol and cloud properties to a resolution of 20 km × 20 km.

**CERES Derived TOA Flux Data**

CERES Single Scanner Footprint (SSF) (Geier et al., 2001) data sets are used here to study changes in cloud radiative forcing during 2011–2013 over Dehradun surroundings. The SSF product is a high-quality compilation of CERES derived radiation measurements includes cloud properties from MODIS and meteorological fields for radiation and climate studies. In the present study, CERES measured fluxes are used to estimate the impact of dust aerosol-cloud interaction on TOA radiation. CERES instruments measure broadband radiances at the TOA with a spatial resolution of ~20 km at nadir in three spectral regions (0.2–5.0 μm, 8–14 μm and 5–100 μm). The radiances measured are converted to TOA fluxes using angular distribution models according to the scene classification by Loeb et al. (2005). Uncertainties in temporal variation and spatial distribution of radiation (solar radiation and outgoing longwave radiation) fluxes in the CERES measured TOA flux are relatively smaller than Earth Radiation Budget Experiment (ERBE) measured flux data, due to better angular distribution models and better scene identification (Smith et al., 2012). To study the impact on radiation fluxes due to the modification of cloud characteristics, the instantaneous TOA shortwave (SW) and longwave (LW) fluxes are associated with the MODIS re-formatted (20 km × 20 km) data sets.

**SPRINTARS Derived Aerosol Species Data**

The Spectral Radiation Transport Model for Aerosol Species (SPRINTARS) is a global aerosol transport-radiation model, based on a general circulation model (Takemura et al., 2000, 2002, 2005), that simulates the transport of tropospheric aerosols (e.g., dust, sulfate, organic carbon, black carbon, and sea-salt) over a global and regional domain. In the present study, SPRINTARS simulated data are used to interpret the temporal distribution of aerosol species (e.g., dust, sulfate, organic carbon, black carbon, and sea-salt). Daily mean data of aerosol distributions on a 1.125° × 1.125° degree grid is acquired for three years (2011–2013). Monthly averaged SPRINTARS simulated data is not used for the validation purpose in this study but it is used to monitor the seasonal variation of different aerosol types.

The European Centre for Medium-Range Weather Forecasts (ECMWF) derived daily relative humidity (RH) and wind data sets at a resolution 0.5°× 0.5° are used to investigate the influences of meteorological variables on aerosol-cloud interaction.

As the aim of this study to investigate the impact of especially dust aerosol on cloud properties, we have performed the aerosol type analysis using AOD and Angstrom exponent (α) to distinguish the dust period. Detail methodology is given in the following section.

The aerosol cloud interaction can be detected using aerosol modified cloud effective radius (r_e). For a constant cloud water path (CWP) an increase in aerosol concentration decreases the r_e, thus, increasing cloud optical depth (COD). This means enhanced backscattering of solar light due to presence of smaller and more numerous cloud droplets known as cloud albedo effect. The cloud albedo effect, expressed as Aerosol-Cloud Interaction (ACI), can be defined as Eq. (1) (Feingold et al., 2003). The term ACI as it is defined here is associated with cloud microphysical responses rather than “indirect effect” or “cloud albedo effect”. In the present study, ACI is estimated for both liquid and ice clouds using Eq. (1). In order to understand the ACI estimation, MODIS measured liquid water path (LWP) and ice water path (IWP) are binned in 12 different bins ranging between 1 and 240 at an interval of 20 g m⁻². The ACI for each bin are calculated using Eq. (1).

\[
ACI = -\frac{\partial \ln r_e}{\partial \ln r_a} \bigg|_{\text{CWP}}
\]
where, $\tau_a$ is the observed aerosol optical depth used as a proxy of aerosol amount, $r_e$ represent both liquid and ice cloud effective radius for fixed LWP and IWP. The LWP and IWP are represented by CWP for both cloud phases. This study helps to reduce the uncertainties in estimating the radiative forcing due to cloud albedo effect.

