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Direct Aerosol Radiative Forcing Uncertainties due to different Factors in Different Environments

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Abstract : The present study aimed to give a comprehensive picture of the parameters which govern the aerosol radiative forcing and subsequent heating rates. Five aspects have mainly been concentrated in this particular study: (1) Absorbing type aerosols (2) Vertical distribution (3) Presence of cloud (4) Surface reflectance and (5) Diurnal variation of aerosols. It has been seen large difference in estimated atmospheric aerosol radiative forcing due to absorbing type aerosols; which are 30.5 Wm-2; 24.5 Wm-2 and 12 Wm-2 for low; average and high SSA values. There is significant difference in atmospheric radiative forcing due to change of vertical profile of aerosol i.e., 24.5 Wm-2 and 27 Wm-2 for default and elevated aerosol layer case.The presence of cloud in the atmosphere dramatically modified the atmospheric radiative forcing. It has gone from 24 Wm-2 to 37 Wm-2 as cloud moved from above the aerosols to below the aerosols. It is also seen that the atmospheric radiative forcing due to aerosols goes from + 24.5 Wm2 to + 45 Wm2 as surface albedo changes from vegetation type to snow type. Finally the study showed that the aerosol radiative forcing also significantly affected by diurnal variation of aerosols and it is + 10.5 Wm2 for averaged optical properties of aerosols and + 12 Wm2 for the instantaneous optical properties of aerosols over Re Union, Nainital and Pune respectively.

Index Terms: Aerosols,Vertical Distribution,Surface refleVertical profilectance,

1. INTRODUCTION

Aerosols are tiny liquid or solid particles suspended in the Earth's atmosphere. When these particles are sufficiently large or dense, we notice their presence as they scatter and absorb solar radiation, leading to the formation of haze, mist, fog, etc. These can produce obvious effects such as reduction in visibility, reddening the sunrises and sunsets and less obvious effects by perturbing the radiation budget of the Earth–atmosphere system. The different sources of natural aerosols, a)Volcanoes, b) Windblown dust, c) Forest fires and d) Sea salt particles and the different sources of anthropogenic aerosols, a) Vehicular pollution, b) Aeroplane exhaust, c) Industrial emissions and d) Thermal power plants.

These aerosols originates from both naturally and human activities. Natural originates includes, volcanoes, dust storms, forest and grassland fires, living vegetation and sea spray. A human activity includes, burning of fossil fuels and the alteration of natural surface cover, also generate aerosols. Aerosols produced from different natural and anthropogenic activities are mixed together and hence each aerosol particle is a composite of different chemical constituents. Chemical composition of aerosols determine their complex (contains real and imaginary parts) refractive index. Particles refractive index is an important parameter while determining the radiative effects. The real part determines its scattering properties and imaginary part, its absorption characteristics. The chemical composition and hence the refractive index depends on the source of particles. The real part of particle refractive index usually lies in the range of 1.3 to 1.6 and imaginary part varies over several orders of magnitude from about 5×10^{-9} to 5×10^{-1}. Particles originating from combustion (burning) processes usually have high absorption properties and hence high imaginary part of refractive index.

Aerosol particles may occur by themselves or may be formed as chains of spheres or cubes. Particles also form as hollow shells with air inside. The condensation of volatile gases over already existing aerosols often form coated spheres with an inner core and an outer shell both having different properties. Hollow shells and coated spheres are very difficult to treat in the estimation of aerosol optical properties and consequently their radiative effects. It is the usual practice that aerosols are either treated as externally mixed or internally mixed. In external mixture, it is assumed that individual particles of different properties exist separately and in internal mixture it is assumed that each particle is a mixture of all kinds of aerosol constituents.In this terminology, atmospheric aerosols are all the particles in the atmosphere larger than a few molecules and smaller than cloud

droplets. In practice, the atmospheric aerosol particles have diameters ranging from 1×10^{-3} to 100 μm.

. Aerosols occur with different shapes and the shape can vary with the formation mechanism. Particles formed by the condensation of water vapour molecules are generally spherical in shape while those formed by breaking larger particles are non-spherical. The shape critically controls the particle's optical and radiative properties (McCartney, 1976; Asano and Yamamoto, 1975; Hill et al., 1984) and hence radiative forcing. Broadly aerosols can be categorized as follows (Reist, 1984), Isometric particles, Platelets and Fibres. In the view of above the present study focussed on the characterisation of aerosols and to assess their radiative forcing over different kinds of environments like maritime clean and urban(Pune) Re union 25 island .

