Contribution of natural and anthropogenic aerosols to optical properties and radiative effects over an urban location

S Ramachandran, R Srivastava, Sumita Kedia and T A Rajesh

Space and Atmospheric Sciences Division, Physical Research Laboratory, Navrangpura, Ahmedabad, Gujarat 380009, India

E-mail: ram@prl.res.in

Received 1 June 2012
Accepted for publication 23 August 2012
Published 13 September 2012
Online at stacks.iop.org/ERL/7/034028

Abstract

A method to determine the contribution of natural and anthropogenic aerosol species to aerosol radiative forcing using surface-based, columnar and vertical profile measurements, optical properties and radiative transfer models is outlined. Aerosol optical properties and radiative fluxes measured during 2008 over Ahmedabad, an urban city located in western India are utilized. Mid-visible aerosol optical depth (AOD) does not show a strong seasonal variation, while $\alpha$, the Ångström exponent, exhibits significant seasonal variation. $\alpha$ is higher during winter and post-monsoon, when fine mode aerosols are dominant, while $\alpha$ is lower during pre-monsoon and monsoon, when coarse mode aerosols are abundant. The contribution of mineral dust to the total aerosol mass is higher than 55% as the study location is in a semi-arid region. Natural aerosols (mineral dust and sea salt) dominate the aerosol mass concentration, while anthropogenic aerosols (water soluble aerosols and black carbon) dominate the aerosol optical depth. The percentage contribution of black carbon to the net atmospheric forcing is larger than 65% throughout the year, corroborating that black carbon aerosol is a strong contributor to global warming on regional scales. Black carbon aerosols contribute 50% or more to the aerosol radiative forcing at the surface, thus, significantly contributing to solar dimming. The large atmospheric warming and the surface forcing due to black carbon aerosols can influence the hydrological cycle. Results emphasize that aerosol radiative forcing is governed more by aerosol optical properties (aerosol optical depth and single scattering albedo) rather than their mass, and there exists no linear relation between mass, optical depth and radiative effects of different aerosol species. These results and the relationship can be used to delineate the anthropogenic influence of aerosols from their natural counterpart, because anthropogenic aerosols in the fine mode (lower mass) give rise to higher AOD, lower SSA, higher aerosol radiative forcing, while natural aerosols which are in the coarse mode (higher mass) give rise to lower AOD, higher SSA and lower aerosol radiative forcing.

Keywords: aerosols, natural, anthropogenic, contribution, radiative forcing

1. Introduction

Atmospheric aerosols can exert a cooling and warming on Earth’s climate through direct (scattering and absorption of
solar and longwave radiation), semi-direct (evaporation of cloud droplets due to solar absorption), and indirect (modify the cloud optical properties, lifetime and albedo) effects (Solomon et al. 2007). The potential for aerosol forcing of climate can vary according to regional differences in aerosol columnar concentration, chemical composition and the age of the air mass. Aerosols are produced from natural processes and anthropogenic activities. Radiative forcing of different aerosol species (sulfate, sea salt, black carbon and mineral dust) are highly uncertain owing to the difficulty in determining their anthropogenic and natural fraction, and their spatiotemporal variations (Solomon et al. 2007). The radiative effects of aerosols depend on their size, abundance and chemical composition, and they differ for different aerosol species. Thus, some aerosol types (black carbon, mineral dust) cause positive forcing (warming), while some others (sulfate, sea salt) cause negative forcing (cooling). In a modeling framework using chemical transport models and/or general circulation models (Solomon et al. 2007) the percentage contribution of aerosol species to the total can be determined in terms of mass concentrations and optical depths. However, it has been noted that uncertainties in model outputs can arise owing to inaccuracies in estimating the aerosol burden, anthropogenic and natural fractions and implementation of their optical properties, because not only the total mass and optical depth but also the contribution from individual species have to be estimated accurately. Results from the global aerosol model intercomparison (AeroCom) initiative suggested that there exists a wide range of diversity in emissions of different aerosol types, particle sizes, residence times, deposition pathways and rate coefficients for sea salt, dust and dry aerosol, aerosol composition, aerosol water content and dispersal of aerosols both in the vertical and horizontal scales, among the sixteen different models which participated (Textor et al. 2006). For example, some models were found to exhibit a tendency to accumulate dust and carbonaceous aerosols at higher altitudes, while other models had more efficient removal schemes. Tropospheric residence times of the fine mode aerosol species were found to vary by ~25% among these models. It was emphasized that an improved representation of these parameters in large scale-aerosol models is a high priority if the uncertainty in aerosol climatic impact is to be reduced. Aerosol radiative forcing of composite aerosols have been/are being estimated directly, utilizing measurements of aerosol optical depths and radiative fluxes (Jayaraman et al. 1998), and/or following a hybrid (measurements and models) approach using measured aerosol optical properties (aerosol optical depth and single scattering albedo), and optical properties and radiative transfer models over oceanic regions and several continental locations (Srivastava and Ramachandran 2012, and references cited therein). However, studies on estimates of radiative forcing due to different aerosol species using measured aerosol characteristics are non-existent.