The co-located CERES TOA fluxes with MODIS observations are used to estimate the cloud radiative properties, namely Short-Wave Cloud Radiative Forcing (SWCRF), Long Wave Cloud Radiative Forcing (LWCRF) and Net Cloud Radiative Forcing (NETCRF). The cloud radiative forcing is estimated by taking a difference between TOA clear-sky and all-sky (Ramanathan et al., 1989; Kiehl, 1994):

\[
SWCRF = SWF_{\text{clear}} - SWF_{\text{all}}
\]

\[
LWCRF = LWF_{\text{clear}} - LWF_{\text{all}}
\]

\[
NETCRF = SWCRF + LWCRF
\]

where, the subscripts ‘clear’ and ‘all’ represent the TOA clear-sky and all-sky fluxes, respectively.

SWF and LWF represent the shortwave forcing and longwave forcing. CERES flux measurements are used to estimate cloud radiative forcing over Dehradun and surroundings.

RESULTS AND DISCUSSIONS

Aerosol Overview

Three years monthly averaged AOD at 550 nm and $\alpha$ from MODIS over Dehradun are shown in Fig. 2. AOD and Angstrom exponent ($\alpha$) indicate the loading of aerosol in the atmosphere and variation in the aerosol size, respectively. The small values of $\alpha$ indicate the loading of coarse mode particles in the ambient atmosphere, while the higher absorption of fine-mode particles lead to the large $\alpha$ values. The result reveals significant variability in both AOD and $\alpha$ indicating the influence of meteorology, variety of aerosol types and sources. The large variation in the atmospheric conditions, boundary layer dynamics and long-range transport lead to the large variation in the range of AOD and $\alpha$ over Dehradun and surroundings.

The aerosol optical properties over Dehradun show large variability, indicates significant seasonal change in atmospheric conditions and the dominance of aerosols of different size, composition, and optical properties. The yearly mean AOD is observed to be decreased (0.49 ± 0.23) during pre-monsoon (March–May) season, while the increment in AOD (0.78 ± 0.40) is observed during monsoon (June–August). The trend of $\alpha$ is observed to be slightly different as compared to AOD. The yearly mean $\alpha$ is found to be increased during winter (0.95 ± 0.46) and post-monsoon (0.87 ± 0.42), while the small decrement is found in $\alpha$ during pre-monsoon (0.66 ± 0.24) and monsoon (0.73 ± 0.35) seasons, which may be due to the presence of coarse mode particles. $\alpha$ value shows the pronounced decrease in pre-monsoon compared to monsoon (Fig. 2). The gradual increase in the AOD and $\alpha$ from post-monsoon to winter can be ascribed to the advection of fine mode anthropogenic aerosols with the easterly and northeasterly wind (Fig. 2). The urbanization and industrial activities have a direct impact on aerosols over Dehradun. The transportation of dust from arid regions with westerly and northwesterly winds shows the sudden decrement in $\alpha$ during pre-monsoon and monsoon (Table 1). The external mixing of the dust and pollution shows relatively high $\alpha$ value in monsoon compared to pre-monsoon (Fig. 2). The analysis reveals that the dominance of coarse mode particle during pre-monsoon and monsoon, while the loading of fine mode particles in winter and post-monsoon over Dehradun.

Fig. 2. Monthly averaged MODIS derived aerosol optical depth (AOD) (Box chart) and Angstrom exponent ($\alpha$) (line plot) over Dehradun and surroundings during 2011–2013.
We have performed aerosol type analysis to confirm the above analysis. The correlation between AOD and $\alpha$ was used for aerosol discrimination along with threshold values on the basis of aerosol range and aerosol characteristics (Kaskaoutis et al., 2007, 2009). The present study is followed by the same threshold values, which are used in Patel and Kumar (2015) over Dehradun to discriminate the aerosol types. In the present study, we have discriminate the desert dust (DD), biomass burning (BB), anthropogenic aerosol (AA) and mixed aerosol (MA) over Dehradun. Fig. 3 describes the percentage contribution of four different aerosol types (MA, AA, DD and BB) over Dehradun.

The large dominance of mixed aerosols (MA) and anthropogenic aerosols (AA) is found during winter (~54.26%) and post-monsoon (~49.59%). The involvement of AA depicts the pollution of the area due to excessive fuel combustion for tourism. The desert dust (DD) contributes 68.98% and 56.15% in pre-monsoon and monsoon, respectively that indicates the high loading of dust in the pre-monsoon (~60%) and monsoon (~56.15%), while it is negative in post-monsoon (~49.59%). The involvement of anthropogenic aerosols (AA) is found during winter (~54.26%) and post-monsoon (~49.59%).

