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Attribution of aerosol radiative forcing over India during the winter monsoon to emissions from source categories and geographical regions

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ABSTRACT

We examine the aerosol radiative effects due to aerosols emitted from different emission sectors (anthropogenic and natural) and originating from different geographical regions within and outside India during the northeast (NE) Indian winter monsoon (January–March). These studies are carried out through aerosol transport simulations in the general circulation (GCM) model of the Laboratoire de Météorologie Dynamique (LMD). The model estimates of aerosol single scattering albedo (SSA) show lower values (0.86–0.92) over the region north to 10°N comprising of the Indian subcontinent, Bay of Bengal, and parts of the Arabian Sea compared to the region south to 10°N where the estimated SSA values lie in the range 0.94–0.98. The model estimated SSA is consistent with the SSA values inferred through measurements on various platforms. Aerosols of anthropogenic origin reduce the incoming solar radiation at the surface by a factor of 10–20 times the reduction due to natural aerosols. At the top-of-atmosphere (TOA), aerosols from biofuel use cause positive forcing compared to the negative forcing from fossil fuel and natural sources in correspondence with the distribution of SSA which is estimated to be the lowest (0.7–0.78) from biofuel combustion emissions. Aerosols originating from India and Africa-west Asia lead to the reduction in surface radiation (−3 to −8 W m^{-2}) by 40–60% of the total reduction in surface radiation due to all aerosols over the Indian subcontinent and adjoining ocean. Aerosols originating from India and Africa-west Asia also lead to positive radiative effects at TOA over the Arabian Sea, central India (CNI), with the highest positive radiative effects over the Bay of Bengal and cause either negative or positive effects over the Indo-Gangetic plain (IGP).

1. Introduction

Human activities causing release of greenhouse gases and aerosols are the main drivers of climate change (Forster et al., 2007). Anthropogenic aerosols alter the earth's energy balance by changing radiative fluxes both at the surface and at the top of the atmosphere (Bellouin et al., 2005) and hence exert radiative forcing of climate. Aerosols reduce the solar radiation reaching the earth's surface by absorbing and scattering respectively the incoming solar radiation and hence cause negative radiative forcing at the surface. Additionally, aerosols such as black carbon absorb solar radiation resulting in a positive radiative forcing at the top-of-atmosphere (TOA). The change in the radiative flux due to aerosols is a function of the atmospheric loading of aerosols and the sign of radiative forcing at the TOA will depend upon the aerosol single scattering albedo as well as the albedo of underlying surface and the distribution of solar zenith angle, the balance between absorption and scattering being the key. On the other hand, the regional climate response to the variation (spatial and temporal) in aerosol radiative effects will depend on the intensity of radiative–convective coupling between the surface and the atmosphere which could be different during the different seasons over the tropical and non-tropical world regions (Ramanathan and Carmichael, 2008). This could lead to climate response due to aerosols which could be different regionally and globally. In this context, it is necessary to examine the radiative effects due to aerosols on a regional basis.

The recent field campaigns over the Indian Ocean [e.g. the Indian Ocean Experiment (INDOEX) and the Indian Space Research Organisation-Geosphere Biosphere Programme (ISRO-GBP) land- and ship-campaigns] have shown large aerosol negative radiative forcing at the surface and a relatively large atmospheric heating compared to the top-of-atmosphere (Ramanathan et al., 2001;

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Jayaraman et al., 2006). The globally and annually averaged direct aerosol radiative perturbation in the Laboratoire de Météorologie Dynamique General Circulation Model (LMD-ZT GCM) have been discussed by Reddy et al. (2005). Recent modelling studies using region-tagged emissions in the LMD-ZT GCM (Verma et al., 2008) showed that the emissions from different regions have disproportionate effects on the surface and columnar loadings of different aerosol species over the Indian subcontinent and ocean. For instance, the surface black carbon (BC) concentrations are mainly contributed from Indian regions while the BC column is more contributed from Africa-west Asia. Hence it is necessary to understand the radiative effects due to aerosols originating from different emission sectors and geographical regions. Modelling studies carried out to understand impact on global annual mean radiative forcing due to aerosol emissions (Koch et al., 2007) or both aerosol emissions and greenhouse gases (Unger et al., 2009; Aunan et al., 2009) originating from sectors such as, residential, industrial, transport, power, biomass showed different extents of radiative effects at the top-of-atmosphere. Using Goddard Institute for Space Studies (GISS) GCM, Koch et al. (2007) showed a significant influence of inter-regional transport of aerosols on aerosol chemical composition over the Arctic and Atlantic oceanic regions. It is required to understand the radiative effects due to aerosols originating from different geographical regions of the world, including those originating from different parts of India and emission sectors (natural and anthropogenic sources) specifically over the Indian subcontinent and ocean. In the present work, we examine the spatial distribution of aerosol radiative effects over the Indian subcontinent and ocean through aerosol transport simulations in the LMD-ZT GCM with the emissions tagged by geographical regions and fuel sectors corresponding to the northeast (NE) Indian winter monsoon when the Indian subcontinent and ocean has the highest pollution loading potential from anthropogenic sources. The specific objectives of the study include the following: (i) evaluate the aerosol single scattering albedo due to contribution from different emission sectors and geographical regions to assess the relative importance of scattering or absorbing aerosols. (ii) analyse the spatial distribution of aerosol radiative effects from different emission sectors and geographical source regions within and outside India.

2. Method of study

Aerosol transport simulations for the period from January to March 1999 (corresponding to the NE Indian winter monsoon) are carried out in the LMD-ZT GCM, version 3.3. A description of the atmospheric model is given in Li (1999) and a specific description of aerosol treatment and atmospheric transport is given in Boucher et al. (2002) and Reddy et al. (2004). Two sets of experiments were carried out for the present work, where aerosols were either tagged by regions or sources. Details of the experimental setup are given in Verma et al. (2007). Fig. 1 shows the masked regions on GCM zoom grid, which include the Indo-Gangetic Plain (IGP), central India (CNI), south India (SI), northwest India (NWI), southeast Asia (SEA), east Asia (EA), Africa-west Asia (AFWA), and rest of the world (ROW). In the region-tagged simulations, the aerosol transport and atmospheric processes are simulated for each geographical region with the emissions outside that region being switched off. In the source-tagged simulations, the aerosol transport and atmospheric processes are simulated for each of the sector – biofuel (BF), fossil fuel (FF), and natural source. The sectors for the biofuel source included wood and crop-waste for residential cooking and heating. The sectors for the fossil fuel source are coal-fired electric utilities, diesel transport, brick kilns, industrial, transportation, and domestic. The natural source included sulphur from