As the radiative effects of the aerosol species differ, and as the knowledge on the contribution of different aerosol species in the various altitudes of the atmosphere is crucial for climate change, for the first time in this study, we use measured aerosol characteristics (surface, column and vertical profile) in conjunction with aerosol optical properties and radiative transfer models to estimate the contribution due to different aerosol species to cumulative aerosol radiative forcing over an urban location (Ahmedabad) in western India. At any given location, aerosols produced from both natural and anthropogenic sources contribute to the aerosol concentration and size distribution. For example, urban locations near arid regions and adjacent to coast, such as Ahmedabad, will have contributions from anthropogenic (local), and natural aerosols (sea salt during monsoon and dust from adjacent arid regions). The proposed methodology provides a linkage to aerosol mass, optical depth and radiative effects of different aerosol species, and can be utilized effectively to delineate the contribution of natural and anthropogenic aerosols. The proposed approach is representative of average conditions and can be widely used for possible application. The results from the proposed methodology using measured aerosol optical properties will enable the development of a useful characterization of the radiative effects of different aerosol species over widely varying environments, which is crucial in the context of regional and global climate change.

2. Measurements and methodology

2.1. Study location

The study location, Ahmedabad (23.03°N, 72.5°E, elevation 55 m above mean sea level), is an urban, densely populated (population of about 5.8 million) and industrialized city in western India (figure 1). The mean synoptic surface winds during winter monsoon (December–January–February) over the western region of India, where Ahmedabad is located, are calm, north/northeasterly and are from the polluted northern hemisphere. During the southwest summer monsoon (June–July–August–September), the winds are stronger, moist and come from the marine and western regions surrounding India (figure 1). The wind patterns start shifting in direction during post-monsoon (October–November) from southwest to northeast. During the pre-monsoon season (March–April–May) the winds originate and travel from the west of the Indian subcontinent. Monthly mean relative humidity over Ahmedabad varied from a low of 24% in February to a high of 93% in July during 2008. Seasonal mean relative humidity (RH) during 2008 over Ahmedabad was about 40% or less during winter, pre- and post-monsoon seasons, which increased to >80% during monsoon (table 1).

2.2. Aerosol optical depths

The most important aerosol parameters required to estimate aerosol radiative forcing are aerosol optical depth (AOD), single scattering albedo (SSA), and asymmetry parameter (g). Aerosol optical depths were measured in the wavelength range of 0.38–1.02 µm using a pair of Microtops-II sun photometers (Solar Light Co., USA) during January–December 2008 at the
Figure 1. Surface level synoptic wind pattern (m s$^{-1}$) over the study region (Ahmedabad in western India) in (a) winter, (b) pre-monsoon, (c) monsoon and (d) post-monsoon during 2008. Colors indicate the magnitude of the seasonal mean wind speeds and the arrows represent the direction.

Table 1. Seasonal mean and ±1σ standard deviation of columnar ozone (Dobson Units (DU)), columnar water vapor (cm), rainfall (cm), relative humidity (%) and surface albedo (0.555 $\mu$m) over Ahmedabad in 2008.

| Atmospheric parameters        | Winter         | Pre-monsoon   | Monsoon       | Post-monsoon  |
|------------------------------|----------------|---------------|---------------|---------------|
| Columnar ozone (DU)          | 249 ± 5        | 272 ± 6       | 275 ± 3       | 257 ± 13      |
| Columnar water vapor (cm)    | 1.40 ± 0.55    | 2.04 ± 0.43   | 4.67 ± 0.61   | 2.32 ± 0.48   |
| Accumulated rainfall (cm)    | 0.61           | 0.71          | 60.2          | 0.03          |
| Relative humidity (%)        | 29.8 ± 6.4     | 37.2 ± 11.8   | 84.4 ± 8.4    | 43.2 ± 6.5    |
| Surface albedo (0.555 $\mu$m)| 0.112 ± 0.008  | 0.126 ± 0.005 | 0.236 ± 0.018 | 0.110 ± 0.002 |

wavelength bands centered around 0.38, 0.44, 0.50, 0.675, 0.87 and 1.02 $\mu$m. The field of view of Microtops-II sun photometers is 2.5°. AOD measurements were taken everyday at 1 h time intervals from 0900 Local Standard Time (LST) to 1700 LST under clear sky conditions. The cumulative error in AOD measurements due to contribution of forward scattering to the measured irradiance, errors associated with the calculation of air mass, deviation in the calibration coefficient, and errors in the estimation of optical depth due to Rayleigh scattering and molecular absorption is found to be 2%–5% for Microtops-II sun photometers at different wavelengths (Kedia and Ramachandran 2011). Daily mean
AODs in the 0.38–1.02 μm wavelength region measured during 2008 are used to determine the monthly and seasonal means.

