

Balloon-borne study of the upper tropospheric and stratospheric aerosols over a tropical station in India

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ABSTRACT

Using sun-scanning/tracking photometers onboard a high-altitude balloon the upper tropospheric and stratospheric aerosol characteristics have been studied over Hyderabad (17.5°N) in April 2001. In the upper troposphere the aerosol extinction coefficients are found to be around 10^{-2} km^{-1} , while in the stratosphere the values are in the range 10^{-3} – 10^{-4} km^{-1} , which are about two orders of magnitude smaller than those measured after the Pinatubo eruption over the same site. The balloon-derived aerosol extinction coefficients are in good agreement with the results obtained from lidar experiments conducted over Mt. Abu (24.6°N). In the upper troposphere the integrated aerosol extinction coefficients at 500 nm are found to show an average increase of about $11 \pm 1\% \text{ yr}^{-1}$ during the period 1985–2001. This increase is found to be consistent with the observed increase in the long-term columnar aerosol optical depth data over selected locations in India. The Ångström wavelength exponent is found to be about 1.33 ± 0.12 in the stratosphere, which is comparable to the Junge size parameter values obtained in 1985. The mode radii of stratospheric aerosols are found to be $0.10 \pm 0.01 \mu\text{m}$ in both the 2001 and 1985 experiments, indicating that the background stratospheric aerosol size distribution has not changed between these years, which is also corroborated by the good agreement between the wavelength exponent and size parameter values obtained in 2001 and 1985. The aerosol optical depth at $1 \mu\text{m}$ in 2001 is about 0.005, which is five times larger than that measured by SAGE in 1979 over the tropics. The stratospheric aerosol optical depth spectra are calculated for both the volcanic and quiescent conditions by taking into account the mode radius and aerosol number density. The estimated and measured aerosol optical depths agree well. We estimate that the sulfur dioxide and aerosol emissions from coal over India have increased by $10\% \text{ yr}^{-1}$ during the past two decades. The increase in emissions from high-speed diesel oil and petrol is higher and is in the range 13 – $14\% \text{ yr}^{-1}$ over India. These increases could possibly be responsible for the observed increase in the upper tropospheric aerosol optical depths over India.

1. Introduction

Vertical profiles of aerosol physical and optical properties are important in understanding the role aerosols play in altering the radiation budget of the Earth's atmosphere. The role of aerosols in altering the atmospheric radiation budget and its possible climate impact has been recognised for a long time. In

the atmosphere aerosol characteristics are altitude dependent and the stratospheric aerosols are quite different from the lower, tropospheric aerosols. Unlike tropospheric aerosols, which are short-lived due to gravitational settling and rainwash and can produce regional and seasonal effects, stratospheric aerosols are long-lived and can produce long-term global effects.

Long-term measurements of aerosol characteristics in the upper troposphere from Stratospheric Aerosol and Gas Experiment (SAGE) I and II showed that the $1 \mu\text{m}$ aerosol optical depth could have undergone

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an increase of a maximum of about $1\% \text{ yr}^{-1}$ when averaged over both hemispheres (Kent et al., 1998). From the 20-yr high-altitude balloon-borne measurements at a midlatitude station at Laramie (41°N), Wyoming, Hofmann (1993) found that there is evidence for a decreasing trend of $1.6\text{--}1.8\% \text{ yr}^{-1}$ in the optically active tropospheric aerosol. The sources of upper tropospheric aerosols and their precursor materials lie both in the stratosphere and in the lower troposphere. The aerosols in the upper troposphere have a longer residence time of a few months when compared to about a week in the lower troposphere. In the lower troposphere aerosols can be produced from surface-blown dust (Tegen and Fung, 1994), biomass burning (Kaufman et al., 1990), biogenic production (Charlson et al., 1987) and industrial effluents (Charlson et al., 1991). These sources exhibit distinct geographical characteristics and can consequently be expected to show differing signatures in the upper troposphere (Kent et al., 1998). A multiyear dataset on upper tropospheric aerosol characteristics from SAGE II showed that there are two main influences on the upper tropospheric aerosol, namely seasonal lifting of material from below and downward transfer of volcanic aerosol from the stratosphere (Kent et al., 1995).