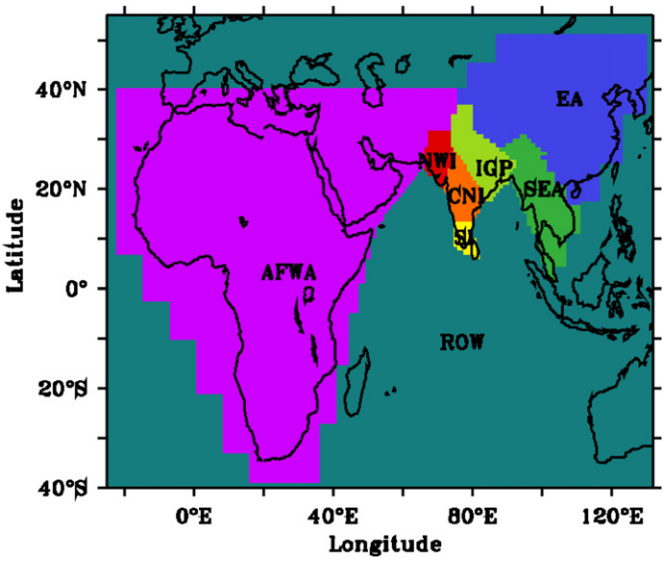

Fig. 1. Masked regions on GCM zoom grid representing tagged source regions taken under study; the classification of source regions are as following with different colours indicated in bracket: 1. IGP (light green) 2. CNI (orange), 3. SI (yellow), 4. NWI (red), 5. SEA (green), 6. EA (blue), 7. AFWA (pink), 8. ROW (dark green). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

volcanic and biogenic sources, terpenes from the vegetation or natural OM, dust from arid regions, and sea-salt. Aerosol emissions over India [SO_2, BC, and organic matter (OM), and inorganic matter (IOM) (fly-ash emissions from coal combustion)] are from the high-resolution ($0.25^\circ \times 0.25^\circ$) emission inventories of Reddy and Venkataraman (2002a, 2002b). Emissions of SO_2, BC, and organic carbon (OC) from fossil fuel and biomass sources over Asia are from Streets et al. (2003). Global carbonaceous aerosol emissions used here are the same as described by Reddy and Boucher (2004). Carbonaceous aerosols are predominantly emitted in the hydrophobic form, but some fraction of the emissions may be in hydrophilic form as well. Here, we assume that black carbon (BC) emissions from both fossil fuel and biomass burning occur as 80% hydrophobic and 20% hydrophilic, whereas organic matter (OM) emissions occur as 50% hydrophobic and hydrophilic. The ageing process of BC and OM is represented by a transfer of the hydrophobic to hydrophilic form with an exponential lifetime of 1.63 days. Aerosol scheme is a mass-only scheme. Aerosol optical properties (mass extinction efficiency, single scattering albedo, and asymmetry factor) for all aerosol species are computed using Mie theory with prescribed size distributions and refractive indices. We consider the relative humidity (RH) effects on particle size and density of sulphate, hydrophilic OM, and sea-salt. The refractive index of sulphate, hydrophilic OM, and sea-salt as a function of RH are calculated as the volume weighted average of the refractive indices of each of the aerosol species and water. Aerosol optical properties for all aerosol species are computed assuming an external mixing.

The radiative code in the LMD-ZT GCM consists of improved versions of the parametrization of Fouquart and Bonnel (1980) (solar radiation) and Morcrette (1991) (terrestrial radiation) as described in Reddy et al. (2005). The model accounts for the diurnal cycle of solar radiation and allows fractional cloudiness to form in a grid box. The reflectivity and transmissivity of a layer are computed using the random overlap assumption (Morcrette and Fouquart, 1986) by averaging the clear and cloudy sky fluxes weighted linearly by their respective fractions in the layer. For each

of the simulation of the region-tagged and source-tagged experimental setup, the radiative fluxes in the shortwave spectrum (0.25–4.00 μm) are computed every 2 h, at the top-of-atmosphere (TOA) and at the surface, with and without the presence of clouds, and with and without the presence of aerosols using a double radiation call at each time-step of the model integration. The all-sky direct aerosol radiative effects can then be estimated as the difference in radiative flux with and without aerosols. The GCM is nudged with European Centre for Medium-Range Weather Forecast (ECMWF) wind fields.

3. Aerosol chemical characteristics over the Indian subcontinent and ocean: scattering vs. absorbing aerosols

A detailed description of evaluation of the model estimates with the observations over Indian ocean and subcontinent have been discussed in our previous work (Reddy et al., 2004; Verma et al., 2008). We present here only a short description of comparison between the mean AOD during the northeast (NE) Indian winter monsoon (January–March) from the GCM estimates during the period of 1999 (Fig. 2a) and the AOD retrieved from MODIS (Moderate Resolution Imaging Spectroradiometer) observations (http://modis.gsfc.nasa.gov/) (Fig. 2b) during the period of 2004 over the Indian subcontinent and ocean. The MODIS AOD data used in the present study are from MODIS Collection 5 Terra Level-3 products, produced globally at a resolution of 1°×1°. These data are cloud screened and have been recommended to be used for scientific investigations. The MODIS data are regridded to the model grid for comparison. The model estimated AOD lies in the range of 0.16–0.24 over the Indian subcontinent and oceanic regions of AS and BoB. The model captures the spatial gradient in AOD over the Ocean showing the higher values near the coastal regions with a decrease as we go away from the coast and show features of high AOD near the eastern part of India as seen in the MODIS data. However, there is a major discrepancy between the modelled and satellite retrieved AOD over the Indo-Gangetic plain (IGP) where the model does not capture the high AOD values as seen in the satellite data. The AOD retrieved from the MODIS is in the range of 0.4–0.5 over the IGP and is in the range of 0.16–0.36 over the rest of India, and oceanic regions of AS and BoB. The modelled AOD is estimated a factor of 2–2.5 lower than the AOD retrieved from MODIS over the IGP, but compares relatively well over the rest of India and the Arabian Sea.

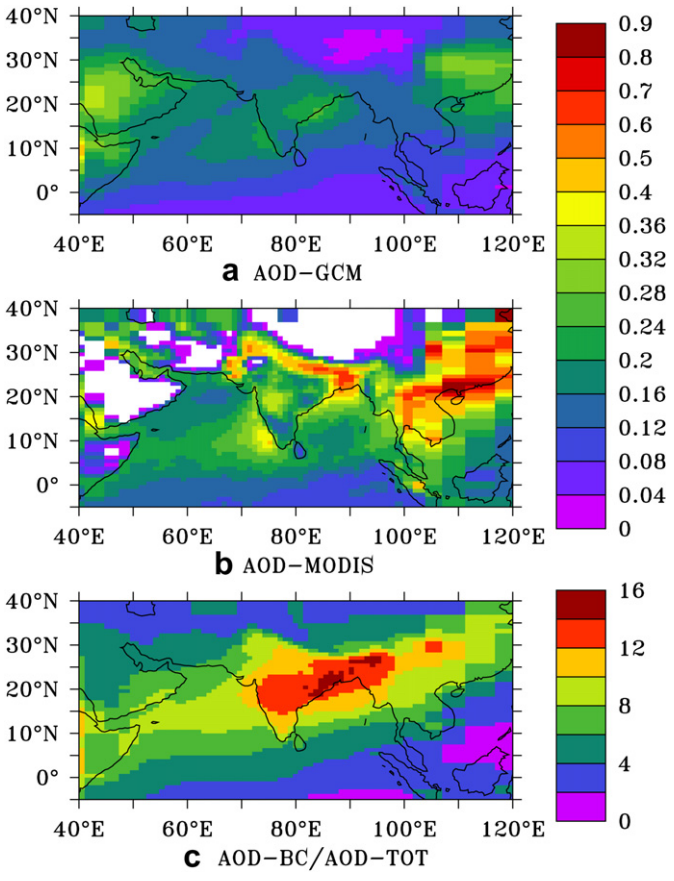

Fig. 2. Spatial distribution of the mean (a) GCM estimated AOD, (b) measured AOD from MODIS, and (c) percentage contribution of GCM estimated AOD-BC to the total AOD during the winter monsoon. The white space in the Fig. 2b represents the missing data.