2.3. Single scattering albedo

Over Ahmedabad single scattering albedo (SSA) values are calculated from the in situ measurements of scattering and absorption coefficients. Scattering and absorption coefficients of aerosols over Ahmedabad were measured using a 3-wavelength (0.45, 0.55 and 0.70 μm) integrating nephelometer (3563, TSI Inc, USA) and a 7-wavelength (0.37, 0.47, 0.52, 0.59, 0.66, 0.88 and 0.95 μm) aethalometer (AE-47, Magee Scientific, USA). Measurements of scattering and absorption coefficients of aerosols were conducted in a continuous mode at 5 min interval. The uncertainty in scattering coefficient taking into account the noise, calibration and truncation error is estimated to be a maximum of 15% (Ramachandran and Rajesh 2008). The cumulative uncertainty in absorption coefficient due to changes in filter scattering caused due to aerosol loading, underestimation of aethalometer signal with increasing filter load, flow rate, filter spot area and detector response is found to be 10%. The absorption coefficients \( \beta_{abs} \) at three wavelengths of 0.45, 0.55 and 0.70 μm are calculated following the wavelength dependence of aerosol absorption expressed as \( \beta_{abs} = K \lambda^{-\alpha} \), where \( K \) and \( \alpha \) are the absorption Ångström coefficients, respectively and \( \alpha \) (absorption Ångström exponent) is a measure of spectral dependence of aerosol absorption. The intercept (ln⁡\( K \)) and \( \alpha \) values varied from a maximum of 5.0 and 2.2 in January to a minimum of 2.4 and 2.0 in July respectively over Ahmedabad in 2008. The difference between the directly measured \( \beta_{abs} \) and those derived using the above equation is found to be <1%. SSA at 0.45, 0.55 and 0.70 μm are determined from the respective scattering and absorption coefficients. The scattering coefficients measured by the nephelometer are sensitive to relative humidity. As SSA is calculated using the \( \beta_{abs} \) measured by aethalometer and \( \beta_{sca} \) measured by nephelometer, \( \beta_{sca} \) are corrected for all the above-mentioned errors but not for RH. The maximum error in SSA taking into account the uncertainties in measured aerosol scattering and absorption coefficients is estimated to be <10%.

2.4. Asymmetry parameter

The angular distribution of light scattered by aerosols, namely, the aerosol phase function, is represented by the asymmetry parameter in aerosol radiative forcing computations. \( g \) is higher for an aerosol size distribution consisting of larger particles; for example, \( g \) is higher for maritime (coarse mode) aerosols when compared to continental/urban (fine mode) aerosols in most of the shortwave region. \( g \) is sensitive to RH; \( g \) increases as RH increases. However, it has been shown that when compared to AOD and SSA, aerosol radiative forcing is less sensitive to changes in asymmetry parameter (Srivastava and Ramachandran 2012). Spectral asymmetry parameter values for Ahmedabad are obtained using an aerosol optical properties model as described in section 2.6.

2.5. Flux observations

Ground reaching broad-band, global and diffuse fluxes in the wavelength range of 0.31–2.8 μm were measured using a set of pyranometers (Kipp and Zonen Model CM21) over Ahmedabad during 2008 (Srivastava and Ramachandran 2010). Global (direct + diffuse) flux measured at 1 s intervals on clear sky days which were subsequently cloud screened are used to calculate the 24 h average global flux and used in the study. The total uncertainty in pyranometer measured fluxes, taking into account the linearity and stability aspects of the pyranometer, the spectral sensitivity, cosine response and directional error is about 2%.

2.6. Methodology and criteria

The Optical Properties of Aerosols and Clouds (OPAC) model (Hess et al 1998) is used in conjunction with the measured aerosol parameters in this study. The necessary input aerosol parameters (AOD, SSA and \( g \)) in the shortwave region are derived by varying the aerosol components that contributed to the aerosol properties over Ahmedabad. In the OPAC model new aerosol mixtures can be defined from the given aerosol components to best fit the observations and derive the required aerosol optical properties. These parameters are calculated on the basis of the micro-physical data (size distribution and spectral refractive index) assuming aerosols as spherical particles that are externally mixed. OPAC outputs the required aerosol inputs at eight different relative humidity conditions in the range of 0–99% (Hess et al 1998), as some of the aerosol components are hygroscopic. The most suitable aerosol components based on aerosol sources, source regions, transport pathways, and earlier results over Ahmedabad are found to be water insoluble, water soluble, black carbon, mineral dust and sea salt (Ganguly et al 2006). Optical (refractive index, SSA and \( g \)) and physical properties (mode radius and density) of hydophobic and hygroscopic aerosol species used in this study are given in tables 2 and 3. Mode radius, SSA, \( g \) and density of black carbon aerosols are the lowest (table 2), while SSA for sea salt is the highest (table 3). Mode radii of insoluble aerosol and mineral dust are higher, and the mode radii of water soluble aerosols and sea salt increase as a function of relative humidity. Water soluble and sea salt aerosols are predominant scatterers; insoluble and mineral dust aerosols are absorbers, while black carbon is the single largest absorber of radiation (tables 2 and 3). SSA is >0.96 for water soluble aerosols and it increases as RH increases, for sea salt SSA is 1.00 (table 3), while SSA for insoluble, black carbon and mineral dust are invariant with RH. This is consistent with the measured SSA values derived from scattering and absorption coefficients, where the scattering coefficient which is sensitive to increase in RH is measured at ambient RH (i.e. \( \beta_{sca} \) is not corrected for RH), while absorption coefficients correspond to dry state (0% RH, hydophobic).