Re Union Island (20o88'N, 55o48'5E, 0 amsl) is situated in the South-Western Indian Ocean. This tiny (area: ~2500 km2) holds two agglomerations with ~100,000 inhabitants: Saint-Denis, located to the North and Saint-Pierre located to the South of it. The island has light/heavy industries, and large number of vehicles such as cars, trucks and buses operate with diesels which are the main sources of anthropogenic aerosols. The site also highly affected by sea spray via winds and a potential sea salt source over this area (Chatrapatty et al., 2001, Smirnov et al., 2009, Chatrapatty et al., 2013).Nainital (29o4'N, 79o5'E, 1958 amsl) is a clean environment and is a high altitude station in the central Himalayan region in India. The station is free from industries around it, and a little number of automobiles that include buses, cars, and two wheelers are running in the city all of which will not significantly contribute to the production of aerosols. Being a high altitude location and free from local emission sources it provides important information on background changes of aerosol concentration, properties and its radiative effects (Sagaret al., 2004, Dumka et al., 2008).Pune (18o53', 73o80', 559 amsl) is an urban environment in the western part of an Indian continent.

1.*1 Instrumentation AND Methodology*

AErosolROboticNETwork(AERONET) is a global network of instrument which is connected remotely and the network is established by National Aeronautics Space Agency (http://aeronet.gsfc.nasa.gov/). The instrument contains sun/sky radio meter (CIMEL), a ground based remote sensor to measure the radiation flux at different wavelengths. Furtherthe project is broadened by contributions from various national/governmental agencies, institutions, universities and independent research labs. Figure 2.1 shows the AErosol Robotic NETwork stations established by NASA over the globe. This network maintains a database of optical, radiative and microphysical properties of aerosolsand the data is continuously updated, and is accessible to the various researchers over the globe. The network station also maintains the standard for instrument calibrations and processing of data.Raw data of the instruments from ground stations is transferred to AERONET in following three ways.First the data from all stations are transferred to the processing station through satellite. Second photometer is connected to a PC which downloads the data from photometer using internet. The download data is sent to the processing server either manually or automatically. In the last way data is manually extracted from the photometer periodically and then send it to the processing station via internet.

The actual processing of raw data involves application of several algorithms. One of these algorithms is used for AOD retrieval and other for AOD cloud screening. The algorithm that is used for sky radiance data inversion is the most computationally expensive and time consuming algorithm. The processed data can be distributed by the data display tool on the AERONET website. The web display tool provides a graphical representation of the data and is accessible to users. AERONET produces three primary products aerosol optical depth (AOD), perceptible water vapour content and inversion products obtained based on measured direct and diffuse radiation flux by AERONET Cimel sun/sky-radiometers.

The inversion products include optical properties like size distribution, complex refractive index, asymmetry factor, single scattering albedo etc. The single scattering albedo and asymmetric factor etc were estimated based on measured direct and diffuse radiation. Other parameters like size distribution, complex refractive index were calculated based on the retrieved aerosol properties (e.g. phase function, single scattering albedo) and measured spectral and broad-band fluxes. In the data products, there are three levels of data quality available at AERONET. Level 1.0 contains unscreened data and it is considered as raw data without undergoing any processing. Level 1.5 contains the data which are cloud screened but not quality assured. This type of processed data is obtained from by removing the cloud influence on the unscreened data.

The data from level 1.5 is further processed for quality to produce level 2.0 data. Level 2.0 contains the data which are both cloud screened and quality assured. In the present study it has been used level 2 data which is quality and cloud screened(Cloud

screening and quality control algorithms for the AERONET data base, submitted to Remote Sensing of Environment, 2000a)

1.*2 SBDART Model*

The fluxes of aerosol free cases were estimated using Sanata Barbara DISORT Atmospheric Radiative Transfer Model (Ricchiazziet al., 1998). Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model is a tool that computes plane-parallel radiative transfer in clear/cloudy sky conditions within and at the surface of the earth's atmosphere. The processes that affect the radiation in all bands (ultraviolet, visible, and infrared) are included in this model.

The model is a sophisticated discrete ordinate radiative transfer model, low-resolution atmospheric transmission model, and Mie scattering results for light scattering by water droplets and ice crystals are included.The model is well suited for a wide variety of atmospheric radiation interactions, radiation flux balance, remote sensing studies and it is also designed so that it can be used for case studies as well as sensitivity analysis.There were many scientific studies on aerosol radiative forcing and earth's radiation balance based on this model (Satheesh et al., 2008, Panicker et al., 2010, Ramachandran et al., 2010, Sumit Kumar et al., 2012).

1.*3 Results and Discussions*

1.3.1.*Direct Aerosol Radiative Forcing Uncertinities due to Different Environments*
1.3.2.Vertical Distribution of Aerosols

Past studies showed the existence of elevated aerosols in the atmosphere over different environments in the atmosphere (Ramana et al., 2004,Satheesh et al., 2008,Hegde et al., 2009, Srivastava et al., 2010, Dimitriset al., 2012). These layers could be due dry convicting lifting of aerosols and subsequent horizontal long range transport from distant locations (Ramana et al. 2004, Hegde et al. 2009). The height and concentrations of elevated aerosols were also highly variable in space and time. For example the height of elevated aerosols changed from 0.5 km to ~ 4 km and the concentration also changed from ~ 10% to ~ 50% as one move from Indian ocean to central India (Satheesh et al. 2008). The concentration of elevated aerosols reaches as high as 70% in the central Himalayan region (Ramana et al. 2004, Hegde et al. 2009). These aerosol layers can leads significant uncertainty in the estimation of aerosol radiative forcing at the surface as well in the atmosphere (Satheesh et al. 2008). There were studies which discuss the importance of vertical distribution of aerosols as an input for calculating the radiative forcing (Gadhavi and Jayaraman2006). They have also shown with specific emphasis on change of the sign of the forcing at tropo-pause level on improper selection of vertical profile of aerosol extinction. This uncertainty increases with the absorbing type aerosols in the elevated aerosol layer (Samsetet al., 2013).