Long-term observations of stratospheric aerosols in the past three decades indicate that the stratosphere has been volcanically disturbed during 90% of the time (Schwartz et al., 1995). Balloon-borne and lidar observations over the northern midlatitudes show that temporal minima were observed in 1979 and in the period 1989–1991 (prior to the Mt. Pinatubo eruption) (Hofmann, 1990; Jäger, 1991; Osborn et al., 1995). During these time periods the stratospheric aerosol loading showed generally stable levels, and these periods are referred to as non-volcanic or background periods. Thomason et al. (1997) compared the global stratospheric aerosol levels between 1979 and 1989–91 periods using Stratospheric Aerosol Measurement (SAM) and SAGE II data. They found that, depending on latitude, the $1\text{-}\mu\text{m}$ aerosol optical depth in 1989–91 was 10–30% higher than that observed in 1979. Balloon-borne data obtained on stratospheric aerosols at Laramie in the 1979–90 period indicate that the background stratospheric aerosol mass has increased by $>5\% \text{ yr}^{-1}$ (Hofmann, 1990).

After being dormant for about 6.5 centuries, the Mt. Pinatubo volcano situated in the Philippines erupted in 1991, producing the largest impact at the strato-

spheric altitudes. The stratospheric aerosol depth following the Pinatubo eruption increased by about two orders of magnitude (McCormick and Veiga, 1992). There has been no major volcanic eruption since then, and as the residence time of stratospheric aerosols is about 3 yr, the stratosphere is in quiescent condition now. The present balloon experiment was conducted to study the characteristics of aerosols over a tropical station in India and to examine long-term changes (if any) in the upper troposphere and stratosphere during volcanically quiescent conditions.

2. Instrumentation

Two multichannel photometer systems, a sun-scanning and a sun-tracking model (Ramachandran et al., 1994a), are employed onboard the balloon to measure the direct as well as the angular distribution of the scattered radiation intensities. The sun-scanning instrument consists of a sensor assembly containing six filter photometers, the sun-tracking mechanism, and a motor assembly for scanning the sky along the solar almucantar, $\pm 90^\circ$ with respect to the sun for scattered sky radiation measurements. The scanning is performed in 18 s corresponding to an altitude ascent of about 90 m by the balloon, which is the lower limit of the altitude resolution of the various quantities that are measured. The spectral bands of the photometers are centered around 290, 310, 500, 670, 850 and 1050 nm, with a typical bandwidth of about 10 nm or less. The sun-tracking photometer, an automatic axis stabilised system was employed to measure altitude profiles of the direct solar radiation intensities uninterruptedly. The wavelengths used in the sun-tracking system are 290, 430, 500, 850, 950 and 1050 nm, with similar bandwidths. Four of the wavelengths are similar to the sun-scanning system to cross-validate the data. The data corresponding to 290 and 310 nm are aimed at retrieving the vertical profiles of ozone, and the 950 nm data are intended for deriving the water vapor profile. The 500, 670, 850 and 1050 nm channels are used for aerosol studies the results of which are presented here. Instruments more or less similar to the present configuration were flown successfully from Hyderabad in 1985 (Jayaraman and Subbaraya, 1988). Two balloon flights were also conducted in 1991 and 1992 using similar instruments, and Pinatubo volcanic aerosol layer evolution and decay at stratospheric altitudes have been studied (Ramachandran et al., 1994a;b).

3. Experiment

A high-altitude balloon (54 000 m³ volume) was launched on 10 April 2001 from the National Balloon Facility located in Hyderabad, India (17.5°N, 78.6°E). The time history of the balloon flight is shown in Fig. 1. The hydrogen gas filled balloon was launched at around 0626 h (local time). The balloon reached the expected ceiling altitude of about 33.5 km by around 0900 h, with an average ascent rate of about 220 m min⁻¹. Ground-based radar of the India Meteorological Department, Hyderabad and a global positioning system tracker kept in the payload assembly were used for balloon tracking. This provided continuous information on balloon position (latitude and longitude of the balloon) and its height in kilometres above the mean sea level during all important phases of the flight. After about an hour of float duration, the balloon was brought down to about 20 km in about 2 h by releasing the gas through an apex valve incorporated in the balloon. The flight was terminated around 1300 h and the instruments were released on a parachute and recovered. Data were collected during both ascent and descent, and the results presented here are the average of both the data wherever applicable. During the experiment all the scientific as well as technical instruments, such as radiosonde and telecommands, worked satisfactorily. Data corresponding to an altitude region of 5–33 km are presented.