We have discussed in our previous work (Verma et al., 2008) the probable reasons for model underestimation which was attributed to inaccuracies in ECMWF meteorological analyses (Bonazzola et al., 2001) which are used to nudge the model, but there is likely to be as well an underestimation of aerosol sources in the model. We also believe that the refinement in the spatial distribution of emissions from residential biomass energy use and the spatial and temporal distribution of emissions from open burning of forest and agricultural biomass may help in closing some of the discrepancy between model predictions and measurements. The possible inaccuracies in the retrieval techniques of the MODIS AOD, including the possibility of lack of efficacy of aerosol model used in the retrieval algorithm (Misra et al., 2008; Levy et al., 2010; Hyer et al., 2010) could also be one of the reasons for a higher discrepancy between the modelled and the MODIS AOD, specifically over the IGP. The ocean retrievals of the MODIS AOD have been shown to be more accurate than the land retrievals and MODIS retrievals over land may be systematically biased high (Remer et al., 2005). A comparison of AOD data from the MODIS with the other ground based observations and the evaluation of measurements with the multiple modelling platforms (Verma et al., 2011, in preparation) can provide a valuable insight into the existing discrepancies between the model estimates and measurements over the Indian subcontinent.

Recently, we have also evaluated the source region information from the GCM estimates with the back-trajectory analysis for the days and locations of Sagar Kanya during its INDOEX-IFP cruise. The regional influence evaluated through region-tagged model estimates corroborated the probable source region evaluated through back-trajectory analysis (Verma et al., 2007). The model estimates were used to assess the origin of surface and columnar INDOEX aerosols from natural or anthropogenic sources, and their transport from adjoining regions or distant geographical regions (Verma et al., 2008). In our previous work, aerosol transport simulations in LMD-ZT GCM have been used as a valuable tool to understand the chemical species composition and the source region evaluation to the aerosol measurements during the INDOEX. In the present study, we investigate the impact on aerosol radiative effects due to emissions originating from the different sectors and geographical regions over the Indian subcontinent and ocean.

Black carbon (BC) contributes 10–16% to the total AOD (Fig. 2c) over the Indian Ocean and subcontinent and the balance of the total AOD is contributed largely by scattering aerosol constituents (mainly composed of sulphate and organic matter from anthropogenic sources). Although, the relative contribution of BC to the total AOD is low as compared to the other aerosol constituents, the presence of trace amounts of BC can cause contrast in the sign (positive or negative) of aerosol radiative effects at TOA. This is because the radiative forcing due to BC (absorbing aerosol) differs

from that of non-BC, e.g. sulphate and organic matter as discussed in the Section 1.

We evaluate the spatial distribution of the aerosol single scattering albedo (ratio of scattering coefficient to the sum of scattering and absorption coefficients) at 550 nm to understand the relative distribution of absorbing and scattering aerosols over the Indian subcontinent and ocean. The spatial distribution of the mean aerosol single scattering albedo during the winter monsoon due to emissions from biofuel use, fossil fuel, and natural are shown in the Fig. 3. There exists a spatial variation in the estimated aerosol SSA due to emissions from a source. This is because of the spatial variation in the distribution of surface mass concentration and AOD of BC and non-BC aerosol constituents (including organic matter and sulphate) arising from the spatial distribution of sectorial emissions of a source category. The different sectors corresponding to a source category are discussed in the Section 2. The spatial distribution of aerosol SSA due to emissions from biofuel use as presented in the Fig. 3a shows lower values (0.7–0.78) of SSA over India and adjoining coastal regions of the Arabian Sea and Bay of Bengal with a patch of the lowest value (0.7–0.72) over the east coast of India. Biofuel (BF) use for residential biofuel combustion is one of the main sources of BC emissions (Habib et al., 2008) over the region and have a higher ratio of BC (absorbing aerosol) to non-BC aerosol constituents (mainly due to organic matter, sulphate) compared to the other source categories (fossil fuel combustion, natural), with their highest impact over the east coast of India comprising of Bihar and West Bengal zones.

The BC surface concentration and AOD due to BC over the Indian subcontinent and ocean are significantly contributed from biofuel combustion (Verma et al., 2008). Fossil fuel (FF) combustion (Fig. 3b) leads to SSA of 0.92–0.98 over most parts of the Indian subcontinent with the patch of a lower value (0.9–0.92) existing over the Indo-Gangetic Plain. Fossil fuel combustion is the main contributor of sulphate aerosols over the Indian subcontinent and ocean. However, emissions of carbonaceous aerosols, including BC and organic matter (OM) from fossil fuel combustion results in a slightly lower aerosol SSA from fossil fuel combustion emissions over the Indian subcontinent as compared to the oceanic regions. The spatial distribution of SSA due to natural sources show values of 0.98–0.984 over the Indian subcontinent due to the transport of dust and natural OM while even larger values (SSA $>$ 0.99) are estimated over the tropical Indian ocean region due to the importance of sea-salt aerosols.

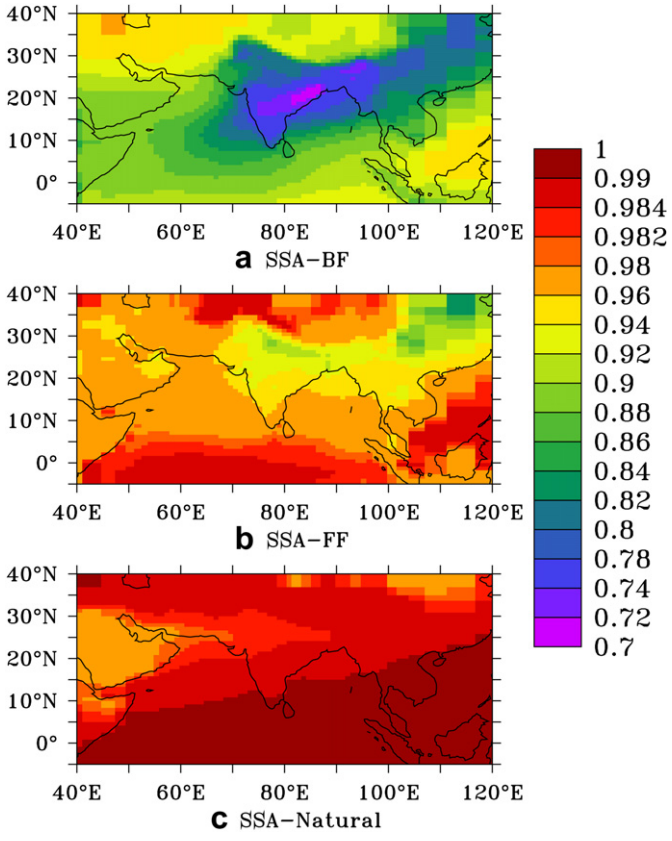

Fig. 3. Spatial distribution of the mean aerosol columnar SSA at 550 nm during the winter monsoon from (a) biofuel sources, (b) fossil fuel sources, and (c) natural sources.