In the view of above it is very important to study the effect of elevated aerosol layer in the estimation of direct aerosol radiative forcing. For the present study, It has been assumed the elevated aerosols in the height region of 0.7 – 1.0 km and their contribution to total aerosols optical depth is about 40%. This profile has been used in the estimation of radiation fluxes along with default profile from radiative transfer model. Figure 5.3 shows two different vertical distribution of aerosol mass concentration in case of standard aerosol profile (radiative transfer model) and in case presence of elevated aerosol layer (elevated aerosol layer with the 40% aerosol contribution). These two different vertical profiles have been used in the estimation of direct aerosol radiative forcing. The same optical properties of aerosols have been used for the both cases, average aerosol optical depth is 0.3 and single scattering albedo is 0.90 at 0.5 μm.

The estimated surface radiativeforcing were -32, -31 $^{Wm-2}$ and radiativeforcing at the top of the atmosphere were -5, - 6.5Wm^{-2}. Subsequently, the atmospheric forcingwere 27 and 24.5 Wm^{-2} respectively, for the case of presence of elevated aerosol layer and without elevated aerosol layer. It is found that the difference in radiative forcing 1Wm^{-2} at surface, 1.5 Wm^{-2} at the top of the atmosphere and 2.5 Wm^{-2} in the atmosphere due to change in the vertical distribution of aerosols. Figure 1 shows the aerosol radiative forcing at different levels of the atmosphere in case of standard aerosol profile and in case of elevated aerosol case.

It is interesting to note from the figure 1 that enhancement in radiative forcing in the elevated aerosols case at 0.75km due to the high concentration of aerosols; whereas no enhancement in radiative forcing found in standard profile case. The corresponding radiative forcing for standard and elevated aerosols cases at 0.75km due to aerosol layer are ~ 16 Wm^{-2} and 20 Wm^{-2} respectively. At the surface radiativeforicng is varied from ~ 22 Wm^{-2} to 23.5 Wm^{-2}, enhancement of about 1.5 Wm^{-2} has been observed in radiativeforcing due to change in vertical distribution of aerosols. It is also clearly evident from the figure 1 that the vertical profiles of radiativeforcing are quite different for elevated aerosols case and standard aerosol profile case at each altitude for the same optical properties of aerosols.

The reduction of radiation fluxes will re-distribute in atmosphere which ultimately reflects in the heating rate leading to modification of the thermal structure of the atmosphere. Figure 2 shows the vertical distribution of heating rate for standard and elevated aerosols cases.It is interesting to note from the figures that enhancement in heating rate is clearly visible in the elevated aerosols case at 0.75km due to aerosol layer; whereas no enhancement in heating rate found in standard profile case. The corresponding heating rates for standard and elevated aerosols cases at 0.75km due to aerosol layer are 0.6 k/day and 0.8 k/day, respectively. At the surface heating rate is varied from ~ 1.8 k/day to 3 k/day, enhancement of about 1.2 k/day has been observed in heating rate due to change in vertical distribution of aerosols. It is also clearly evident that the vertical profiles of heating rates are quite different for elevated aerosols case and standard aerosol profile case at each altitude for the 25 same optical properties of aerosols.

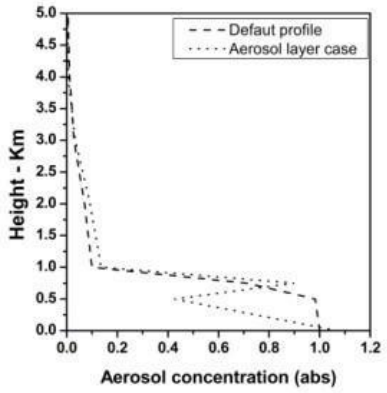

Figure 1 : vertical distribution of aerosol mass in case of standard aerosol profile and in case of elevated aerosol case.

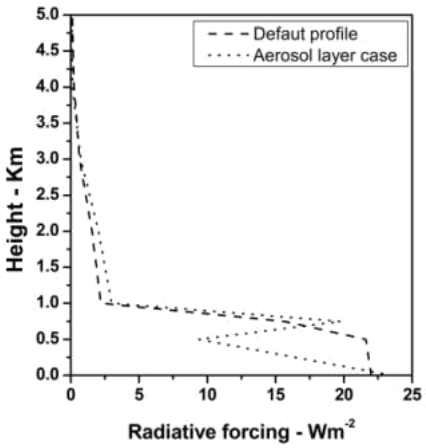

Figure 2 : Direct aerosol radiative forcing at different levels of the atmosphere in case of standard aerosol profile and in case of elevated aerosol case.