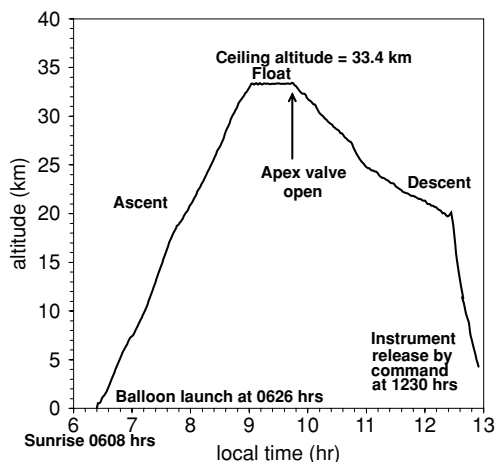


Fig. 1. The altitude trajectory for the balloon flight conducted on 10 April 2001.

4. Results and discussion

4.1. Aerosol extinction coefficients

The data analysis mainly involves the estimation of the attenuation of the incoming solar radiation at each altitude. If I is the intensity of the solar radiation at altitude z , then the total atmospheric extinction coefficient β (km⁻¹) at z can be written using the Beer–Lambert law as

$$\beta(z) = \frac{dI}{I(z) dz \sec \chi} \quad (1)$$

where χ is the solar zenith angle at the time of observation and $\sec \chi$ gives the atmospheric airmass. The total atmospheric extinction coefficient β is made up of

$$\beta = \beta_{\text{ma}} + \beta_{\text{rs}} + \beta_{\text{aerosol}} \quad (2)$$

where β_{ma} is the absorption coefficient due to molecular gases such as ozone, nitrogen dioxide, water vapor etc., β_{rs} is the Rayleigh scattering coefficient (scattering due to air molecules), and β_{aerosol} is the aerosol extinction coefficient.

The air density profile (not shown) constructed from the meteorological balloon soundings on the flight day 2 h prior to the launch of the balloon and the mean ozone density available over Hyderabad (Lal et al., 1989) are used to correct the extinction coefficient profiles for Rayleigh scattering and ozone absorption. Absorptions due to nitrogen dioxide in the 400–450 nm region, as well as by water vapor at 800 nm, are less than 1% of the total extinction coefficients at these wavelengths (Jayaraman and Subbaraya, 1993). For the altitudes where measured air density values are not available, NASA (1966) data for 15°N are used. The measured and the standard atmosphere air densities are found to exhibit a very close correspondence. A broad tropopause is seen on the flight day, starting from 16 km and extending up to about 18 km with temperatures < -80 °C.

Using the sun-tracking mechanism the solar radiation intensities, $I(\lambda)$ are measured with an accuracy better than 1%. The analog I values are digitised and recorded on magnetic tapes for detailed analysis. However, as the aerosol extinction coefficients are derived from the total extinction coefficients after correcting for the contribution due to scattering and absorption by the molecular species, the accuracy of the final results depends on the accuracy of the input parameters. In the absence of meteorological radiosonde data on

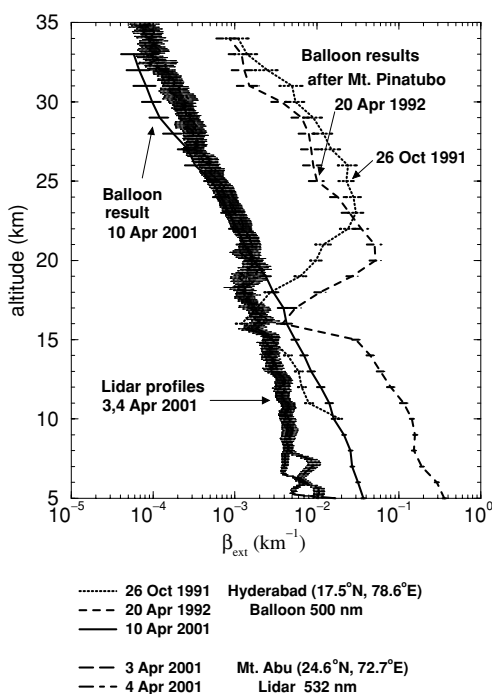


Fig. 2. Aerosol extinction coefficient profile obtained in April 2001 over Hyderabad (17.5°N, 78.6°E). The aerosol extinction coefficients are compared with the profiles obtained over the same site in 1991 and 1992 after the Pinatubo eruption. Results obtained from the Nd:YAG lidar measurement conducted on 3 and 4 April 2001 over Mt. Abu (24.6°N, 72.7°E) are also shown for comparison.

the air density profile, model values (NASA, 1966) are used above 25 km. In general, the derived aerosol extinction coefficient profiles have a maximum uncertainty of 10% in the lower altitudes, which increases to about 30% at stratospheric altitude levels.