The aerosol type analysis supports the statement that the dominance of dust-like coarse mode aerosols over Dehradun and surroundings.

In addition to this, we have used three years SPRINTARS simulated different aerosol types distribution data. Fig. 4 describes the monthly variation of aerosol species distribution (e.g., dust, sulfate, black carbon, organic carbon and sea-salt) over Dehradun and surroundings for 2011–2013. The analysis reveals that the relative increment in dust concentration (30%–60%), increase the AOD during pre-monsoon and monsoon season, this leads to the decrease in $\alpha$ value (20%–40%). Patel and Kumar (2015) reported the transportation of dust along with the westerly and southwesterly wind from the arid and semi-arid region over Dehradun, which supports the SPRINTARS simulation of dust aerosol distributions (Fig. 4). SPRINTARS simulated aerosol distributions indicate the presence of sulfate in the atmosphere throughout the year, which probably transports from Southeast China.

The contributions from organic and black carbon are very small to the total aerosol optical depth for all the seasons. The external mixing of sulfate or carbonaceous aerosols with the dust may respond to the regional level cloud dynamics and radiative properties. The analysis supports the large contribution of dust during pre-monsoon and monsoon over Dehradun (Table 1).

### Table 1. Monthly mean variation of aerosol and cloud properties over Dehradun and surroundings.

<table>
<thead>
<tr>
<th>Month</th>
<th>MODIS AOD</th>
<th>Angstrom exponent ($\alpha$)</th>
<th>SPRINTARS Dust AOD</th>
<th>RH (%)</th>
<th>Cloud Fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Jan</td>
<td>0.53 ± 0.31</td>
<td>0.96 ± 0.46</td>
<td>0.01097</td>
<td>29.12 ± 3.45</td>
<td>0.22 ± 0.11</td>
</tr>
<tr>
<td>Feb</td>
<td>0.43 ± 0.20</td>
<td>0.97 ± 0.47</td>
<td>0.03833</td>
<td>39.17 ± 4.95</td>
<td>0.24 ± 0.12</td>
</tr>
<tr>
<td>Mar</td>
<td>0.46 ± 0.25</td>
<td>0.71 ± 0.30</td>
<td>0.1573</td>
<td>32.01 ± 4.60</td>
<td>0.34 ± 0.17</td>
</tr>
<tr>
<td>Apr</td>
<td>0.45 ± 0.20</td>
<td>0.63 ± 0.18</td>
<td>0.23961</td>
<td>33.04 ± 4.65</td>
<td>0.44 ± 0.15</td>
</tr>
<tr>
<td>May</td>
<td>0.57 ± 0.25</td>
<td>0.65 ± 0.25</td>
<td>0.39673</td>
<td>36.81 ± 4.84</td>
<td>0.54 ± 0.20</td>
</tr>
<tr>
<td>Jun</td>
<td>0.83 ± 0.44</td>
<td>0.67 ± 0.28</td>
<td>0.3646</td>
<td>59.97 ± 7.99</td>
<td>0.83 ± 0.31</td>
</tr>
<tr>
<td>Jul</td>
<td>0.97 ± 0.49</td>
<td>0.68 ± 0.33</td>
<td>0.29468</td>
<td>80.33 ± 9.01</td>
<td>0.94 ± 0.40</td>
</tr>
<tr>
<td>Aug</td>
<td>0.55 ± 0.28</td>
<td>0.85 ± 0.45</td>
<td>0.10722</td>
<td>83.04 ± 10.15</td>
<td>0.89 ± 0.34</td>
</tr>
<tr>
<td>Sep</td>
<td>0.40 ± 0.17</td>
<td>0.83 ± 0.42</td>
<td>0.05778</td>
<td>74.33 ± 7.71</td>
<td>0.66 ± 0.30</td>
</tr>
<tr>
<td>Oct</td>
<td>0.67 ± 0.42</td>
<td>0.86 ± 0.43</td>
<td>0.03761</td>
<td>52.11 ± 5.60</td>
<td>0.46 ± 0.21</td>
</tr>
<tr>
<td>Nov</td>
<td>0.68 ± 0.43</td>
<td>0.9 ± 0.41</td>
<td>0.01513</td>
<td>48.02 ± 4.40</td>
<td>0.38 ± 0.14</td>
</tr>
<tr>
<td>Dec</td>
<td>0.52 ± 0.28</td>
<td>0.91 ± 0.43</td>
<td>0.00837</td>
<td>35.39 ± 3.77</td>
<td>0.27 ± 0.23</td>
</tr>
</tbody>
</table>