The model estimated mean aerosol single scattering albedo during the winter monsoon from simulations with region-tagged emissions is shown in the Fig. 4. The total SSA (i.e. with emissions from all the regions) as presented in the Fig. 4a shows the presence of lower SSA values (0.86–0.9) over the Indian subcontinent compared to the adjoining oceanic regions of AS and BoB (0.88–0.94) and lies in the range 0.94–0.98 over the region south to 10°N. The SSA estimated from the present study is consistent with the SSA values inferred based on aerosol measurements on various platforms over the Indian Ocean and subcontinent. The modelled SSA over the AS based on aerosol data collected using ship-borne and island platforms from 1995 to 2002 averaged during November to March was 0.94 at 500 nm (Satheesh et al., 2006) and over BoB was deduced to be between 0.84 and 0.96 at 550 nm (Nair et al., 2008). The SSA estimated from measurements over the Indian subcontinent, including stations over central India during February 2004 was found to be 0.84 at 525 nm (Jayaraman et al., 2006). The SSA estimated from aeronet observations during December 2004 at Kanpur (north India) at 440 nm was 0.94 and 0.87 at ambient RH and for dry aerosols respectively (Tripathi et al., 2006).

The lower SSA estimated over the Indian subcontinent and the adjoining oceanic regions of AS and BoB is attributed to impact from aerosols originating due to emissions from the Indian subcontinent itself and Africa-west Asia which mainly contribute to the aerosol optical depth. The aerosol SSA due to emissions originating from Indian regions (Fig. 4b) lie in the range 0.82–0.92 over the Indian subcontinent, Bay of Bengal, and parts of the Arabian Sea which is lower than SSA due to aerosols originating from Africa-west Asia (0.88–0.92, Fig. 4c) and rest of the world ($>$0.96, Fig. 4d). The lower value of aerosol SSA estimated due to the emissions from the Indian subcontinent (Fig. 4a) is mainly attributed to emissions from the biofuel use (discussed in previous paragraph) prevalent over the Indian subcontinent. This is consistent with our previous study (Verma et al., 2008) which showed the surface BC mass concentration over the Indian subcontinent and ocean to be significantly influenced due to emissions from biofuel sources from India (mostly from Indo-Gangetic plain). Aerosols originating due to emissions from Africa-west Asia arise largely from open biomass burning which influence BC loading in the higher atmospheric layers but chiefly contribute to OM (organic matter) over the Arabian Sea, Indian subcontinent, Bay of Bengal (Verma et al., 2008) and hence have a slightly higher SSA compared to the aerosols from the Indian subcontinent. The SSA ($>$0.98) due to emissions from rest of the world is mainly due to sea-salt aerosols south to 10°N and a slight decrease in SSA (0.96–0.98) is due to the presence of sulphate (from natural sources and long-range transport from fossil fuel combustion) and dust re-circulated over the Indian subcontinent and ocean. We evaluate the impact on the aerosol radiative effects due to the spatial variation in the aerosol single scattering albedo in Section 4.

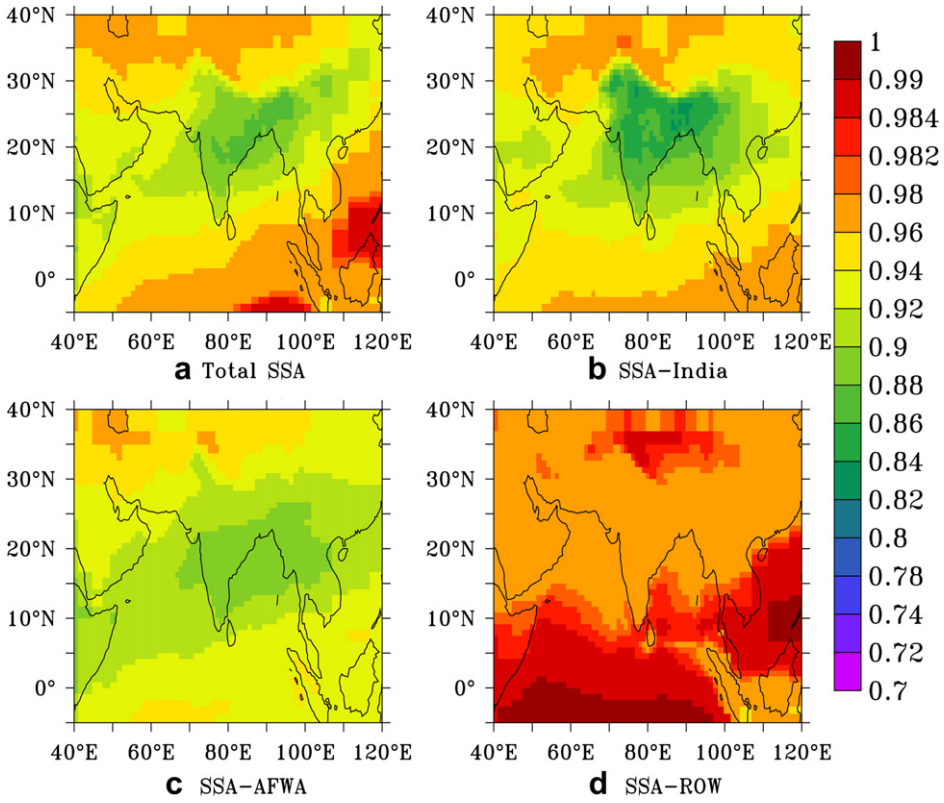

Fig. 4. Spatial distribution of the mean SSA at 550 nm during the winter monsoon from (a) all regions, (b) India, (c) Africa-west Asia, and (d) rest of the world.

4. Aerosol radiative effects over the Indian subcontinent and ocean

4.1. Impact due to emissions originating from natural and anthropogenic sources

The spatial distribution of the GCM estimated mean all-sky direct aerosol radiative effects during the NE Indian winter monsoon due to emissions from anthropogenic sources (biofuel use, fossil fuel combustion), and natural sources at the surface, top of the atmosphere, and within the atmosphere are shown in the Fig. 5. Aerosols from all the emission sectors exert negative radiative effects at the surface (left column of the Fig. 5) indicating reduction in the radiation at the surface. While aerosols originating from the natural sector lead to the reduction in the surface radiation by 0.5–1 W m^{-2}, aerosols from fossil fuel combustion and biofuel combustion lead to the reduction by 1–5 W m^{-2} over most of the parts of the Indian subcontinent, AS, and BoB. The emissions from fossil fuel combustion reduce the surface radiation by a factor 6–8 times more, compared to 2–6 times only from biofuel, than the aerosols from the natural sector over most of the parts of the Indian subcontinent and ocean.