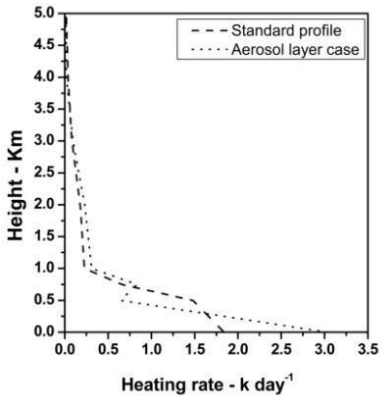

Figure 3 : Vertical profile of heating rates (k/day) in case of standard aerosol profile and in case of elevated aerosol case. Gadhavi and Jayaraman(2006), showed the importance of vertical distribution of aerosols as an input for calculating the radaitive forcingwith specific emphasis on change of the sign of the forcing at tropopause level on improper selection of vertical profile of aerosol extinction.

It has been observed that the negative temperature gradient in the vertical profile of heating rate in the standardaerosol profile case throughout the lower atmosphere. This type of temperature profiles will leads to high thermal convections in the vertical column of the atmosphere. The temperature gradiant in vertical profile of heating rate in elevated aerosols case is varying with the altitudes. In the present case there is negative temperature gradient from 0 to 0.5 km and from 0.75 to the 5 km whereas the temperature gradiant is positive from 0.5 to 0.75. Due to the negative temperature gradient there is a possibility of mixing of aerosols up to 0.5 km. From 0.5 to 0.75 km the atmosphere is highly stable due to positive temperature gradient. This acts as a inversion layer which will not allow mixing of aerosols from below this layer. The pollutants which are emitted at the surface of the atmosphere will be trapped in the atmosphere below 0.75 km. This condition will also affect the cloud formation as the water vapour will not move to sufficient height to form a cloud. Here, present study reconfirm and emphasizes the importance of vertical profile of aerosols along with the properties of aerosols in the estimation of aerosol radiative forcing.

1.4 Presence of Cloud

The presence of cloud can dramatically enhance the radiative impact of absorbing aerosols, particularly when aerosol layer is above the cloud layer.

Few studies reveal the presence of elevated aerosol layers in the atmosphere is a common phenomenon. Height and width of aerosol layer can vary from few meters to kilometres (Podogornyet al., 2001, Satheeshet al.,2008, Chandet al.,2008). The width of cloud can vary from few meters to about 500 m and the existence of stratus cloud between 0 and 8 km altitude range with different frequency of occurrence over Indian summer monsoon region. Different types of cloud and the amount of cloud presence also studied over the Indian sub continent during different months (Devasthaleet al. 2009). From the above studies, it can be concluded that the presence of aerosol layer can occur above and below thecloud layer. In the view of above it is important to study the sensitivity of aerosol radiative forcing in the cloudy atmosphere. In the present work it has been estimated the effect of cloud reflectance in the estimation of radiative forcing. To calculate the aerosol radiative forcing in cloudy sky, it has been assumed an optically thin stratus continental low level cloud above and below the aerosol layer. The properties of cloud are assumed based on the previous studies on global cloud climatology (Dong et al., 1997, Margetet al., 2009, Devasthaleet al., 2009, Subramanyamet al., 2013). The cloud droplets number distribution is assumed to follow

modified Gamma distribution after Deirmendjian (1969):

$dN/dr = Nar\alpha\exp(-B r\gamma)$ (1)

With $B = \alpha/(\gamma r^{\wedge}\gamma \text{ mod})$,

Where N is the total number density in particles per cubic centimetre and the mode radius in micro-meters. The constants α and γ describes the slope of the size distribution, while "a" is a normalization constants ensuring that the integral of the size distribution over all radii yields N.The assumed optical depth of a status continental optically thin low level cloud is 4 at 500 nm with the vertical width of 0.2 km extended in the atmosphere. Cloud coverage has been assumed to be 25 % in the both cases when aerosol layer is above and below the cloud. Column optical properties of aerosol are assumed to be same as the previous study. The presence of elevated aerosol has been assumed at 0.75 km with the vertical width of 0.2 km. The direct aerosol radiative forcing at the top and surface of the atmosphere are – 5 Wm-2 and -32Wm $^{-2}$ in the clear sky. Subsequent atmospheric aerosol radiative forcing is + 27 Wm^{-2} as discussed in the previous section.

In the first case the cloud layer has been assumed above aerosol layer at 1.5 km with the vertical width of 0.2 km as shown in figure 4.