The different aerosol species (water insoluble, water soluble, black carbon (BC), mineral dust and sea salt) are distributed according to the seasonal mean vertical profiles.
et al vertical profiles similar to those obtained during 2002–5 using available in 2008 for Ahmedabad, seasonal model aerosol simultaneous LiDAR measured aerosol profiles were not Table 3. Real and imaginary parts of refractive index, single scattering albedo (SSA), asymmetry parameter (g), mode radius (µm) and density (g cm⁻³) of hygroscopic aerosol species at 0.55 µm, as given in Hess et al (1998) and used in the current study.

| Aerosol species       | RH (%) | Refractive index | SSA    | g    | Mode radius | ρ    |
|-----------------------|--------|------------------|--------|------|-------------|------|
| Insoluble             | 0      | 1.53 − i 6.00 × 10⁻³ | 0.962  | 0.614 | 0.021       | 1.80 |
| Black carbon          | 50     | 1.53 − i 6.00 × 10⁻³ | 0.977  | 0.672 | 0.026       | 1.42 |
| Mineral dust          | 70     | 1.53 − i 6.00 × 10⁻³ | 0.981  | 0.690 | 0.029       | 1.33 |
| Water soluble         | 80     | 1.53 − i 6.00 × 10⁻³ | 0.984  | 0.704 | 0.030       | 1.27 |
| Sea salt              | 90     | 1.53 − i 6.00 × 10⁻³ | 0.989  | 0.725 | 0.035       | 1.18 |
|                       | 95     | 1.53 − i 6.00 × 10⁻³ | 0.993  | 0.743 | 0.040       | 1.09 |

of aerosol extinction and the total AOD is estimated. As simultaneous LiDAR measured aerosol profiles were not available in 2008 for Ahmedabad, seasonal model aerosol vertical profiles similar to those obtained during 2002–5 using micropulse LiDAR (Ganguly et al 2006) are constructed and utilized. By doing so the vertical structure of aerosols similar to those measured earlier over Ahmedabad is maintained, while AOD (which is measured) corresponds to 2008. The year to year intra-seasonal variations in aerosol vertical profiles measured over Ahmedabad during 2002–5 were much less (Ganguly et al 2006). Therefore, uncertainty, if any, due to the non-inclusion of aerosol vertical profiles pertaining to 2008 will be quite small.

A flow chart describing the methodology adopted to retrieve the aerosol parameters in the shortwave region and estimate aerosol radiative forcing, using measured aerosol characteristics (aerosol optical depths, single scattering albedo and black carbon aerosol mass concentrations) in conjunction with an aerosol optical properties model and a radiative transfer model is given in figure 2. The number concentrations of the water insoluble, water soluble, black carbon, sea salt and mineral dust aerosol species are varied iteratively until the following conditions are satisfied (figure 2): (1) the root mean square (rms) error between the measured and modeled AOD spectra is <0.03, thus, constraining the rms error to within 0.10 AOD, (2) Ångström exponents α determined from the measured AODs in the 0.38–1.02 µm wavelength range are consistent with model-derived α values, (3) BC mass concentration in OPAC should be the same (within ±1σ, table 4) as that of the aethalometer measured BC mass, and (4) model-derived SSA should match with SSA obtained from in situ measurements over Ahmedabad. The contribution of the optical depth due to each aerosol species to the total AOD is estimated by assigning the optical depth of the other aerosol species as zero. Following the above procedure, necessary aerosol inputs (AOD, SSA and asymmetry parameter) corresponding to the monthly mean relative humidity are obtained in the shortwave region for composite aerosols and for different aerosol species. Relative humidity was identified as the single most important parameter in determining aerosol radiative forcing, and the increase of aerosol mass resulting from water uptake was found to be the most important process (Pilinis et al 1995). In this study, the size (mode radius), composition (refractive index) and density of aerosol species, which will impact AOD, SSA and g, change as a function of relative humidity (table 3), and therefore, AOD, SSA and g are retrieved at the corresponding relative humidity. AOD, SSA and α retrieved following the above methodology are found to compare very well with observations (table 4), thus revealing that the approach (figure 2) to retrieve aerosol parameters in the entire shortwave region utilizing the measured aerosol characteristics is quite appropriate. In this study, since an exact match is obtained between the measured and the aerosol properties retrieved assuming that aerosols are spherical and externally mixed (table 4), any impact on the aerosol optical properties and radiative forcing due to aerosols being partly (core-shell) or fully (internal) mixed and their asphericity will not be significant.

2.7. Additional inputs

The additional inputs that are necessary to perform aerosol radiative forcing calculations include atmospheric profiles of temperature, pressure, columnar ozone, water vapor
Figure 2. Flow chart describing the methodology adopted to retrieve the aerosol parameters in the shortwave region and estimate aerosol radiative forcing, using measured aerosol characteristics (aerosol optical depths, single scattering albedo and black carbon aerosol mass concentrations) in conjunction with an aerosol optical properties model and a radiative transfer model.