**Estimation of Aerosol Cloud Interaction (ACI)**

Aerosol induced change in cloud properties and cloud radiative forcing can be understood in three different components resulting from changes in droplet size and its concentration (Twomey effect/first indirect effect), liquid/ice water path (second indirect effect), and cloud fraction. So far it is difficult to distinguish these three components but they characterize the respective changes in physical cloud properties. The aerosol cloud interaction (ACI) effect on cloud droplet effective size is estimated using Eq. (1) for both phase of clouds, which is the principle source of uncertainties in the climate model. Fig. 5a illustrates the mean seasonal variation of ACI for liquid and ice clouds. The result reveals the aerosol cloud interaction vary substantially as a function of time. The ACI is found to be positive for ice cloud during pre-monsoon and monsoon, while it is negative during winter and post-monsoon (Fig. 5a). The positive ACI for ice cloud indicates the long-range transportation of dust aerosol from the arid and semi-arid regions, serves as an ice nuclei (IN), forms the ice cloud with more number of ice crystals and less precipitation efficiency, and in turn the high reflection of solar radiation. The ACI is observed to be maximum during pre-monsoon (~0.17 ± 0.06) for ice clouds (Table 2), when high concentration of dust aerosols (Fig. 4) in the ambient atmosphere. Supplementary Fig. 1. shows the CALIPSO observation of vertical feature mask, aerosol types and cloud types present over Dehradun during 14th May 2013, shows observational evidence of presence of dust induced ice clouds.

The negative ACI for both phase of clouds during winter...
and post-monsoon indicates the absence of aerosol-cloud interaction due to lack of cloud condensation nuclei (CCN)/ice nuclei (IN). It may be also possible that the less moist environment (Fig. 1) prevents the hygroscopic growth of fine mode polluted aerosols, inhibiting cloud formation. The non-hygroscopic nature of dust aerosol can be a reason for negative ACI in liquid cloud during pre-monsoon. There are two possible reasons for the positive value of ACI in the case of liquid clouds during monsoon. First, the presence of favourable meteorological environments (high relative humidity) during monsoon increase the growth of cloud droplets, which supports the formation of clouds. Second the hygroscopic growth of anthropogenic aerosol coated dust aerosol in the vicinity of moist environment can act as a good CCN, enhancing the cloud formation. Though it is difficult to decouple the individual impacts on cloud properties. However, further study, to investigate the meteorological influence on aerosol-cloud interaction is necessary, because untangling the response of aerosol on cloud properties with meteorological influences are not the correct approach to study the aerosol-cloud-climate interaction. The external mixing of dust with anthropogenic aerosol lead to decrease in ACI in monsoon compared to pre-monsoon for ice cloud. The high concentration of dust particles serves as ice nuclei.

Fig. 3. Percentage contribution of each aerosol types to the total in each season over Dehradun (MA = mixed aerosol, DD = desert dust, AA = anthropogenic aerosol, BB = biomass burning).

Fig. 4. SPRINTARS simulated monthly variation of aerosol types (dust, sulfate, carbonaceous (black and organic) and sea-salt) over Dehradun for the period of 2011–2013.
in cloud, which modifies the ice cloud microphysical and radiative properties compared to more pristine conditions. For the detail analysis, we have monthly analyzed the ACI variation particularly during dust period (pre-monsoon and monsoon; Fig. 5(b)). The monthly variation of ACI describes the gradual increase in ACI values (Fig. 5(b)) for ice cloud with an increase in dust concentrations (Fig. 4) from March to May, which is more pronounced in May (~0.24 ± 0.05; Table 3), indicates the dust serve as good ice nuclei, and reinforce the cloud formation. It is gradually decreased in monsoon with decreased in dust aerosols in the atmosphere. In case of liquid cloud, the hygroscopic growth of aerosols in the vicinity of moist environment modifies the cloud properties during June-August. So dust aerosols influence the ice cloud properties in great extent in the absence of favorable meteorological conditions in pre-monsoon, whereas during monsoon season, both aerosol and meteorology have an influence on cloud properties and it is difficult to decouple individually.