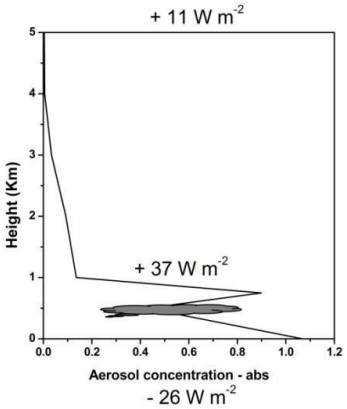

Figure 4: Vertical distribution of aerosols in clear sky. The ordinate represents height above the ground level.

In this case we observed direct aerosol radiative surface forcing at top of the atmosphere and surface are + 2 Wm $^{-2}$ and – 26 Wm^{-2} respectively. Subsequent atmospheric forcing is + 24 Wm^{-2} which is lower than the forcing in clear sky case. In the second case, we assumed the cloud layer at 0.5 km with the vertical width of 0.2 km which is below the aerosols layer as shown in figure 5. Here we got radiativeforcing at top of the atmosphere and surface are + 11 Wm^{-2} and - 26 Wm^{-2}.

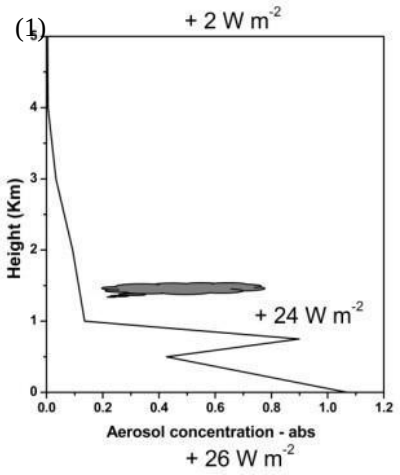

Figure5: Vertical distribution of aerosols in cloudy sky, a thin cloud layer exists above the aerosol layer. The ordinate represents height above the ground level.

Subsequent atmospheric forcing is + 37 Wm^{-2} which is higher than the forcing in clear sky case. Figure 6 – 8 shows the direct aerosol radiative forcing at top of the atmosphere, surface and atmospheric forcing in three cases, clear sky, when the cloud is above the aerosol layer and when the

cloud is below the aerosol layer. It is observed that the surface radiative forcing in the cloudy sky are higher than the clear sky. This is due to the additional effect of cloud to the to the aerosols layer. The atmospheric forcing are high and low if the cloud is below the aerosol and above the aerosol when compare to clear sky

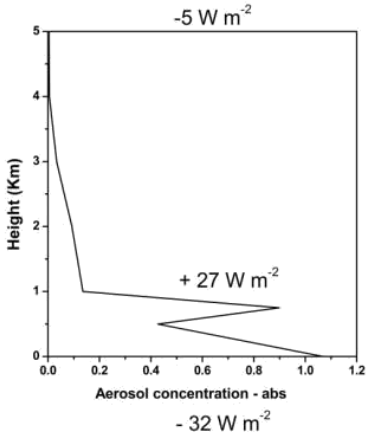

Figure 6: Vertical distribution of aerosols in cloudy sky, a thin cloud layer existsbelow the aerosol layer. The ordinate represents height above the ground level.

In the first case as there is absorption in the aerosol layer for to and fro ways of the radiation, and radiation budget is high when compare to second case as most of the radiation will be reflected back to space from cloud itself, so outgoing radiation is high compare to first case makes difference in radiative forcing at the top of the atmos phere.Atmospheric aerosol

radiative forcing is highly sensitive to the amount of absorbing aerosols in case of cloud layer is below the aerosol layer whereas it has no significant effect when cloud layer is above the aerosol layer. Radiative forcing at the top of the atmosphere also highly sensitive to the size distribution of cloud drops when the cloud is below the aerosol layer (Podogornyet al., 2001, Satheeshet al.,2008). The size distribution of clouds plays an important role in changing the albedo

of cloud as the small drops reflects more radiation in backward direction and vice versa.

From the above study it is concluded that, the height of cloud layer (whether it is above the aerosols layer or below the aerosol layer), microphysical properties and type of cloud, vertical profiles of aerosols along with the properties are important in order to estimate radiative forcing in the cloudy atmosphere. If the cloud is above the aerosol layer most of the radiation will be reflected back from cloud (depends on albedo), the available radiation to interact with the aerosol layer is less which implies less radiation budget. When the cloud is below the aerosol layer, incoming radiation will interact with aerosol layer and after passing through the aerosol layer it gets reflected by cloud and reflected radiation will again interacts with aerosol layer in this case which leads to high radiation budget.

