Abstract

Aerosols exhibit large spatio-temporal variabilities in their optical, physical and chemical properties, and can influence our planet by interacting with incoming solar and outgoing terrestrial radiation. The objectives of the thesis are to characterize the spatial and temporal variabilities in optical and physical properties of aerosols, source apportion black carbon aerosols, and to estimate the aerosol radiative forcing and their seasonal variability over distinct environments (urban, and high altitude remote). Aerosol characteristics were measured and examined over an urban (characterized by high aerosol concentrations dominated by anthropogenic aerosols) (Ahmedabad (23.03°N, 72.55°E, 55 m above mean sea level (AMSL)), and a high altitude remote region (with low aerosol concentration dominated by transport mechanisms) (Gurushikhar (24.65°N, 72.78°E, 1680 m AMSL). These study locations in western India are influenced by similar meteorology. The influence of atmospheric aerosols on the Earth-atmosphere radiation budget is examined using radiative transfer model. The shortwave aerosol radiative forcing is estimated using two single scattering albedo (SSA) values, one derived from the surface measurements of aerosol scattering and absorption coefficients (Method 1), and the other derived from remote sensing satellite measurement (Method 2). Further, to delineate the impact of black carbon (BC) aerosols on the Earth-atmosphere radiation budget the shortwave radiative forcing is computed for BC aerosols only over both the study locations.

Over the urban site, Ahmedabad, high values of scattering ($\beta_{\text{sca}}$) and absorption ($\beta_{\text{abs}}$) coefficients are found during morning and late evening due to a substantial increase in the anthropogenic activities and the atmospheric boundary layer dynamics. The scattering and absorption coefficients decrease as day advances (due to the evolution of the atmospheric boundary layer) and
attains a minimum value around afternoon. On the contrary, $\beta_{sca}$ and $\beta_{abs}$ over Gurushikhar are higher in the afternoon hrs when compared to forenoon and night because of atmospheric boundary layer dynamics which when accompanied with strong thermal convection aid an upward movement of pollutants to the observational site from the surrounding foothills.

The surface single scattering albedo shows a rare diurnal variability over Gurushikhar when compared to Ahmedabad. The near surface SSA is lower over Ahmedabad than Gurushikhar due to the dominance of absorbing aerosols over Ahmedabad from the anthropogenic emissions. The diurnal variation in Ångström exponent ($\alpha$), backscatter fraction ($b$), and asymmetry parameter ($g$) over Gurushikhar do not show any morning or evening peaks as observed over Ahmedabad consistent with $\beta_{sca}$ and $\beta_{abs}$ variations. The maximum $\alpha$ observed during winter suggests the dominance of smaller size aerosols. The minimum $\alpha$ and $b$, and maximum $g$ found during monsoon suggest the dominance of larger particles reaching the observational site from the marine region (Arabian Sea). The aerosol optical depth ($AOD$) over Gurushikhar is lower than Ahmedabad, as Ahmedabad is consistently influenced by the high magnitude of anthropogenic emissions, whereas the remote high altitude Gurushikhar is influenced by local and longrange transported aerosols.

The black carbon (BC) mass concentrations, and its equivalent BC from fossil fuel ($BC_{FF}$) and wood burning ($BC_{WB}$) exhibit strong diurnal variations over Ahmedabad compared to Gurushikhar due to the combined effects of the diurnal evolution of atmospheric boundary layer and consistent anthropogenic emissions. A distinct BC variation is observed over Gurushikhar with an increase in BC concentration during noontime as seen in $\beta_{abs}$. The diurnal contribution of $BC_{FF}$ in total BC dominates throughout the day at both the observational sites. The annual mean contribution of $BC_{FF}$ to total BC mass concentration is 80 and 72% over Ahmedabad and Gurushikhar respectively. This comparison indicates that even a high altitude remote site can have com-
parable fossil fuel contribution due to emissions produced over urban regions.

The study highlights the roles of single scattering albedo and aerosol optical depth ($AOD$) in the aerosol radiative forcing estimate. The differences in the forcing ($ARF$) for composite aerosol following Methods 1 and 2 is attributed to the differences in $SSA$ values viz; surface and column. $ARF$ estimated using surface $SSA$ (lower) (Method 1) is always higher than column $SSA$ (higher) (Method 2). The spectral aerosol properties for the black carbon (BC) aerosols exhibit significant variation in the $AOD$ for BC aerosols only, but $SSA$ and $g$ remain the same. The forcing for BC aerosols only over Ahmedabad is higher by a factor of 2-3 than Gurushikhar when $AOD$ also varies by the same factor, which confirms the linear dependence of $AOD$ on the $ARF$. Over an urban location (Ahmedabad), TOA forcing is comparable for Method 1 and BC aerosols only, whereas significant variations are found in SFC and ATM forcing due to $AOD$. On the contrary, the TOA forcing flips sign from +ve to -ve following Method 2 as compared to BC aerosols only. Over a high altitude remote location (Gurushikhar), the forcing values are comparable from both the methods as the SSA values are comparable. The TOA forcing is always negative as SSA is higher over Gurushikhar. The study reveals that over an urban and a high altitude remote locations the BC aerosols alone can contribute in the range of 20 to 60% to the shortwave atmospheric forcing.

It is to be noted that when a high altitude remote site is in the same region as that of an urban aerosol source location, and both theses locations are governed by the same meteorology and atmospheric dynamics, then aerosol measurements over the high altitude region can serve as regional background which is the case here. Results indicate that although Gurushikhar is a high altitude remote site, it is significantly influenced by the local and longrange transported aerosols through convection and advection. The study reveals that Gurushikhar lacks anthropogenic emissions and the aerosol properties over Gurushikhar do not exhibit any significant inter-annual variability, confirming that Gurushikhar is a regional background site for aerosols in western
India. These results can be used as inputs in regional and global climate models for the estimation of climate forcing, to further improve our understanding on the spatio-temporal variability and radiative effects of aerosols over different environments.

**Keywords:** Atmospheric Aerosols, Scattering Coefficient, Absorption Coefficient, Single Scattering Albedo, Ångström Exponent, Backscatter Fraction, Asymmetry Parameter, Aerosol Optical Depth, Black Carbon, Source Apportionment, Radiative Forcing, Heating Rate, Nephelometer, Aethalometer, Remote Sensing, Optical Properties Model, Radiative Transfer Model, Observations, Urban Region, High Altitude Remote Site.