Several studies report ACI estimates over different places of the world. The ACI values reported in this study are in agreement with the earlier studies. Pandithurai et al. (2009) reported the indirect effect values of 0.03–0.18 over the East China Sea region. Feingold et al. (2003) reported indirect effect values of 0.07–0.11 on different observational days over Oklahoma using ground-based method. Few studies reported negative indirect values in certain environmental conditions (Myhre et al., 2007; Yuan et al., 2008). Panicker et al. (2010) reported indirect effect values for the two contrasting monsoon seasons in India. It is found that the aerosol indirect values are negative (~–0.007 and –0.22 for different regions) in active monsoon and positive (0.15–0.31) in break monsoon.

Dust-induced Cloud Properties

Cloud microphysical properties are intrinsically linked to cloud macrophysical characteristics through dynamic and thermo-dynamic processes. As the primary focus of this study to investigate the dust impact on cloud properties, we have limited our further analysis for the period of March-July, where the dust is dominant species over Dehradun (Fig. 4). To investigate the AIE, the cloud parameters (e.g., liquid/ice droplet radius, liquid/ice water path and liquid/ ice cloud optical depth) are classified into ten bins of AOD from 0.0–1.0 with an interval of 0.1. Fig. 6 describes the correlation between AOD and droplet effective radius, water path and cloud optical depth of both phase of clouds. The analysis shows the significant response of dust to the ice cloud. It is observed that the increase in AOD decrease the ice cloud effective radius (ICER), but it does not have any response to the liquid cloud effective radius (LCER). The decrease in ICER is more pronounced during dust period, indicates the strong ice nucleation due to long-range transportation of dust (Chylek et al., 2006; Myhre et al., 2007; Dipu et al., 2013). The good correlation (~–0.847)

Table 2. Seasonal mean variation of aerosol-cloud interaction (ACI) for ice and liquid clouds over Dehradun.

<table>
<thead>
<tr>
<th>Season</th>
<th>ACI (Ice Cloud)</th>
<th>ACI (Liquid Cloud)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Winter</td>
<td>–0.16 ± 0.04</td>
<td>–0.03 ± 0.01</td>
</tr>
<tr>
<td>Pre-monsoon</td>
<td>0.17 ± 0.06</td>
<td>–0.05 ± 0.02</td>
</tr>
<tr>
<td>Monsoon</td>
<td>0.12 ± 0.04</td>
<td>0.09 ± 0.04</td>
</tr>
<tr>
<td>Post-Monsoon</td>
<td>–0.08 ± 0.03</td>
<td>–0.02 ± 0.01</td>
</tr>
</tbody>
</table>
Table 3. Monthly mean variation of ACI for ice and liquid clouds along with SWCRF, LWCRF and NETCRF over Dehradun.

<table>
<thead>
<tr>
<th>Month</th>
<th>ACI (Ice Cloud)</th>
<th>ACI (Liquid Cloud)</th>
<th>SWCRF</th>
<th>LWCRF</th>
<th>NETCRF</th>
</tr>
</thead>
<tbody>
<tr>
<td>March</td>
<td>0.09 ± 0.02</td>
<td>-0.08 ± 0.04</td>
<td>-24.09 ± 11.54</td>
<td>15.83 ± 6.47</td>
<td>-8.26 ± 6.52</td>
</tr>
<tr>
<td>April</td>
<td>0.18 ± 0.03</td>
<td>-0.05 ± 0.03</td>
<td>-35.37 ± 13.87</td>
<td>23.45 ± 10.67</td>
<td>-11.91 ± 9.52</td>
</tr>
<tr>
<td>May</td>
<td>0.24 ± 0.05</td>
<td>-0.03 ± 0.02</td>
<td>-61.75 ± 15.78</td>
<td>34.89 ± 13.53</td>
<td>-26.86 ± 11.25</td>
</tr>
<tr>
<td>June</td>
<td>0.2 ± 0.04</td>
<td>0.12 ± 0.05</td>
<td>-41.25 ± 13.25</td>
<td>25.56 ± 10.25</td>
<td>-15.69 ± 10.52</td>
</tr>
<tr>
<td>July</td>
<td>0.12 ± 0.04</td>
<td>0.09 ± 0.03</td>
<td>-18.26 ± 8.52</td>
<td>12.52 ± 5.26</td>
<td>-5.70 ± 4.25</td>
</tr>
</tbody>
</table>

