

Comments on “A Method to Estimate Aerosol Radiative Forcing from Spectral Optical Depths”

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Satheesh and Srinivasan (2006, henceforth SS06) discussed a method to determine aerosol radiative forcing from spectral optical depths. A more refined methodology applicable to a wide variety of environments, which employs stringent criteria and is statistically rigorous, for the first time has been published earlier by Ramachandran and Jayaraman (2003), the details of which are given below. In addition the shortcomings in the method given in SS06 are pointed out in this work.

The details given in the first few pages of SS06, including Eqs. (1)–(6), are well known. The calculation of aerosol optical depths (AODs) from the extinction (scattering + absorption) is done using the well-known Beer–Lambert’s law and Mie theory. Information on different properties and refractive indices of aerosols given in Tables 1–3 of SS06 are taken from Hess et al. (1998), as well as the information required to plot Fig. 1 of SS06. It is well known that a given change in any individual aerosol species would affect the spectral variation of AODs differently. At a given location the aerosol population will have contributions from local sources and long-range transport. Number concentration of each aerosol component is important in determining the composite aerosol optical depth; relative humidity can affect the mode radius and the number concentration of both the smaller- (sulfate) and larger-size (sea salt) particles. For example, during the Indian Ocean Experiment (INDOEX; Ramanathan et al. 2001) it was shown that continental aerosols can be transported thousands of kilometers across the Indian Ocean and can contribute up to 80% to the measured AODs. Hess et al. (1998) clearly mention that all the

aerosol types defined may have additional components with varying number densities according to the actual location for which they are assumed to be valid. It is stated that in coastal areas an additional sea salt component would be present and urban locations, which are near arid regions, will be influenced by mineral aerosols.

SS06 state that they make an initial assumption of the zero-order aerosol composition, such as continental clean, marine polluted, desert, urban, etc., which are given in Hess et al. (1998). Then the scaling factors (S_i), which are the total number densities of the respective aerosol components, are varied iteratively to make the absolute values of spectral optical depths simulated by aerosol mixtures consistent with those estimated from observations. Ramachandran and Jayaraman (2003) adopted the same methodology and then derived the aerosol optical depth spectra. Ramachandran and Jayaraman (2003) wrote,

We maintained the aerosol components that form the aerosol type but changed the number concentrations to derive the AODs. These number concentrations may not be unique, but we scrutinize our choice of the aerosol components and their number concentrations. We looked into the number mixing ratios, mass ($\mu\text{g m}^{-3}$) and the single scattering albedos (ω), and compared with the standard aerosol types given in Hess et al. (1998).

Several stringent criteria were adopted in choosing the particular optical depth spectra, which are discussed in detail in Ramachandran and Jayaraman (2003) and Ramachandran (2005a,b). Briefly, for the measured aerosol optical depth spectra several estimated AOD spectra are reconstructed using the different aerosol components given in Hess et al. (1998). Following the criteria listed below a suitable estimated AOD spectrum is then chosen (Ramachandran and Jayaraman

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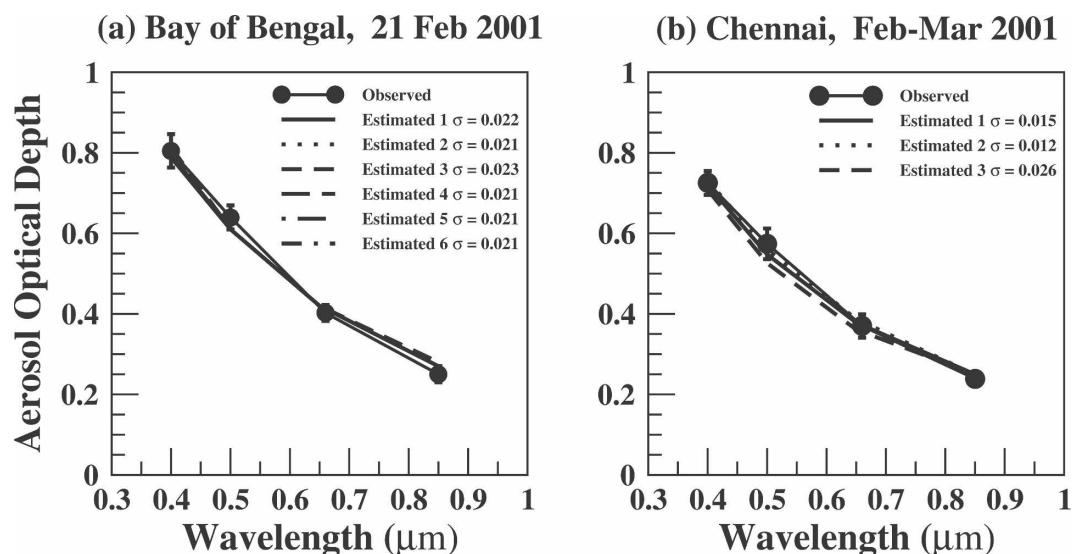


FIG. 1. Comparison of the measured aerosol optical depth spectra over (a) Bay of Bengal (21 Feb 2001) and (b) Chennai (February–March 2001) with the model estimated aerosol optical depths. The estimated AOD spectra are obtained using the Optical Properties of Aerosols and Clouds (OPAC) software, taking into account the measured relative humidity values and aerosol components suggested by Hess et al. (1998). See Table 1 for more details.