Figure 2 shows the aerosol extinction coefficient profile obtained after correcting for molecular scattering and absorption at 500 nm. The aerosol extinction coefficient profiles obtained after the Pinatubo eruption on 26 October 1991 and 20 April 1992 (Ramachandran et al., 1994a) from the same location are shown for comparison. Results obtained from the Nd:YAG lidar system (Jayaraman et al., 1995) operating at 532 nm over Gurushikhar (24.6°N, 72.7°E) on Mt. Abu on 3 and 4 April 2001 (a week before the balloon flight) are also shown.

One of the uncertainties in determining an aerosol vertical profile from lidar measurement arises due to the lidar ratio, which is the ratio of the backscatter-

ing coefficient to extinction, expressed as B_a , and has the units sr^{-1} . For a given wavelength B_a depends on the aerosol size distribution, which can be different at different altitudes. Using lidar and simultaneous balloon-borne optical particle counter data Jäger and Hofmann (1991) have obtained values for B_a at 532 nm. The observations covered the period 1980–1987 and hence include the effects of the 1982 El Chichon (17.3°N, 93.2°W) volcanic aerosols on aerosol extinction. Balloon-borne optical particle counter measurements were made over Laramie, Wyoming (41°N), and lidar measurements were made over Garmisch-Partenkirchen (47.5°N). From their results we calculated the average values of B_a for the lower stratospheric altitudes (15–30 km) from 1980 until the El Chichon eruption (i.e. April 1982) and then for 1986 and 1987. This corresponds to about 4 yr of data. The values are found to be in the range 0.017–0.022 with a mean value of about 0.019. The lowest B_a value during these years was 0.016 and the highest was 0.03. In our calculations we derived the aerosol extinction coefficients for three B_a values, namely 0.015, 0.02 and 0.025, which include the measured range of values over Garmisch. The uncertainty in the derived aerosol extinction using the above B_a values is of the order of 20–25% in the 15–30 km region, while it is less than 20% in the 5–15 km altitude region.

The aerosol extinction coefficient obtained on 10 April 2001 is about $4 \times 10^{-2} \text{ km}^{-1}$ at 5 km, which decreases by an order of magnitude in the tropopause region (Fig. 2). The aerosol extinction decrease further in the lower stratosphere and reaches a minimum of about $5 \times 10^{-5} \text{ km}^{-1}$ at 33 km. The lidar-derived aerosol extinctions on 3 and 4 April 2001 are in close agreement with the balloon-measured data in the lower stratosphere. In the troposphere the lidar-derived extinctions are lesser than the balloon results. The lidar observation site at Mt. Abu is located at about 1.7 km above MSL and is a cleaner region devoid of any low-altitude atmospheric pollution and has a population of only about 35 000. Hyderabad is densely populated urban/industrial city with a population of about 3.5 million. Similar differences in the lower atmosphere have been seen before during April 1992 (Ramachandran et al., 1994a). We see that there can even be day-to-day differences in the values in the lower atmosphere: the profile obtained on 3 April shows a peak around 7 km while on 4 April there is a decrease around the same altitude. The aerosol extinction coefficients in the lower stratosphere are about two orders of

magnitude smaller than those measured after the Pinatubo eruption.

4.2. Upper troposphere

The features of upper tropospheric aerosols are much less documented when compared to the lower tropospheric or for that matter the stratospheric aerosols, in spite of the fact they are important in understanding the global change and long-range transport mechanisms. Long-term measurements on upper tropospheric aerosols by satellites are hampered by high-altitude clouds (Kent et al., 1998). Here, the aerosol extinction coefficients obtained in the 5–15 km region over Hyderabad from the balloon flights conducted in 1985 and 2001 are discussed. The SAGE II upper tropospheric aerosol extinction for the entire Indian sub-continent in the latitude and longitude range of 5–35°N and 60–100°E from 1985 to 2000 is analysed for comparison. The aerosol extinction coefficients obtained in the 5–15 km altitude region are integrated and used in the study.