Fig. 6. Variation in particle radius, water path and cloud optical depth as a function of AOD for both the phase of clouds (liquid and ice).

between ICER and AOD in the dust period, indicating the increase in dust (30%–50%) reduces the ICER (40%). The smaller ICER with an increase in AOD associated to the “first aerosol indirect effect” and “second aerosol indirect effect” would enhance the cloud optical depth and cloud albedo. The non-hygroscopic nature of dust particle prevents to act as a cloud condensation nuclei (CCN) resulting in no distinct impact on liquid cloud microphysics (correlation ~0.071).

To quantify the second aerosol indirect effect is the challenging task, where the cloud lifetime increases with the increase in liquid/ice water path and cloud optical depth known as a cloud lifetime effect. Figs. 6(b) and 6(c) illustrate the variation in water path and cloud optical depth as a function of AOD for liquid and ice clouds during dust period (March–July). The more and smaller ice crystals have less chance to coalescence and increase the ice cloud optical depth (ICOD) with an increase in ice water path (IWP) in the clouds. The high correlation (0.876 and 0.918) between AOD with IWP and ICOD indicates the positive response of dust aerosol on ice cloud. The extra latent heat to invigorate the growth of clouds, enhance the water path increase the corresponding cloud optical depth (Fig. 6(c)) and prolong the cloud lifetime (Albrecht, 1989). The aerosol effects on liquid/ice water path, precipitation and cloud lifetime, in general are known as second aerosol indirect effect (cloud lifetime effect; Albrecht, 1989). In general, the aerosol semi-direct effect associated with aerosol absorption of sunlight, which leads to heating of the atmosphere and evaporates the ice crystals in the dust induced ice clouds, which reduce the cloud optical depth and cloud coverage for ice cloud (Hansen et al., 1997). In the present study, the increase in cloud optical depth with small and more ice crystal increases the cloud coverage (Table 1) as per the second indirect effect over Dehradun during the dust period, which may be an influence of meteorological variables. It is observed that the substantial increase in relative humidity (RH) due to the onset of Indian summer monsoon, which reinforces the cloud formation. Mahowald and Kiehl (2003) reported the negative correlation between the high cloud amount and dust along the equator across North Africa and the Atlantic in a region of a relatively high large cloud amount. They also suggested that there was a positive anomaly between desert dust and ice phase cloud to the west, indicating a shift in the location of ice clouds, although there might be a net increase in the ice cloud cover. Therefore, the aerosol impacts on cloud properties are not independent of the meteorological environment. Untangling the two may not be a fully correct approach for the investigation of aerosol effects, as they may include the meteorological response. This needs further investigation with the high quality of air-borne and ground-based observations.

Dust-induced Cloud Radiative Forcing

Aerosols in the troposphere can modify the cloud microphysical properties and in turn their reflectivity, thereby exerting a radiative influence on climate (SCEP, 1970). This section describes the observational evidence of change in cloud radiative forcing by dust induced ice clouds during dust period over Dehradun.

Fig. 7(a) describes the monthly mean variation of shortwave cloud radiative forcing (SWCRF), longwave cloud radiative forcing (LWCRF) and net cloud radiative forcing (NETCRF) at TOA over Dehradun. The result
Fig. 7. (a) Monthly variation of SWCRF, LWCRF and NETCRF over Dehradun during dust period. The scatter plot between NETCRF as a function of (b) liquid cloud optical depth and (c) Ice cloud optical depth.