Table 4. Comparison of seasonal mean (±1σ) aerosol parameters aerosol optical depth (AOD) at 0.5 μm, single scattering albedo (SSA) at 0.55 μm, Ångström exponent (α) and black carbon aerosol mass (μg m⁻³) obtained from measurements over Ahmedabad with model (OPAC) retrieved values. Species wise aerosol optical depths derived from the model following the methodology illustrated in figure 2 are also given.

| Source/parameters | Winter    | Pre-monsoon | Monsoon   | Post-monsoon |
|-------------------|-----------|-------------|-----------|--------------|
| AOD               | 0.43 ± 0.09 | 0.41 ± 0.07 | 0.42 ± 0.01 | 0.46 ± 0.02 |
| SSA               | 0.69 ± 0.01 | 0.71 ± 0.03 | 0.79 ± 0.07 | 0.68 ± 0.01 |
| α                 | 1.20 ± 0.09 | 0.67 ± 0.35 | 0.69 ± 0.28 | 1.06 ± 0.15 |
| Black carbon mass | 10.6 ± 2.8  | 3.8 ± 2.5   | 2.0 ± 0.6  | 8.9 ± 3.5   |

The radiative transfer algorithm SBDART (Santa Barbara DISORT Atmospheric Radiative Transfer) developed by...
Ricchiazzi et al (1998) is used to perform the radiative transfer calculations in the shortwave (0.31–2.8 μm) region. The aerosol radiative forcing calculations are performed using 8 radiation streams at 1 h interval for a range of solar zenith angles and 24 h averages are obtained for each month, which are then used to construct the seasonal means. Aerosol radiative forcing (ARF) at the top of the atmosphere (TOA) and surface (SFC) is calculated as the change between the net (down minus up) flux with and without aerosols as

\[
\text{ARF}_{\text{TOA,SFC}} = \text{Flux(Net)}_{\text{with aerosol TOA,SFC}} - \text{Flux(Net)}_{\text{without aerosol TOA,SFC}}.
\]

The difference between the radiative forcing at TOA (100 km in this case) and surface is defined as the atmospheric forcing (ATM) and can be written as

\[
\text{ARF}_{\text{ATM}} = \text{ARF}_{\text{TOA}} - \text{ARF}_{\text{SFC}}.
\]

ARF_{\text{ATM}} represents the amount of energy trapped by the aerosols present in the atmosphere. A positive ARF_{\text{ATM}} indicates a net gain of radiative flux in the atmosphere, and hence warming, while a negative ARF_{\text{ATM}} indicates a net loss, and thereby cooling. The relative standard error in radiative forcing reported in the study, taking into account the uncertainties in aerosol input parameters, additional inputs and flux estimates, is estimated to be <15% (Srivastava and Ramachandran 2012). Lack of information on the vertical profile of aerosols can introduce uncertainty in aerosol radiative forcing estimates as a function of altitude (Solomon et al 2007). The inclusion of a vertical distribution of aerosols was found to modify only negligibly the net aerosol radiative forcing at the top of the atmosphere, surface and in the atmosphere (Srivastava and Ramachandran 2012), thus, ascertaining that the net energy content trapped in the atmosphere remains the same with and without including the vertical profiles, but only its vertical distribution varies. The emphasis of the present study is to estimate the net aerosol radiative forcing at the top of the atmosphere, at the surface and in the atmosphere, and therefore, including the vertical profile information is not expected to modify the overall outcome and conclusions. Aerosol radiative forcing is calculated including aerosol properties of all the species (total) and for different aerosol species independently. Total aerosol radiative forcing and aerosol radiative forcing corresponding to each aerosol species are calculated for each month in 2008 following the above procedure, which are then used to calculate the respective seasonal means.

3. Results and discussion

3.1. Aerosol properties

The seasonal mean variation in AOD and Ångström exponent (α derived using τ = βλ−α, where τ is AOD and β is the total columnar aerosol loading) is shown in figure 3. 0.50 μm AODs are ≥0.4 during the year over Ahmedabad (figure 3). AOD at 0.5 μm during monsoon is comparable to winter. Although wet removal is more effective in reducing the near surface aerosols, owing to a deeper boundary layer 0.50 μm AOD during monsoon is almost the same as in winter (figure 3). However, α shows a strong seasonal variation, with a high value during winter and post-monsoon, and a low value in pre-monsoon and monsoon. AODs in the visible wavelength region are more influenced by fine mode aerosols; on the other hand when coarse mode aerosols are dominant, AODs in the longer wavelength are affected more significantly. Dominance of fine mode aerosols gives rise to higher α values while an increase in coarse mode aerosols reduce α (figure 3).

The contribution of mineral dust is quite high (≥55%) to the total aerosol mass concentration throughout the year, as the study location is in a semi-arid region (figure 1); however, all aerosol species including mineral dust show significant seasonal variability. Mineral dust contribution peaks in pre-monsoon (76%). Sea salt contribution increases from 6% in pre-monsoon to 18% during monsoon. Black carbon aerosol mass concentration is high during post-monsoon and winter, and contributes about 7% to the total mass (figure 4). The contribution of water soluble aerosol species, which originate from gas to particle conversion and are made up of various kinds of sulfates, nitrates, organic and water soluble substances, is also high during winter and post-monsoon over Ahmedabad. During winter over Ahmedabad the winds are north/northeasterly and bring in anthropogenic pollutants. In addition to fossil fuel combustion, in winter, owing to colder temperatures, a significant increase in the amount of open biomass burning occurs as people burn dry leaves, shrubs, paper and waste materials to keep them warm. This leads to an increase in the amount of biomass burning aerosols; fossil fuel emissions and the additional biomass burning accompanied with the shallow boundary layer results in higher BC and water soluble aerosol mass concentrations. The insoluble aerosol species, mainly re-suspended soil/road dust particles with a certain amount of organic material, contribute in the range of 5%–8% to aerosol mass.