At the TOA (middle column of the Fig. 5), the all-sky direct aerosol radiative effects from biofuel differ from fossil fuel and natural sources in terms of sign. The all-sky direct aerosol radiative effects at the top-of-atmosphere is negative due to the emissions from fossil fuel (−1 to −2 W m^{-2}) combustion and natural (−0.2 to −0.5 W m^{-2}) sector. In contrast, the emissions from biofuel combustion lead to a positive forcing (0.5–1 W m^{-2}) everywhere with values as large as 1–2 W m^{-2} over the Bay of Bengal, some parts of the Arabian Sea, and over central India. This follows from the lower aerosol SSA estimates from BF emissions compared to the fossil fuel and natural as discussed in the Section 3. We analyse the positive TOA aerosol radiative effects in the context of the transport features of BC later in the subsequent paragraphs. The positive value of mean aerosol radiative effects during the NE winter monsoon at TOA from the biofuel source as estimated in the present study is in corroboration with the study by Koch et al. (2007). Using the GISS GCM, the annual mean aerosol radiative forcing at TOA (Koch et al., 2007) from residential fuel sector emissions averaged over the globe was estimated to be +0.04 W m^{-2}. The spatial distribution of annual mean aerosol radiative forcing at the TOA showed values to be between 0 and 0.3 W m^{-2} over the Indian subcontinent (Koch et al., 2007). At present there is a disagreement on the net radiative effect of emissions from the residential cooking sector with some studies claiming the annual mean radiative forcing to be a net negative at TOA (Aunan et al., 2009), but others a net positive (Koch et al., 2007; Unger et al., 2009). The present study indicates a larger positive TOA forcing from the residential cooking sector than the previous estimates (Koch et al., 2007; Unger et al., 2009) over the Indian subcontinent and ocean. One possible reason could be due to the period of averaging in the present study which is NE winter monsoon when the Indian subcontinent and ocean has the highest pollution loading potential from anthropogenic sources. The other possible reason could be the lower amount of organic carbon (OC) emissions from the residential sector in the present study using the inventory of Reddy and Venkataraman (2002b) over India than in the previous studies, using the inventory of Bond et al. (2004). This leads to a higher BC/OM ratio in the emissions over India from the residential sector in the present study (BC/OM = 0.33) compared to that in the previous studies (BC/OM = 0.17).

The atmospheric forcing is estimated as the difference between the radiative effects at the top-of-atmosphere and the surface. As

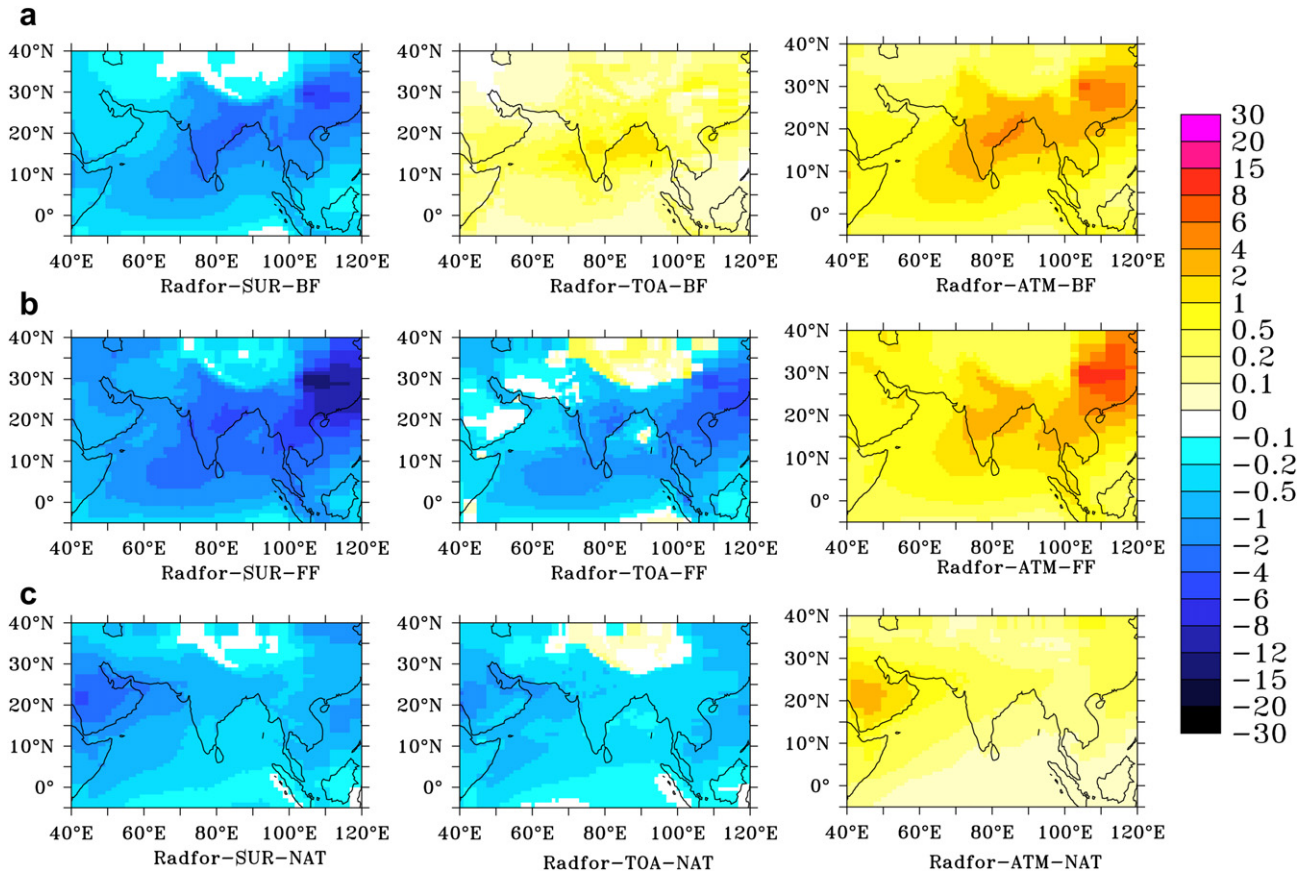

Fig. 5. Spatial distribution of the mean shortwave all-sky aerosol radiative forcing during the winter monsoon at the surface (left column), top of the atmosphere (middle column), and atmosphere (right column) from biofuel (top row), fossil fuel (middle row), and natural (bottom row).

shown in the Fig. 5 (right column), aerosols from anthropogenic emissions arising from biofuel combustion for energy and fossil fuel combustion lead to a higher positive forcing within the atmosphere (2–5 W m^{-2}), which is a factor of about 10 times more than the positive forcing from natural aerosols (0.2–0.5 W m^{-2}). Interestingly, the aerosol radiative forcing at the atmosphere, due to biofuel emissions (2–5 W m^{-2}), is estimated to be widely spread over most of the parts of the Indian subcontinent, Arabian Sea, and Bay of Bengal, as compared to the fossil fuel emissions (2–3 W m^{-2}) for which the aerosol radiative forcing at the atmosphere is estimated to be more concentrated over the central India and the Indo-Gangetic plain. This is because biofuel being a dispersed source whereas emissions from fossil fuel combustion are mainly from large point sources in industrialized regions (Reddy and Venkataraman, 2002b).