2003). The criteria employed are 1) the sigma value (Eq. 9 of SS06) between the measured and the fitted aerosol optical depth spectra, which should be in the 0.02–0.04 range, 2) the feasible combination of atmospheric aerosol components (by comparing the ω values with known results and aerosol models), and 3) a good agreement between the measured and estimated aerosol mass concentrations. It was shown that a priori the ambience or the environment of the location where AOD measurements were made should be known to choose the proper AOD spectra (Ramachandran and Jayaraman 2003).

In Fig. 1 and Table 1 the number density of aerosol components that contribute to the aerosol optical depths measured over a polluted oceanic region (20.05°N, 89°E in Bay of Bengal on 21 February 2001) and a continental location (Chennai, 13.04°N, 80.17°E, population \sim 4.2 million, an urban industrial city on the eastern coastline of India, February–March 2001) (Ramachandran and Jayaraman 2003) are given and plotted. Aerosol optical depths measured over Bay of Bengal and Chennai could be estimated by using insoluble, water soluble, and soot aerosol components. It is to be noted that AOD spectra measured over Bay of Bengal on 21 February 2001 could be fitted with six different combinations of the above-specified aerosol components. The February–March mean AOD spectra over Chennai could be fitted with three different compositions with varying number density of each species. Note that the σ value in Eq. (9) of SS06 is about 0.02 in all six

cases. For Chennai σ in fact is found to be 0.015 and 0.012 in two cases, respectively, while the third case has a σ of 0.026. Note that these σ values are smaller than the error in the measured AODs, which is about 15% (Ramachandran and Jayaraman 2003).

The fitted AOD spectra are almost indistinguishable from the observed AOD spectra (Fig. 1). The second case of AOD spectra measured in Chennai, which is reconstructed only with water soluble aerosol particles, expectedly has a higher ω value of 0.987. It should be noted that a typical densely populated urban location will have other aerosol components, such as soot, dust, etc., in addition to sulfate particles. The third case for Chennai, as well as for Bay of Bengal, clearly illustrates the need to know a priori the ambience where the aerosol measurements were made to choose the proper aerosol spectra and reject the other ones. If we did not know that the AOD measurements were made in an urban or over a polluted ocean we would have chosen the fitted AOD spectra from case 3 and would have obtained an atmospheric forcing greater than 100 W m^{-2} , which is erroneous. It is clear from Table 1 that for the same optical depth spectra the aerosol radiative forcings can vary for different ω values; it is shown that when ω increases the surface forcing decreases. The surface aerosol radiative forcing over Bay of Bengal for case 1 is about -35 W m^{-2} , when $0.5\text{-}\mu\text{m}$ ω is 0.947, while for case 6 with a ω of 0.788 the forcing is about -56 W m^{-2} . Note that between the two cases ω values differ by less than 20%, while the variation in forcings

TABLE 1. Composition of aerosol types estimated for measured aerosol optical depth spectra and the computed single scattering albedos (ω) at 0.5 μm over Bay of Bengal and Chennai (Ramachandran and Jayaraman 2003). Aerosol radiative forcings at the top of the atmosphere (TOA), at the surface (SFC), and in the atmosphere (ATM) for Bay of Bengal and Chennai (Ramachandran 2005b) are given.

Date (No.)	No. density of components (cm^{-3})			ω (0.5 μm)	Aerosol radiative forcing		
	1 ^a	2 ^b	3 ^c		TOA	SFC	ATM
21 Feb 2001 (1)	0.3	27 800	34 300	0.934	-16.8	-34.9	18.1
21 Feb 2001 (2)	0.6	26 700	53 000	0.905	-13.1	-37.6	24.5
21 Feb 2001 (3)	6×10^{-5}	3300	462 000	0.309	5.1	-105.8	110.9
21 Feb 2001 (4)	0.3	58 500	54 300	0.947	-17.6	-33.0	15.4
21 Feb 2001 (5)	0.3	49 500	205 800	0.838	-11.7	-49.6	37.9
21 Feb 2001 (6)	—	45 500	274 400	0.788	-9.5	-56.2	46.7
Location (No.)							
Chennai (1)	0.15	19 600	130 000	0.774	8.8	-52.9	61.7
Chennai (2)	—	28 000	—	0.987	-10.7	-20.0	9.3
Chennai (3)	—	140	445 000	0.230	37.8	-108.3	146.1

^a Insoluble: soil particles with a certain amount of organics.