reveals the positive response of dust to the SWCRF and LCRF. The gradual increase in both SWCRF and LWCRF by modifying the cloud properties during March (−24.09 ± 11.5/15.83 ± 6.47 W m⁻²) to May (−61.75 ± 15.78/34.89 ± 13.53 W m⁻²) with an increase in dust concentration (Table 3). However, the decrease in dust may decrease the SWCRF and LWCRF during May (−61.75 ± 15.78/34.89 ± 13.53 W m⁻²) to July (−18.52 ± 8.52/12.52 ± 5.26 W m⁻²). Dust modifies the radiation budget via direct, indirect and semi-direct effects and it is difficult to distinguish those using satellite observations, however it needs further investigation. The reduction of ICER in the dust induced ice cloud increase the ice crystals and cloud coverage, which reflects more solar radiation back to the space and cools the surface (known as cloud albedo effect), resulting in the increase in SWCRF over Dehradun. The dust induced greenhouse effect results in the increase in LWCRF. In general, dust is not that sensitive to LWCRF (Su et al., 2008), but the heterogeneous nucleation of dust shifts the occurrence distribution of ice clouds to a lower altitude. The reduction in occurrence frequency, cloud top height or increase in emissivity can increase the outgoing longwave radiation, which cools the atmosphere resulting in the increase in LWCRF. The magnitude of SWCRF is observed to be large compared to LWCRF, resulting in the negative NETCRF. For smaller dust particles, which scatter solar radiation, smaller absorption would be produced in the atmosphere, leading to a negative cloud radiative forcing at TOA (Gu et al., 2006). The negative NETCRF during dust period (−13.68 W m⁻²) indicates the cooling effect at TOA.

Aerosol first and second indirect effects enhance the cooling effect, whereas the aerosol-induced cloud warming is mainly the result of the dust induced semi-direct effect. Figs. 7(b) and 7(c) compare the NETCRF at TOA with the liquid cloud optical depth and ice cloud optical depth, respectively, to distinguish the contribution from both phases of clouds to the NETCRF. It is observed that the increase in ice cloud optical depth increase the magnitude of NETCRF, whereas there is no correlation between liquid cloud optical depth and NETCRF. The high correlation (~0.781) between NETCRF and ice cloud optical depth reveals the dust induced ice clouds are contributing more to the radiative budget over Dehradun. The aerosol indirect and semi-direct effects due to dust induced ice clouds play an important role to modify the cloud dynamics and radiative properties.

CONCLUSIONS

The aerosol characteristics over Dehradun and its interaction with both liquid and ice clouds are studied using MODIS derived aerosol and cloud properties. CERES derived TOA flux measurements are used to understand the associated changes in cloud radiative forcing for the dust period. The main conclusions of our study are summarized as follows:

Dehradun is characterized by significant seasonal variability and the dominance of aerosols of different size and composition. The site is dominated by long-range transportation of dust during pre-monsoon and monsoon
resulting in the decrease in $\alpha$. The relative increase in $\alpha$ during winter and post-monsoon, indicates the influence of pollution over Dehradun. Aerosol type analysis (68.98% and 56.15%) and SPRINTARS simulated aerosol species (30%–60%) confirms the large contribution of dust during pre-monsoon and monsoon.

Seasonal aerosol cloud interaction (ACI) is estimated for low level liquid and ice clouds over Dehradun and surroundings. The loading of dust serves as an ice nuclei resulting in the positive ACI for ice clouds during pre-monsoon (-0.17 ± 0.06) and monsoon (-0.12 ± 0.04). The meteorological influence along with anthropogenic aerosol coated dust-cloud interaction leads to the positive values of ACI (~0.09 ± 0.04) for liquid clouds during monsoon, whereas the non-hygroscopic nature of dust resulting in the negative ACI (~−0.08 ± 0.04) for liquid clouds in pre-monsoon. The high correlation between aerosol optical depth and ice cloud properties indicates the positive response of dust to the ice cloud, but it is difficult to make firm conclusions regarding ice cloud and dust aerosol interactions, where both dust and cloud changes are driven by same meteorological conditions.

The reduced ice crystal effective radius due to the aerosol first indirect effects reflects more solar radiation back to the space. The aerosol indirect effects enhance the cloud cooling, whereas the semi-direct effect of dust lead to the aerosol-induced cloud warming over Dehradun and surroundings. The results will be useful to model the aerosol-cloud interactions and its impact to the climate for high altitude site in the climate models.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aqr.org.

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