1.5.Surface Reflectance

Surface reflectance or surface albedo is one of the highly variable parameters in space and time. The surface albedo mainly depends on the properties of surface material for example Ocean contains water and the reflectance depends on the properties of water surface. Same way the reflecting properties of pure water is different from Ocean water as it contains lot of minerals and salts. Similarly Land, snow, grass and each body has their own reflecting properties and gives different surface albedos. It also variable seasonally for example during the monsoon period most of the land will be covered by trees and grass. In addition the surface also wet with lot of water on it. When summer comes most of the wet will washout by large heating due to high solar radiation and surface becomes dry and make trees and grass dry. This phenomenon causes the variation of surface albedo with time.The surface reflectance of fresh snow is about 1 for the short wave solar radiation whereas for fresh water it is about 0.3. From this it can be observed that it is highly variable and causes variation in estimation of aerosol radiative forcing.

The other optical properties of aerosols were taken same as the previous section. Vertical profiles of aerosols have been taken according to standard model. Figure 7 shows the estimated direct aerosol radiative forcing at TOA (Top of Atmosphere), BOA (Bottom of Atmosphere) and AF (Atmospheric Forcing) as a function of surface reflectance (the surface type is going from vegetation to snow).

The estimated radiative forcing at top of the atmosphere increased (from negative to positive) as surface reflectance changed from vegetation to snow. It is increased from – 6.5 Wm^{-2} to 26 Wm^{-2} with the rate of 6.5. Snow surface has high albedo which reflects more solar radiation and the reflected radiation will again pass through the atmosphere gives more forcing at the top of the atmosphere. Vegetation surface has relatively less surface albedo which reflects less amount of radiation and the small amount of radiation will again pass through the atmosphere gives less forcing at the top of the

atmosphere. The surface radiative forcing decreased from – 31 Wm^{-2} to -19Wm^{-2} with the rate of 4.2 as the surface changes from vegetation to snow. Less radiative forcing over snow surface is due to high surface albedo which leads less net flux in case of aerosols when compared to no aerosol case. Subsequently the atmospheric radiative forcing increased from 24.5 Wm^{-2} to 45Wm^{-2} with the rate of 2.3 as surface changes from vegetation to snow. Figure 8 shows the direct aerosol radiative forcing over two different surface reflectance,vegetation and snow.

As shown in figure 9, radiative forcing at the surface is reduced by factor 1.6 and the atmospheric radiative forcing increased by factor 1.8 as the surface changes from vegetation to snow.

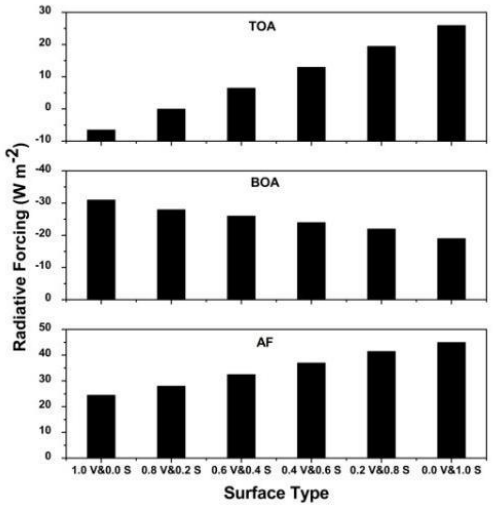

Figure 7: Estimated aerosol radiative forcing at TOA (Top of Atmosphere), BOA (Bottom of Atmosphere) and AF (Atmospherig) as a function of surface reflectance (the surface type is going from vegetation to snow).

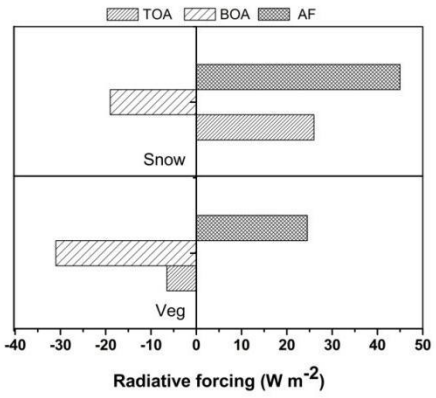

Figure 8: Direct aerosol radiative forcing over two different surface types which have different reflectance,vegetation and snow.

1.6.Diurnal variations of aerosols

From this study, it is observed that there is an increment in radiative forcing in the atmosphere as reflecting surface goes from vegetation type (relatively less reflecting) to snow type (relatively highly reflecting). This will further amplify with the increment of soot concentration in the atmospheric aerosol composition. If there is an elevated aerosol layer in the atmosphere the effect will amplify leads high stability of the atmosphere. In this case the atmosphere will not allow cloud formation as the temperature of the atmosphere increases and will not allow any pollutant to disperse. It is clearly understood from the above study that the radiative forcing is highly sensitive to the surface reflectance. The aerosol radiative forcing are different for the different surface reflectance for the same optical properties of aerosols. In this contest it is highly essential to use accurate surface reflectance to get more realistic of aerosol radiative forcing.