^b Water-soluble: particles from gas to particle conversion that contain sulfates, nitrates, organic, and water soluble substances.

^c Soot: absorbing black carbon (Hess et al. 1998).

is higher than 35%. Thus it is clear that for a reliable estimate of aerosol radiative forcing it is essential to know both columnar aerosol optical depths and ω values.

It is difficult to state that soot and sulfate contribute to smaller size particles while dust and sea salt contribute in the larger particle mode. Several observational studies in different environments have shown that, for example, sulfate particles were not present as pure sulfates, and instead were found mixed with organic matter and soot in the submicron-size range and with sea salt and dust in the supermicron-size range. Dust and sea salt also contain small-size particles (Chung et al. 2005). Not only are the mass of absorbing aerosol species (e.g., soot and dust) and their percentage contribution to the total mass important in determining the ω value, the size distribution to a greater extent, is crucial, as the value of ω (whether high or low) is determined by the ratio of the number of absorbing to scattering particles in a size distribution. Considering these results and the earlier discussion it is not clear how one can differentiate scattering and absorbing aerosols using the approach proposed by SS06.

SS06 state that using their proposed approach the mean difference between observed and estimated forcing is $\pm 2 \text{ W m}^{-2}$, which was about 6% of the aerosol forcing reported over the tropical Indian Ocean, and referred the work of Satheesh and Ramanathan (2000). Satheesh and Ramanathan (2000) have stated that the mean forcing value at the surface in February and March 1998 was -12 W m^{-2} . Taking this value a $\pm 2 \text{ W m}^{-2}$ difference would correspond to 17% and not 6% as mentioned by SS06. In addition, in Fig. 8a of SS06

the observed forcing values are found to vary from 5 to 20 W m^{-2} . The mean difference plotted in Fig. 8b does not exhibit any significant AOD dependence and is found to vary by about $\pm 2 \text{ W m}^{-2}$ in the AOD range of 0.1 to 0.3. Taking into account these variations the estimated uncertainty in aerosol radiative forcings will be higher and can vary from 10% to 40%, depending on the observed fluxes and AODs.

Figures 9 and 10 in SS06 make a comparison of the estimated and measured absorption AODs over six island locations, which are located over the Atlantic, Pacific, and Indian Oceans. It is to be noted that the measured absorption AODs are low; most of the absorption AODs are less than 0.05 in these chosen locations, going as low as 0.015. The number of observations for which correlation between measured and estimated absorption AODs obtained by SS06 varies by more than an order of magnitude between the six chosen locations, the lowest being 11 (Reunion St. Denis) and the highest 1021 (Mauna Loa). Note that in SS06 only the absorption AODs are compared, in contrast Ramachandran (2005a), using the methodology discussed in Ramachandran and Jayaraman (2003), showed that the correlation between the total (scattering + absorption) measured and estimated AODs over coastal India, the Arabian Sea, and the tropical Indian Ocean was greater than 0.98 where the AODs varied in the range of 0.05 to 0.5. Chung et al. (2005) estimated the annual mean ω (ratio of scattering AOD to the total AOD) values for the globe, Northern Hemisphere (NH), Southern Hemisphere (SH), land, and ocean. The 0.55- μm ω values were found to be 0.942 (global), 0.939 (NH), 0.947 (SH), 0.928 (land), and 0.952 (ocean). These ω values

clearly indicate that the absorption AOD on a global mean scale contributes a maximum of only 7% to the total AOD.

SS06 compare the ω values obtained from earlier measurements/campaigns and the ω values estimated using the proposed method. SS06 compare the measured and estimated black carbon mass concentrations over Bay of Bengal during April/May and October/November and in Bangalore, a continental location. It should be noted that the above comparison is for the “surface” level measured black carbon mass concentrations, while the other comparisons, which include the Scientific Training and Access to Aircraft for Atmospheric Research throughout Europe-Mediterranean (STAAARTE-MED) experiment and Zambian International Biomass Burning Emissions Experiment (ZIBBEE) are either for the columnar measurements or column-integrated values obtained by integrating the vertical profiles of aerosol characteristics. The columnar aerosol optical properties are dependent on the scale height, which varies from 1 km over the oceans to 8 km over the continent (Hess et al. 1998). The columnar aerosol property is a function of the aerosol burden throughout the column and not only at the surface. The surface and columnar aerosol properties can be entirely different because of the presence of aerosol layers above (Ramanathan et al. 2001). Incidentally the reference of Eck et al. (2001) given by SS06 in the paper is wrong as it corresponds to Maldives in the Indian Ocean and not to the ZIBBEE field campaign as stated in SS06.

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