Four balloon flights were conducted from Hyderabad during the period 1985–2001. The aerosol extinction results obtained from experiments conducted on 26 October 1991 and 20 April 1992 are strongly influenced by the Mt. Pinatubo eruption (Ramachandran et al., 1994a) and hence are not considered in the present work. The balloon experiment of 22 October 1985 was conducted 3.5 yr after the El Chichon eruption and can be considered to represent a volcanically quiescent condition. The integrated aerosol extinction coefficients on 22 October 1985 and 10 April 2001 are 0.0092 ± 0.0006 and 0.19 ± 0.012 , respectively. This corresponds to an average increase of about $11 \pm 1\%$ yr^{-1} in the integrated aerosol extinction in the upper troposphere over the 16-yr period of analysis.

In Fig. 3 the upper troposphere (5–15 km) integrated aerosol extinction measured by SAGE II at 525 nm along with the error bars are plotted for the 1985–2000 period. The upper tropospheric aerosol optical depths are in the range of 0.01–0.02. To make a comparison with the balloon-derived results we chose the SAGE II extinction profiles for periods close to the launch of balloon flights. The SAGE II data are obtained for 10 October 1985 and 3 April 2000 (about a year earlier than the balloon flight). The values are found to be 0.0064 ± 0.0022 and 0.0096 ± 0.0025 , respectively. This corresponds to a modest increase of about 3% yr^{-1} in the upper troposphere aerosol extinction. The striking feature of the data in Fig. 3 is that the

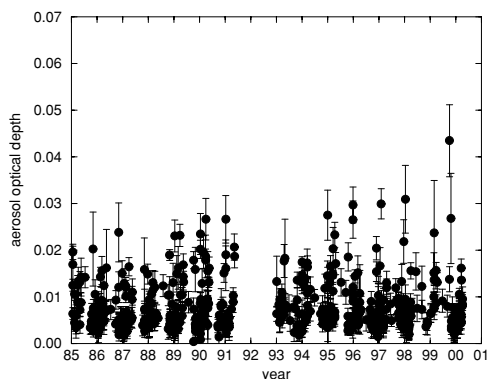


Fig. 3. Time evolution of the upper troposphere aerosol optical depth (integrated aerosol extinction coefficients in the 5–15 km altitude region) at 525 nm measured by SAGE II in the latitude–longitude range 5–35°N to 60–100°E.

variability in the aerosol optical depths has increased with time, and towards 2000 aerosol optical depth values in excess of 0.025 are present. Due to the high opacity of the atmosphere after the Pinatubo eruption reliable transmission measurements could not be made by SAGE II, and hence there are no data in the upper troposphere from June 1991 until the end of 1992. During the southwest monsoon over India (June–August) there are many fewer data due to the presence of clouds. SAGE II measures the scattered radiation by the limb-viewing technique, while the balloon-borne instrument measures the extinction. This could be one of the major reasons for the differences observed between the balloon and SAGE II results. This becomes important in the upper troposphere, as there can be aerosols comprising wind-blown dust, soot etc. Recent INDOEX results (Ramanathan et al., 2001) showed that non-sulfate particles can also be lifted to upper troposphere; as a result these particles can participate in the troposphere–stratosphere exchange. Franke et al. (2003) found a negative correlation for the northern Indian aerosols when they correlated the lidar-derived aerosol extinction and lidar ratio with relative humidity, which indicated that a large fraction of these particles was not only highly absorbing but also hydrophobic. It has been seen that, particularly over the central and north Indian regions, the dust storm activities which are quite active in the March–April period inject many dust particles into the atmosphere (Jayaraman, 1991).

Hofmann (1993) found that during the period 1972–1990 the tropospheric column burden in the optically active size range ($r \geq 0.15 \mu\text{m}$) over Laramie

decreased by about $1.6\% \text{ yr}^{-1}$. The decreasing trend could be due to a similar reduction in the SO_2 emissions in the United States over this period (Hofmann, 1993).