Water soluble aerosols and BC contribute in excess of 80% to total AOD during winter and post-monsoon (figure 4). Sea salt contribution to AOD is maximum during monsoon at 11%. The results show that though mineral dust contributes...
Figure 4. Percentage contribution of aerosol species to total aerosol mass concentration (µg m⁻³) obtained using OPAC model during (a) winter (102.9 ± 17.9), (b) pre-monsoon (99.8 ± 16.0), (c) monsoon (85.9 ± 17.2) and (d) post-monsoon (100.5 ± 9.2). Percentage contribution of water insoluble, water soluble, black carbon, sea salt and mineral dust aerosol components to the total aerosol optical depth obtained using model in (e) winter (0.43 ± 0.09), (f) pre-monsoon (0.41 ± 0.07), (g) monsoon (0.45 ± 0.05) and (h) post-monsoon (0.46 ± 0.02). Seasonal mean AODs measured at 0.5 µm and total aerosol mass concentration obtained from model (OPAC) alongwith ±1σ standard deviation are given in parentheses.

Aerosol mass concentration (µg m⁻³)

Aerosol optical depth (0.5 µm)

about 60% or more to the total aerosol mass, its contribution to the total AOD is in the 15%–36% range. During pre-monsoon, when the mineral dust aerosol contribution is more than three-fourths (76%) to the total mass, the contribution to AOD is only about one-third (36%). On the other hand, BC aerosols contribute only about 2% (monsoon) to mass to a maximum of 7% (winter), but the contribution of BC aerosols to the total AOD is higher than 30% during winter, pre- and post-monsoon. Even a 2% contribution by BC aerosol (which is in the fine mode (table 2)) to total mass results in a 22% contribution in AOD during monsoon. As the relative humidity is higher during monsoon (table 1) the mode radius of sea salt aerosol increases (table 3), and as the mineral dust mode radius is higher (table 2), their contribution to the optical depth is lower, despite their higher contribution to total aerosol mass (figure 4). Thus, it is clear that the contributions of different aerosol species to AOD vary as a function of wavelength depending on whether the particular species is in the fine mode or coarse mode. The percentage contribution of smaller size particles (e.g., black carbon, water soluble aerosols) to total aerosol mass will be quite small. It should be noted that the mass per particle decreases by 2–3 orders of magnitude when the mode radius of the particle decreases from 0.3 to 0.03 µm (Hess et al 1998). In a given volume of aerosols the number of bigger aerosol particles is orders of magnitude less compared to that of small particles; hence, coarse mode particles contribute insignificantly to aerosol optical depth. The optically effective aerosol radius range of 0.05–10 µm contributes the maximum to aerosol optical depth, as Mie scattering contribution of smaller particles (<0.05 µm) is only marginal (d’Almeida et al 1991). Though aerosols smaller than 0.1 µm are abundant in the atmosphere, their residence times are shorter, as they get converted into accumulation mode particles through gas to particle conversion mechanism. In addition, their contribution to total mass will be less. Although bigger aerosols such as mineral dust and sea salt can contribute significantly to total aerosol mass, their role in optical properties and radiative transfer are however limited, because of lower number density and shorter residence times (d’Almeida et al 1991). The results confirm that there exists no direct relationship between aerosol mass concentrations and optical depths over an urban location.

In contrast to AODs, SSA exhibits seasonal variations (figure 5). SSA at 0.55 µm increases steadily from winter to monsoon (figure 5). SSA is lower (≤0.8) during post-monsoon and winter due to the dominance of fine mode urban aerosols (mainly BC, figure 4), which are more absorbing in nature. SSA increases during pre-monsoon due to the increase in the abundance of mineral dust particles and a decrease of BC in the atmosphere (figure 4). BC mass concentrations are lowest (∼2 µg m⁻³) during monsoon over Ahmedabad. A decrease in the amount of BC accompanied
with an increase in water soluble aerosols owing to higher RH, and sea salt aerosols gives rise to a higher SSA during monsoon. The very high correlation between the measured and modeled SSA (figure 5) confirms that the modeling approach (figure 2) adopted to retrieve the measured aerosol properties and the contribution of different aerosol species is indeed robust, and the problem of retrieving aerosol optical properties is well constrained for the solution of inversion to be unique.

3.2. Aerosol radiative forcing

The radiative transfer model estimated radiative fluxes are validated against the observed fluxes over Ahmedabad (figure 6). A very good agreement is evident between the diurnally averaged (24 h) measured clear sky global (direct + diffuse) and modeled fluxes (W m\(^{-2}\)) derived using SBDART radiative transfer model (figure 6). The measured radiative fluxes correspond to the measured AOD and SSA on clear sky days during 2008 over Ahmedabad. The radiative fluxes from SBDART model are derived using the measured aerosol optical properties (AOD and SSA), atmospheric parameters and surface reflectance appropriate to the season and location as inputs (section 2). The modeled radiative fluxes are systematically lower than the measured radiative fluxes. This underestimate could arise due to the uncertainties in aerosol inputs, surface albedo or water vapor (McComiskey et al. 2008), and the differences could be larger when SSA is \(\leq 0.8\) (which is the case in Ahmedabad, figure 5). Nevertheless, the good agreement between the measured and modeled fluxes \((r^2 = 0.99)\) confirms the robustness of the methodology utilized to derive the input aerosol parameters, radiative fluxes, and hence radiative forcing.