4.2. *Impact due to emissions originating in different geographical regions*

We present the spatial distribution of the GCM estimated mean all-sky aerosol radiative effects during the NE Indian winter monsoon at the surface, top-of-atmosphere, and atmosphere due to the aerosols originating from the different geographical regions within and outside India in the Fig. 6. The topmost row of the Fig. 6 shows estimates of the total mean aerosol radiative effects during the Indian winter monsoon. At the surface, aerosols lead to a decrease in the radiation over the Indian subcontinent by 10–15 W m^{-2} and over the adjoining oceanic regions by 4–10 W m^{-2} (AS: −8 to −10; BoB: −4 to −8 W m^{-2}) with a maximum reduction (−15 to −20 W m^{-2}) present along the east coast of India. The negative aerosol surface radiative forcing over the Indian subcontinent and ocean is mainly due to the emissions originating from the Indian regions (second row first column of Fig. 6) and Africa-west Asia (third row first column of Fig. 6). Aerosols originating from India and Africa-west Asia emissions lead to a reduction of the radiation at the surface by 3–8 W m^{-2} each over most of the parts of the Indian subcontinent and ocean with a maximum reduction (8–12 W m^{-2}) estimated along the east coast of India due to the Indian emissions. There is a small negative radiative effects at the surface of 1–2 W m^{-2} due to the emissions from east Asia which mainly influences the Bay of Bengal. Aerosols originating from rest of the world emissions lead to a negative aerosol surface radiative forcing of about 0.5–1 W m^{-2} uniformly distributed all over the subcontinent and ocean. Overall, aerosols originating from India and Africa-west Asia emissions contribute to the reduction in the incoming solar radiation by 40–60% of the total reduction of surface solar radiation by aerosols over the Indian subcontinent and the adjoining ocean. The surface dimming of solar radiation due to aerosols originating from India has a higher impact over the BoB and the Indian subcontinent. Aerosols originating from India emissions lead to the surface dimming of solar radiation 3–9 times and 6–15 times more than the surface dimming due to aerosols from rest of the world over the BoB and the Indian subcontinent respectively. On the other hand, aerosols due to emissions from Africa-west Asia show a higher impact over the AS and leads to the surface dimming of solar radiation 6–9 times more than the surface dimming due to aerosols from rest of the world.

At the top-of-atmosphere (first row, middle column of Fig. 6), aerosols exert both negative and positive radiative effects north to 10°N. It is to be noted that the negative aerosol radiative effects at the surface as discussed in the previous paragraph also show the maximum impact north to 10°N. These features of aerosol radiative

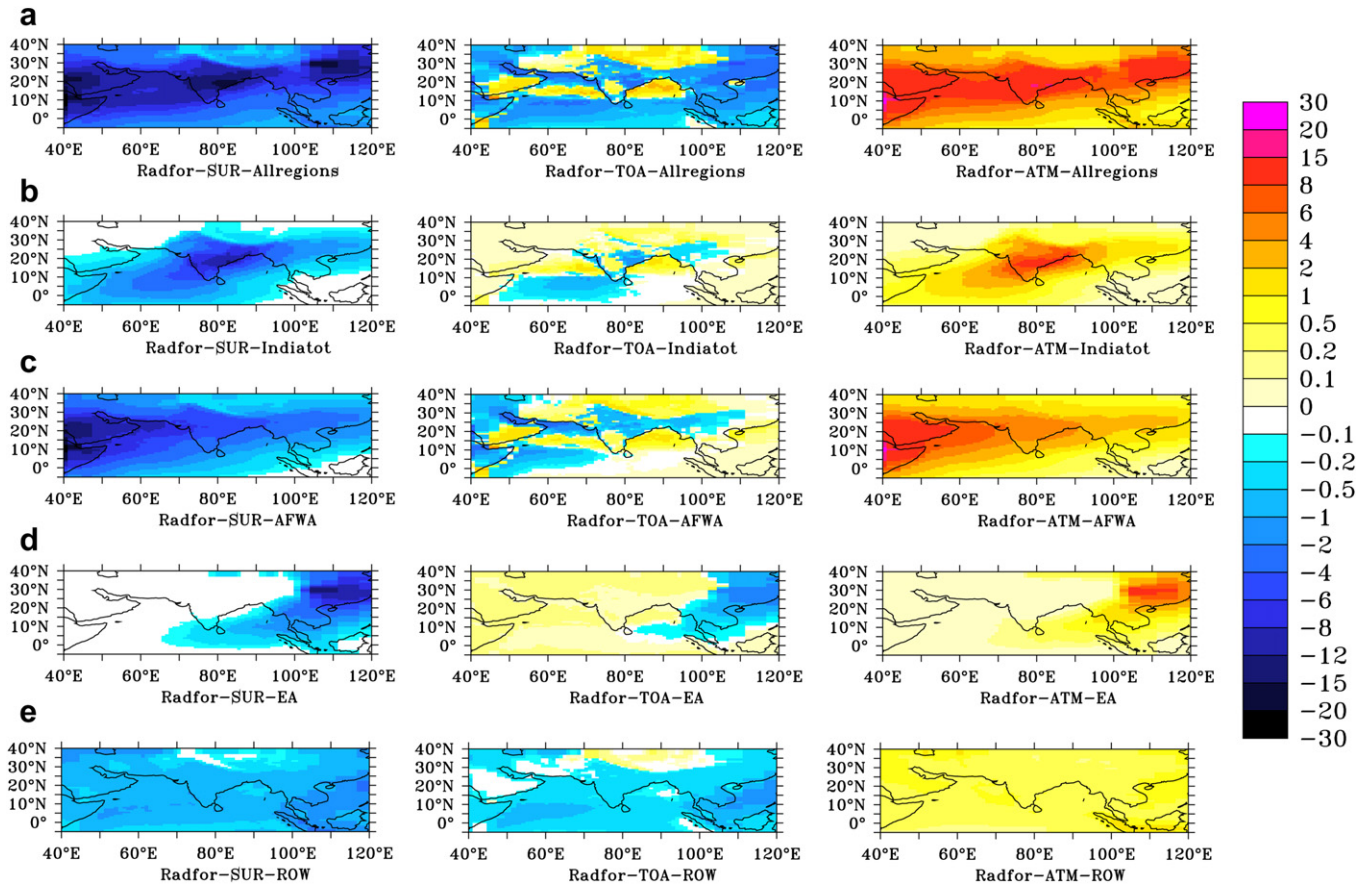

Fig. 6. Spatial distribution of the mean shortwave all-sky aerosol radiative forcing during the winter monsoon at the surface (left column), top of the atmosphere (middle column), and atmosphere (right column) from All regions (first row), Indian regions (second row), Africa-west Asia (third row), east Asia (fourth row), and rest of the world (fifth row).

effects are in correspondence with the spatial distribution of the aerosol SSA (Fig. 4a) showing the presence of lower values of SSA north to 10°N indicating the larger influence from anthropogenic aerosols over the region north to 10°N. The positive aerosol radiative effects of 0.5–1 W m^{-2} at the TOA are seen over most of the parts of the AS, BoB, and the parts of central India, with the values as high as 2–4 W m^{-2} over the BoB. It is interesting to see somewhat higher intensity of positive radiative effects at the TOA over the BoB than the AS, which is due to the presence of higher concentration of BC in correspondence with the lower values of SSA over the BoB than the AS. The aerosol radiative effects at the TOA are estimated to be negative over east and the west tropical Indian oceanic regions (south to 10°N).