Most of the previous studies considered daily averaged optical properties of aerosols in the estimation of direct aerosol radiative forcing (Ramana et al., 2004, Dumka et al., 2006, Satheesh et al., 2010, Marque et al., 2010, Ramachandran et., 2010, Paniker et al., 2010). This approach gives reasonable assessment on aerosolradiative forcing when the diurnal variations of column integrated aerosol mass concentrations are constant. Previous studies also showed negligible diurnal variation in the aerosol optical depth and other optical properties over most of the low altitude station such as Bangalore,

Gadanki, etc (Satheesh et al., 2010, Gadavi and Jayaraman, 2010). However, it is also observed that the column integrated 10 aerosol mass concentration exhibits large diurnal variation over a high altitude stations (Dumka et al., 2006, Marque et al., 2010). This leads huge diurnal variability in aerosol optical depth and other optical properties attributed to boundary layer dynamics and thermal convections. This phenomenon increases aerosol optical depth from forenoon to after noon and it is also noticed that the increment is due to transport of anthropogenic aerosols from nearby valley (Dumka et al., 2006, Marque et al., 2010). Due to this diurnal variation of aerosols, the average optical properties of aerosols could underestimated /overestimate the instantaneous properties and considering the averaged optical properties of aerosols leads significant uncertainty in the estimation of aerosol radaiative forcing. Hence, it is important to consider the instantaneous aerosol optical properties for accurate estimation of radiative forcing. In this context, it is attempted to understand the contribution of diurnal variation of aerosol radiative forcing over high altitude station.

To see the effect of diurnal variation of aerosols on direct aerosol radiative forcing, it has been considered the diurnal variation of aerosol optical properties along with the averaged optical properties of aerosols in the estimation of radiative forcing over Nainital. There are rarely available data that shows the diurnal variation of aerosol optical propertieswhich constraint the number of days. In the present study it has been considered for a test day during November which shows large diurnal variation.

1.7.Diurnal variation of aereosol optical properties

The measured averaged aerosol optical depth at 440 nm is 0.12, and varied from 0.08 (minimum) to 0.19 (maximum). Figure 9 shows the hourly averaged diurnal variation of aerosol optical depth at 440 nm. As shown in the figure, the measured aerosol optical depth showed large diurnal variations with low values 0.08 during early morning hours and high values during afternoon hours with peak values 0.19.The diurnal variation has been observed in aerosol optical depth at 440 nm with high values during afternoon (factor of 1.5) when compared to forenoon.These types of variations were also reported previously by Dumka et al. (2006) over the same locations. Few other reports also concluded that the aerosols show significant diurnal variation over hill stations, for example (Marcque et al., 2010). This leads high variation of aerosol optical properties as day progress. The diurnal variation of aerosol optical depth and other properties shows negligible variation over low altitude stations such as Bangalore, Gadanki (Satheesh et al., 2010, Gadavi and Jayaraman, 2010). This could be due to the fact that there is redistribution of aerosols within the boundary layer and this may not imply any change of properties and concentration of aerosols (Satheesh et al., 2010). However, the aerosol optical depth over any high altitude stations is unique showing the increment during daytime.

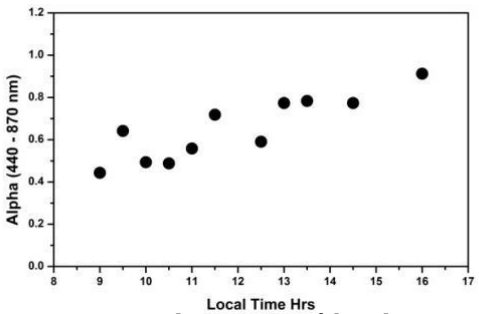

Figure 9 :Diurnal variation of hourly averaged angstrom exponent (440 – 870 nm).

This could be due to the addition of aerosols from nearby valleys due to boundary layer dynamics. Previously there were reports which also reveals the same diurnal variation in aerosol optical depth during winter season (Dumka et al. 2006) and they also explained the high values of aerosol optical depth during afternoon is mainly due to transport of aerosol from nearby plane areas or valleys.

The derived angstrom exponent (α) also followed the similar diurnal variation and is directly related to aerosol optical depth as shown in Figure 10.The angstrom exponent shows low value 0.44 during morning and high value 0.91 during afternoon with an average value of 0.65 during daytime.Low values of αduring forenoon is attributed to the background aerosols where as high values during afternoon imply the transport of anthropogenic aerosols. The up lift of fine mode aerosols is highly significant when compared to coarse mode aerosols due to their lighter weight. This could be one of the reasons for high values of angstrom exponent as the day is progress.The single scattering albedo (SSA) also showed high diurnal variation and is inversely related to aerosol optical depth and angstrom exponent as shown in figure 11. The single scattering albedo shows high value 0.96 during morning and low value 0.86 during afternoon with an average value of 0.90 during daytime. High value of SSAduring forenoon is attributed to the background aerosols where as low value during afternoon imply the anthropogenic aerosols.