The seasonal mean shortwave (0.31–2.8 \(\mu\)m) ARF for Ahmedabad at TOA, surface and in the atmosphere due to different aerosol species are given in figure 7. TOA forcing due to different aerosol species can be either positive or negative (figure 7), unlike the greenhouse gas forcing which is always positive. TOA forcing becomes more positive when absorbing aerosols are present over high reflecting surfaces (continent, snow) (Solomon et al. 2007). Surface albedo is higher during pre-monsoon and monsoon when compared to winter and post-monsoon (table 1). Black carbon mass concentration is higher during winter and post-monsoon in Ahmedabad. TOA forcing due to BC aerosols is more or less the same throughout the year in Ahmedabad. This suggests that the higher surface albedo during pre-monsoon and monsoon compensates the decrease in BC aerosols and gives rise to similar TOA forcing. TOA forcing for water soluble and BC aerosols in winter are of the same magnitude but have different signs, and cancel each other, thereby yielding a low net TOA forcing (figure 7).

The atmospheric forcing, calculated as the difference between TOA and SFC forcing, can be higher or lower depending on aerosol type. For scattering particles (sulfate) TOA forcing is negative (figure 7), and the surface forcing has the same sign and similar magnitude as that of TOA forcing, yielding a lower atmospheric warming. In the case of absorbing aerosols (black carbon) TOA forcing is positive, the surface forcing is of opposite sign (negative) and is about 3 times larger than TOA forcing, thus resulting in a higher atmospheric warming (figure 7). Black carbon aerosols dominate the surface and atmospheric forcing over an urban location throughout the year (figure 7). BC contributes 2% (monsoon)–7% (post-monsoon) to the total mass, which translates into an AOD contribution of 22%–37% respectively (table 2, figure 4). Nevertheless the percentage contribution of BC to net atmospheric forcing is >65% throughout the year, suggesting that BC aerosol contributes significantly to warming on regional scales. Black carbon aerosols contribute about 55%–70% of the aerosol radiative forcing at the surface. The large positive atmospheric warming and negative surface forcing can contribute to solar dimming, and can significantly influence the atmospheric stability and cloud formation in the tropics, thereby leading to a weaker hydrological cycle (e.g., Solomon et al. 2007). Though the contribution of natural aerosols (dust and sea salt) is higher (69%–82%) to the total mass concentration (figure 4), it is clear that anthropogenic aerosols (water soluble and black carbon aerosols) dominate (57%–82%) the optical properties and the radiative effects (mainly black carbon, >50%). This confirms that aerosol radiative forcing is dependent more on AOD and the chemical

---

**Figure 5.** Seasonal mean single scattering albedo from observations in comparison with model estimates.

**Figure 6.** Correlation between diurnally averaged, clear sky, measured and modeled global (direct + diffuse) fluxes (W m\(^{-2}\)) over Ahmedabad in 2008.
Figure 7. Top of the atmosphere aerosol radiative forcing due to water insoluble, water soluble, black carbon, sea salt and mineral dust aerosols in (a) winter (0.2), (b) pre-monsoon (2.6), (c) monsoon (2.5) and (d) post-monsoon (1.8). Percentage contribution of above aerosol species to atmospheric forcing during (e) winter (43.5), (f) pre-monsoon (51.1), (g) monsoon (41.4) and (h) post-monsoon (50.5). Percentage contribution of different aerosol species to aerosol radiative forcing at the surface in (i) winter (−43.3), (j) pre-monsoon (−48.5), (k) monsoon (−38.9) and (l) post-monsoon (−48.7). Total aerosol radiative forcing at the top of the atmosphere, in the atmosphere and at the surface are given in brackets.

4. Concluding remarks

Measured aerosol characteristics have been used in conjunction with an aerosol optical properties model and a radiative transfer model to estimate aerosol radiative forcing due to natural and anthropogenic aerosols over Ahmedabad, an urban, industrialized location in western India. Aerosol optical depths in the 0.38–1.02 μm spectral range and single scattering albedo in the 0.45–0.70 μm wavelength range measured during 2008 over Ahmedabad are used in the study. Ground reaching broad-band global and diffuse fluxes in the 0.31–2.8 μm wavelength range measured during 2008 over Ahmedabad are utilized.

The major conclusions from the study include:

Mid-visible aerosol optical depth (AOD) does not show a strong seasonal variation and is about 0.4 over Ahmedabad during 2008. α, the Ångström exponent, exhibits significant seasonal variation. α is higher during winter and post-monsoon when fine mode aerosols are dominant, while α composition (SSA) than on the mass concentration of aerosols, and there exists no linear relation between aerosol mass, optical depth and their radiative contribution.
is lower during pre-monsoon and monsoon when coarse mode aerosols are dominant. The contribution of mineral dust is >55% to the total mass, as the study location is in a semi-arid region. The contribution of all the aerosol species exhibit significant seasonal variation. The contribution of natural aerosols (dust and sea salt) is higher than 65% to the total mass concentration. While, the anthropogenic aerosols (water soluble and black carbon) dominate the aerosol optical depth by contributing more than 57% to the total.

SSA is lower than 0.82 during winter and post-monsoon owing to the dominance of fine mode absorbing aerosols. SSA is higher during pre-monsoon and monsoon due to the increase in the abundance of mineral dust and sea salt aerosols, which are more scattering in nature.