As seen from the Figures (middle column of second and third rows of Fig. 6), the positive and negative aerosol radiative effects over the Indian Ocean and the subcontinent are mainly due to the emissions from India and Africa-west Asia. Specifically, emissions from the Indian regions cause positive TOA aerosol radiative effects over the central India, BoB, parts of AS, and both positive and negative effects over the Indo-Gangetic plain. Compared to the emissions originating from the Indian regions, the emissions from Africa and west Asia also lead to a positive TOA aerosol radiative effect over the central India which extend towards south India but cause negative effects over the entire Indo-Gangetic plain. The positive radiative effects correspond with the lower SSA (0.86–0.9) (Fig. 4b and c) estimated due to aerosols originating from the Indian regions and Africa-west Asia. The disparity (positive or negative) in sign of aerosol radiative effects at TOA estimated due to emissions from Indian region and Africa-west Asia is also seen in region-tagged emissions transport simulations in GISS by Koch et al. (2007). The global annual mean radiative forcing at TOA from aerosols arising from south Asia emissions was estimated to be 0.01 with a neutral to slight negative forcing (0–0.3 W m^{-2}) over the Indian subcontinent and ocean (including AS and BoB) in comparison to the aerosol radiative forcing from African biomass emissions which was estimated to be neutral to slight positive (0–0.1 W m^{-2}) or negative (0–0.1 W m^{-2}) over the region (Koch et al., 2007). In contrast, the emissions from rest of the world (Fig. 6) lead to a smaller negative aerosol radiative effects at the TOA which is almost uniformly distributed over the entire Indian subcontinent and ocean. The existence of positive TOA aerosol radiative effect estimated over some parts of the Indian subcontinent and ocean is linked with the transport of BC in elevated atmospheric layers as discussed later in the subsequent paragraphs.

Within the atmosphere (right column of Fig. 6), aerosols trap 8–15 W m^{-2} of solar radiation over the Indian subcontinent and ocean due to the influence from all the regions. The atmospheric absorption of solar radiation over the BoB is estimated to be mainly influenced due to emissions from India, and is about 5–15 times the atmospheric absorption due to aerosols from rest of the world. Over the Indian subcontinent, the emissions from India lead to the atmospheric absorption 10–15 times the absorption due to emissions from rest of the world. On the other hand, the emissions from Africa-west Asia mainly impact the atmospheric absorption of solar radiation over the AS, which is about 10–15 times the atmospheric absorption due to emissions from rest of the world. A small positive radiative effects (0.2–1 W m^{-2}) at the atmosphere is estimated due to the emissions originating from east Asia which mainly influences the Bay of Bengal. The emissions from rest of the world lead to the positive

aerosol radiative effects at the atmosphere of 0.2–0.5 W m^{-2} uniformly distributed all over the subcontinent and ocean.

The spatial distribution of the mean aerosol radiative effects during the NE Indian winter monsoon due to emissions from the different Indian regions is shown in the Fig. 7. We present the estimates of aerosol radiative effects due to emissions originating from the Indo-Gangetic plain (IGP) and central India (CNI) only but do not show the estimates due to emissions originating from south India and northwest India which have small impacts on the radiation. Aerosols originating from the IGP and CNI decrease the surface radiation equally by values as large as 6–8 W m^{-2}. Interestingly, there is a contrast in the aerosol radiative effects at the TOA due to the emissions originating from the Indo-Gangetic Plain and the central India. While the emissions from the IGP lead to the negative radiative effects over parts of the Indian subcontinent and either a negative or almost a neutral effect over the Indian Ocean, the emissions from central India cause positive radiative effects (0.5–1 W m^{-2}) over the central India, Bay of Bengal, and the Arabian Sea along with a smaller negative impacts over the west tropical Indian Ocean. The IGP is dominated by the emissions of sulphur and BC arising mainly from fossil fuel and biofuel use, respectively, and is the main contributor to surface mass concentration of sulphate and BC aerosols over the Indian subcontinent and ocean. Emissions from the central India arising mainly from forest burning (Venkataraman et al., 2006) have a higher impact on the AOD due to BC than they have on surface concentrations. It is to be noted that the open biomass burning is prevalent in central India during January to March, whereas the crop residue burning is in western India during these months (Venkataraman et al., 2006). Within the atmosphere, there is a trapping of solar radiation by 2–6 W m^{-2} over the IGP and CNI due to the emissions arising from these regions. Aerosol radiative effects over the Indian subcontinent due to the influence of emissions from the Indian regions reveal that the control of BC emissions specifically over the IGP and CNI would be helpful to reduce the major part of atmospheric radiation absorption over India.

We discuss here the positive aerosol radiative effects estimated at TOA over some parts of the Indian subcontinent and ocean, as already presented in the previous paragraphs, in the context of aerosol transport features from Indian regions and other regions. The positive radiative effects at the TOA over central India, AS, and BoB shows correspondence with the transport of BC in higher atmospheric layers from central India and Africa-west Asia (Verma et al., 2008) arising from elevated emissions of BC from forest burning in the central part of India as discussed before and from open biomass burning from Africa-west Asia. The BC surface mass concentration over the BoB was estimated to be mainly contributed from Indian regions from the Indo-Gangetic plain over the BoB. Additionally, there is a contribution from lofting of surface emissions of BC above the boundary layer by air mass convergence and lofting, for example, shown along the west coast of India during parts of the INDOEX-IFP (Verma et al., 2006).

It is worth noting the presence of a higher AOD and a lower SSA in the model estimates over the land region (refer Figs. 2 and 3) compared to the oceanic regions of Bay of Bengal and Arabian Sea. However, positive aerosol radiative effects are estimated at the TOA over the BoB and the AS, which is linked with the transport of BC in elevated atmospheric layers. The presence of BC in the atmospheric layers above cloud (surface with higher surface reflectivity than the land) further enhances its absorbing characteristics which is the reason for positive TOA aerosol radiative effects over the oceanic regions of BoB and AS where a high cloud fraction is estimated in the model at the lower levels. It is to be noted that these features of the positive aerosol radiative effects at the TOA are not seen in the model estimates for the clear-sky condition (Figure not shown) over the BoB and AS, but they do exist over central India, west Asia, and over Himalayas as also estimated in the present study for the all-sky condition. Positive radiative effects estimated over central India is due to the presence of lower SSA accompanied by the transport of BC in higher layers from open biomass burning (forest biomass) prevalent in central India during the period of study (Venkataraman et al., 2006). On the other hand, the presence of desert over west Asia and northwest India and that of snow over Himalayas leads to positive aerosol radiative effects at TOA over these regions as aerosols are suspended over surfaces of high surface reflectivity, a feature similar to that seen over Antarctica by Gadhavi and Jayaraman (2004).