1.8.Direct aerosol radiative forcing(DARF)

The aerosol radiative forcing has been estimated for two cases one is average aerosol optical properties and other one is instantaneous aerosol optical properties. Figure 12 shows the averaged direct aerosol radiative forcing at top, surface and in the atmosphere for two cases. In the first case, the averaged radiative forcing value at surface is - 11 Wm^{-2} and top of the atmosphere is – 0.5 Wm^{-2} and subsequent atmospheric forcing is + 10.5 Wm^{-2}. However, the forcing values in the second case are higher when compared to the first case.The forcing values at surface is - 12 Wm^{-2}, top of the atmosphere is 0 Wm^{-2} and the subsequent atmospheric forcing is + 12 Wm^{-2}.

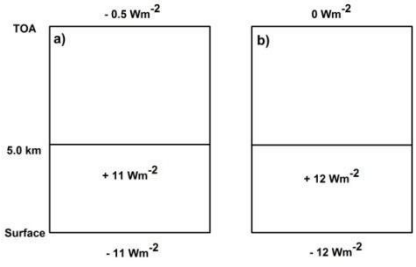

Figure 10:Diurnal variation of hourly averaged single scattering albedo at 441 nm.

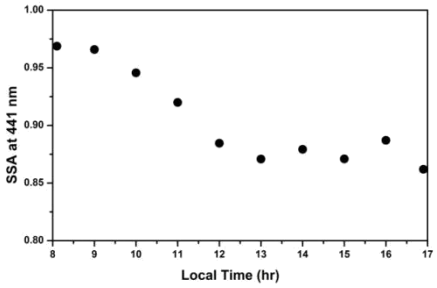

Figure 11 : Direct aerosol radiativeforcing at the top, surface and in the atmosphere at averaged and instantaneous optical properties of aerosols.

The differences in radiative forcing (second case – first case) at the surface is 1.5 Wm $^{-2}$ leading to ~ 9 % and difference in atmospheric radiative forcing (second case – first case) are 1.5 Wm^{-2} leading to a contribution of ~14%, which is significant 10 in respect to the magnitude of aerosol radiativeforcing.It should be noted that the variations in the atmospheric radiativeforcing are highly sensitive to the thermal structure of the atmosphere that affects the convection, cloud formation process, etc (Chien Wang, 2013).It is worth mentioning that, if the warming in the atmosphere is considerably large as compared to the near surface could decrease the upward movement of moisture and inturn reduce the cloud cover (Ackerman et al., 2000; Koch et al., 2010; Chien Wang, 2013). Therefore, utmost care should be taken in considering the above discussed parameters for reasonable assessment of aerosol radiative forcing. We surmise that the absorbing type, vertical profiles, presence of cloud, surface reflectance and diurnal variations of aerosols as well as boundary layer dynamics play crucial role in accurate estimation of aerosol radiative forcing.Apart from the above discussed parameters uncertainties in aerosol radiative forcing may also arise due to mixing state of aerosols. As the recent studies on aerosol optical properties indicated that the existence of mixed state(internal/ core shell) of absorbing aerosols such as black carbon with other aerosols. Internal or core shell mixing of black carbon with other aerosols shows large radiative impact when compare to external mixing of black carbon with other aerosols(Jacobson, 2001,McMeeking et al., 2011; Christopher et al., 2012).In the present report, the study has been restricted to few parameters (absorbing type, vertical profiles, presence of cloud, surface reflectance and diurnal variations of aerosols) that affect the radiative forcing. Hence, considering the above said parameters .along with mixing state of aerosols will yield more accurate estimation of aerosol radiative forcing and heating rate

1.9.Summary and Conclusions

Owing to the increasing importance to the aerosol radiative forcing in global warming aspects our study aimed to give a comprehensive picture of the parameters which govern the aerosol radiative forcing and subsequent heating rates. Five aspects have mainly been concentrated in this particular study: (1) Absorbing type aerosols (2) Vertical distribution (3) Presence of cloud (4) Surface reflectance and (5) Diurnal variation of aerosols.

It has been seen large difference in estimated atmospheric aerosol radiative forcing due to absorbing type aerosols; which are 30.5 Wm^{-2}; 24.5 Wm^{-2} and 12 Wm^{-2} for low; average and high SSA values. There is significant difference in atmospheric radiative forcing due to change of vertical profile of aerosol i.e., 24.5 Wm^{-2} and 27 Wm^{-2} for default and elevated aerosol layer case.The presence of cloud in the atmosphere dramatically modified the atmospheric radiative forcing. It has gone from 24 Wm^{-2} to 37 Wm^{-2} as cloud moved from above the aerosols to below the aerosols. It is also seen that the atmospheric radiative forcing due to aerosols goes from + 24.5 Wm^{2} to + 45 Wm^{2} as surface albedo changes from vegetation type to snow type. Finally the study showed that the aerosol radiative forcing also significantly affected by diurnal variation of aerosols and it is + 10.5 Wm^{2} for averaged optical properties of aerosols and + 12 Wm^{2} for the instantaneous optical properties of aerosols.

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