The correlation between the modeled and measured radiative fluxes is very high ($r^2 = 0.99$), confirming the robustness of the proposed approach to derive the input aerosol parameters and fluxes.

Black carbon aerosols contribute 2% (monsoon)–7% (post-monsoon) to the total aerosol mass, which results in a contribution of 22%–37% respectively to aerosol optical depth. The percentage contribution of black carbon to the net atmospheric forcing is >65% throughout the year, suggesting that black carbon aerosol contributes significantly to warming on regional scales. Black carbon aerosols contribute 50% or more to the aerosol radiative forcing at the surface, which has important implications to solar dimming and hydrological cycle.

Results reveal that the aerosol radiative forcing is governed more by AOD and SSA rather than their mass, and there exists no linear relation between the mass, optical depth and radiative effect of different aerosol species.

Both scattering and absorbing aerosols cool Earth’s surface, while their radiative effects in the atmosphere vary with altitude. For a perfect scatterer (e.g., sulfate, sea salt) in the shortwave, the top of the atmosphere aerosol forcing is quite similar to the surface forcing. In contrast, for absorbing aerosols (e.g., BC) surface forcing is 2–3 times larger than the top of the atmosphere forcing and, thus, absorbing aerosols result in a larger atmospheric warming. For the same type of aerosols (same SSA) aerosol radiative forcing at the surface is linearly related to AODs. However, the top of the atmosphere aerosol forcing significantly depends on SSA, and can be either positive (lower SSA) or negative (higher SSA), unlike the greenhouse gas forcing, which is always positive. In addition, TOA forcing can become less negative and/or positive when aerosols with lower SSA are present in abundance over high reflecting surfaces (such as continent, desert, snow) (Solomon et al 2007). Therefore, in locations where absorbing aerosols are less abundant, the surface aerosol forcing will be governed more by the AODs and to a lesser extent by the SSA. The methodology is general and can be widely and effectively used over a variety of environmentally distinct regimes to determine quantitatively the natural and anthropogenic fraction in the measured aerosol properties and in radiative forcing, and their temporal and regional variations.

Acknowledgments

Relative humidity and water vapor are obtained from NCEP Reanalysis. Columnar ozone values are downloaded from the GES-DISC, NASA. Winds are downloaded from www.cdc.noaa.gov. Rainfall over Ahmedabad is obtained from NOAA, NESDIS National Climatic Data Center (NCDC), USA.

References

d’Almeida G A, Koepe P and Shettle E P 1991 Atmospheric Aerosols: Global Climatology and Radiative Characteristics (Hampton, VA: A. Deepak Publishing) p 561
Ganguly D, Jayaraman A and Gadhavi H 2006 Physical and optical properties of aerosols over an urban location in western India: seasonal variabilities J. Geophys. Res. 111 D24206
Hess M, Koepe P and Schult J 1998 Optical properties of aerosols and clouds: the software package OPAC Bull. Am. Meteorol. Soc. 79 831–44
Jayaraman A, Lubin D, Ramachandran S, Ramanathan V, Woodbridge E, Collins W D and Zalpuri K S 1998 Direct observations of aerosol radiative forcing over the tropical Indian Ocean during the January–February 1996 pre-INDOEX cruise J. Geophys. Res. 103 13827–36
Kedia S and Ramachandran S 2011 Seasonal variations in aerosol characteristics over an urban location and a remote site in western India Atmos. Environ. 45 2120–8
McClatchey R A, Fenn R W, Salby J E A, Volz F E and Garing J S 1972 Optical Properties of the Atmosphere (Environ. Res. Pap.) 3rd edn, vol 411 (Cambridge, MA: Air Force Cambridge Research Laboratory) p 108
McComiskey A, Schwartz S E, Schmid B, Guan H, Lewis E R, Ricchiazzi P and Ogren J 2008 Direct aerosol forcing: calculation from observables and sensitivities to inputs J. Geophys. Res. 113 D09201
Pilinis C, Pandis S N and Seinfeld J H 1995 Sensitivity of direct climate forcing by atmospheric aerosols to aerosol size and composition J. Geophys. Res. 100 18739–54
Ramachandran S and Rajesh T A 2008 Asymmetry parameters in the lower troposphere derived from aircraft measurements of aerosol scattering coefficients over tropical India J. Geophys. Res. 113 D16212
Ricchiazzi P, Yang S, Gautier C and Bowle D 1998 SBDART, a research and teaching tool for plane-parallel radiative transfer in the Earth’s atmosphere Bull. Am. Meteorol. Soc. 79 2101–14
Solomon S et al (ed) 2007 Climate Change 2007. Fourth Assessment Report of the Intergovernmental Panel on Climate Change (New York: Cambridge University Press)
Srivastava R and Ramachandran S 2010 Aerosol radiative forcing over an urban location: observations and model estimates Indian Aerosol Sci. Technol. Assoc. Bull. 19 387–9
Srivastava R and Ramachandran S 2012 The mixing state of aerosols over the Indo-Gangetic Plain and its impact on radiative forcing Q. J. R. Meteorol. Soc. at press (doi:10.1002/qj.1958)
Textor C et al 2006 Analysis and quantification of the diversities of aerosol life cycles within AeroCom Atmos. Chem. Phys. 6 1777–813