The model estimated all-sky radiative effects at the surface(atmosphere) as discussed in the previous paragraphs are slightly underestimated compared to those inferred from measurements as -20 ± 3 W m^{-2} (18 ± 3 W m^{-2}) over the north Indian Ocean (0–20°N) during the INDOEX-IFP (Ramanathan et al., 2001). The model estimated clear-sky aerosol radiative effects (Figure not shown) at the surface (TOA) are within the range of the estimated values from the observations over the Arabian Sea ($-16.2(-6.1)$ W m^{-2}), and over the Indian region ($-19.9 \pm 9(+0.3 \pm 2.5)$ W m^{-2}) (Dey and Tripathi, 2008). However, model estimates do not show values as high as -27 to -30 W m^{-2} at the surface retrieved from measurements over the BoB (Ganguly et al., 2005) and central India (Jayaraman et al., 2006). We believe that the refinement in the estimation of aerosol optical depth as discussed in the Section 3 and a better representation of spatial and temporal distribution of surface albedo in the model may help in closing some of the discrepancy between model predicted aerosol radiative effects and those inferred from measurements. It is

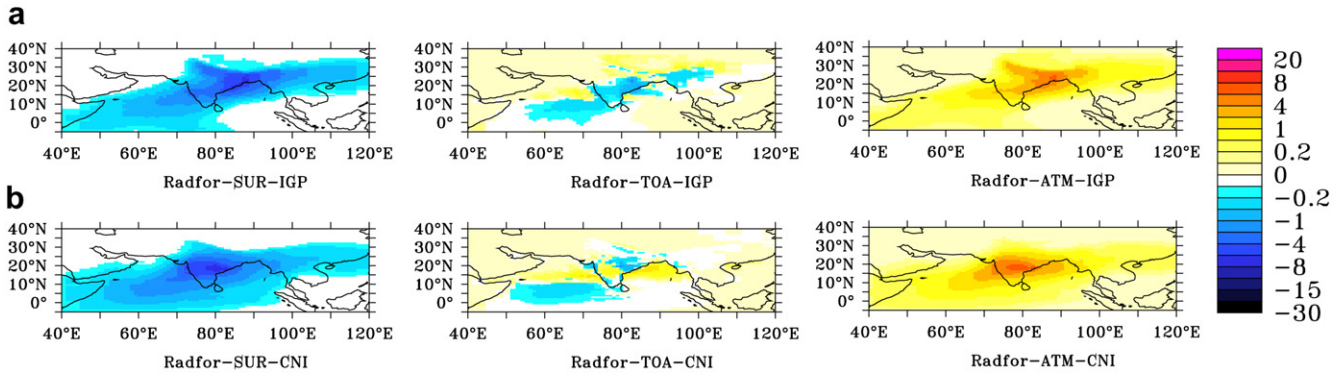

Fig. 7. Spatial distribution of the mean shortwave all-sky aerosol radiative forcing during the winter monsoon at the surface (left column), top of the atmosphere (middle column), and atmosphere (right column) from IGP (first row), and CNI (second row).

interesting to see that the slightly positive values or the lower negative values estimated from the GCM at TOA over the central India and the Indo-Gangetic plain are consistent with the measurements (Dey and Tripathi, 2008; Jayaraman et al., 2006).

In summary, aerosols from the anthropogenic sources, mostly arising from India and Africa-west Asia emissions, perturb the climate by enhancing the dimming of radiation at the surface thereby leading to a surface forcing by a factor of 10–20 times larger over the AS and BoB, and 20–30 times larger over the Indian subcontinent than the aerosols from the natural sources. Within the atmosphere, anthropogenic aerosols perturb the climate by absorbing 40–80 times more solar energy than the aerosols from the natural sector over the AS and BoB. Taking into account the weak coupling between the surface and atmosphere during the Indian winter monsoon as reported in the previous studies (Ramanathan and Carmichael, 2008), it is speculated that the aerosol radiative effects scenario over the Indian subcontinent point towards the IGP getting cooler and the central India getting warmer. The large surface cooling and the differential warming of the atmosphere with respect to the surface could have impact on the regional circulation patterns.

5. Conclusions

All-sky mean aerosol radiative effects at the surface, top of the atmosphere, and within the atmosphere during the NE Indian winter monsoon in the shortwave spectrum were evaluated due to aerosols emitted from the different sectors (natural and anthropogenic) and geographical regions over the Indian subcontinent and adjoining oceanic regions using region- and source-tagged emissions transport in the LMD-ZT GCM. The spatial distribution of aerosol single scattering albedo was estimated to be lower over India and the adjoining oceanic regions of the Arabian Sea and the Bay of Bengal compared to the tropical Indian oceanic regions. The lower values of SSA estimated were mainly attributed to aerosols emitted from India and Africa-west Asia which have a higher impact over the Indian subcontinent and the adjoining ocean. Among the sectors contributing aerosol emissions, aerosols from biofuel combustion emissions exhibited the lowest SSA (0.7–0.78) over the entire Indian subcontinent and ocean.

The spatial distribution of the GCM estimated mean total all-sky aerosol radiative effects during the NE winter monsoon showed that the aerosols from fossil fuel combustion emissions reduce the incoming solar radiation at the surface 6–7 times more, compared to 2–5 times only from the biofuel use, than the aerosols from the natural sector. The region-tagged model estimates showed that the reduction in surface radiation (-3 to -8 W m^{-2} each) is mainly due to the aerosol emissions from the Indian regions and Africa-west Asia. It is worth noting that the aerosols from emission sectors lead to a homogeneous distribution in terms of sign of the aerosol radiative forcing at TOA, e.g. aerosols from BF use exert positive radiative forcing everywhere, while fossil fuel exert negative. However, aerosols originating due to the emissions from the different regions lead to both positive and negative radiative effects at the TOA depending upon the relative contribution from regional emissions to the fraction of scattering or absorbing aerosols. At the TOA, north to 10°N, aerosols emitted from India and Africa-west Asia lead to positive all-sky radiative effects over AS, BoB, central India and both positive and negative effects over the IGP in correspondence with the estimated lower SSA above 10°N and the transport features of BC in higher layers above the surface of high albedo. Aerosol radiative effects over the Indian subcontinent due to the influence of emissions from the Indian regions reveal that the control of BC emissions specifically over the IGP and CNI could be helpful to reduce the major part of atmospheric radiation absorption over India. Besides, the long-range transport of dust mixed with regional BC emissions could also lead to an enhanced atmospheric warming as suggested in the previous studies. Hence, it is also required to examine the aerosol radiative forcing with the mixing effects of the different aerosol components. Overall, taking into account the weak coupling between surface and atmosphere existing during the northeast Indian winter monsoon as reported in previous studies, radiative effects scenario over the Indian subcontinent could lead to the Indo-Gangetic plain getting cooler while the central India warmer. This could have impact on the wind circulation and the hydrological cycle and the Indian monsoon. These need to be investigated through incorporating aerosol radiative effects into a coupled atmosphere–ocean model.

We believe that the refinement in the spatial distribution of emissions and the meteorological fields used in the model may help in closing some of the discrepancy between the modelled AOD and the MODIS AOD. The possible inaccuracies in the retrieval techniques of the MODIS AOD, including the possibility of lack of efficacy of aerosol model used in the retrieval algorithm could also be one of the reasons for a large discrepancy between the modelled and the MODIS AOD, specifically over the IGP. A comparison of AOD data from the MODIS with the other ground based observations and the evaluation of measurements with the multiple modelling platforms (Verma et al., 2011, in preparation) can provide a valuable insight into the existing discrepancies between the model estimates and the measurements over the Indian subcontinent.

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