Over tropical India long-term measurements of columnar aerosol optical depths are being made using a network of multiwavelength solar radiometers over Trivandrum (8.5°N , 76.9°E), Mysore (12.3°N , 76.5°E) and Vishakapatnam (17.7°N , 83.2°E) (Moorthy et al., 1993). The aerosol environments have been classified as a combination of tropical, coastal and rural for Trivandrum; tropical, rural and continental for Mysore and tropical, coastal and industrialised urban for Vishakapatnam. Long-term (1986–1998) trends of aerosol optical depths obtained over these locations show that at 400 nm the AODs have increased by about $3\% \text{ yr}^{-1}$ over Trivandrum, 9% over Mysore and 2% over Vishakapatnam during the 13-yr period of analysis (Moorthy, 2001). These trends are obtained after removing inter-annual fluctuations in the AODs. Also different wavelengths are found to exhibit varied trend rates. The trends in AOD increase from a coastal ambience (Trivandrum) to a continental environment (Mysore) by a factor of three. The increase in trends could be due to the variations in meteorological processes, such as winds, sea breeze/land breeze activities in coastal stations and urban/industrial activities of local significance. Since the upper tropospheric aerosols exhibit significant regional features on the observed aerosol extinctions it is quite clear that while over northern hemisphere midlatitudes there is a decreasing trend, in the tropics there is a significant increasing trend, which is corroborated by the balloon and columnar aerosol optical depth measurements.

4.3. Mode radius

Yue and Deepak (1983; 1984) proposed a method to retrieve aerosol mode radius from the aerosol extinction coefficients β measured at two wavelengths. Assuming that the stratospheric aerosol size distribution can be best fitted using a lognormal distribution function of the form

$$\frac{dn(r)}{dr} = \frac{A}{\sqrt{2\pi} \ln \sigma} \frac{1}{r} \exp \left[-\frac{\ln^2 \left(\frac{r}{r_m} \right)}{2(\ln \sigma)^2} \right] \quad (3)$$

where A is the total number concentration (cm^{-3}), σ is the width of the lognormal curve and r_m is the mode radius, one can retrieve r_m . A value of 1.86 for σ is considered acceptable in the literature (Pinnick et al., 1976) for stratospheric aerosols.

The method involves computing β as a function of r_m for $A = 1$ using Mie theory, given by the expression

$$\beta_\lambda = \int_{r_1}^{r_2} \frac{dn(r)}{dr} Q(m, \lambda, r) \pi r^2 dr \quad (4)$$

where $dn(r)/dr$ is the number of particles per cm^3 whose radii are between r and $r + dr$, $Q(m, \lambda, r)$ is the Mie extinction efficiency factor and m is the refractive index of the aerosol particle. Mie computations are made for the selected photometer wavelengths of 500 and 1050 nm taking the m values as $1.431 - i1 \times 10^{-8}$ and $1.423 - i1.5 \times 10^{-6}$ (d'Almeida et al., 1991) for the two wavelengths, respectively. The ratio of aerosol extinctions, R , is defined as

$$R = \frac{\beta_{500}}{\beta_{1050}} = \frac{\int_{r_1}^{r_2} \frac{dn(r)}{dr} Q(m, \lambda = 500, r) \pi r^2 dr}{\int_{r_1}^{r_2} \frac{dn(r)}{dr} Q(m, \lambda = 1050, r) \pi r^2 dr} \quad (5)$$

where r_1 and r_2 are the lower and upper radii limits of the integration and chosen such that the aerosol number density falls by 1×10^{-6} with respect to the maximum value in the size distribution.

R values are plotted as function of r_m in Fig. 4 for a σ of 1.86. When R is about 1 or less the mode

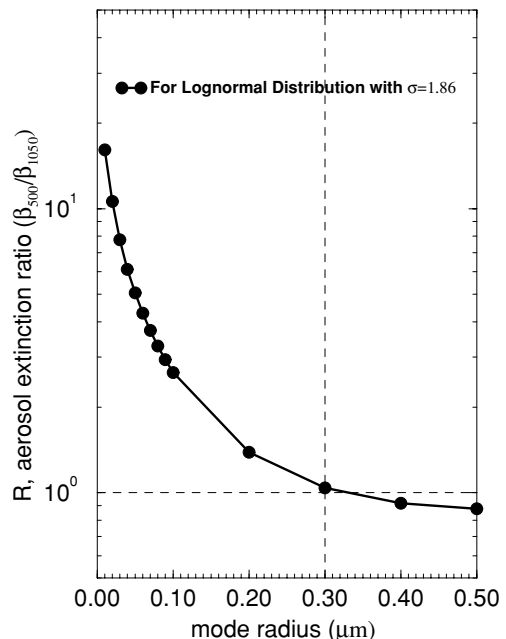


Fig. 4. The aerosol extinction ratio as a function of the variable parameter mode radius (r_m) for a lognormal distribution having a σ value of 1.86.

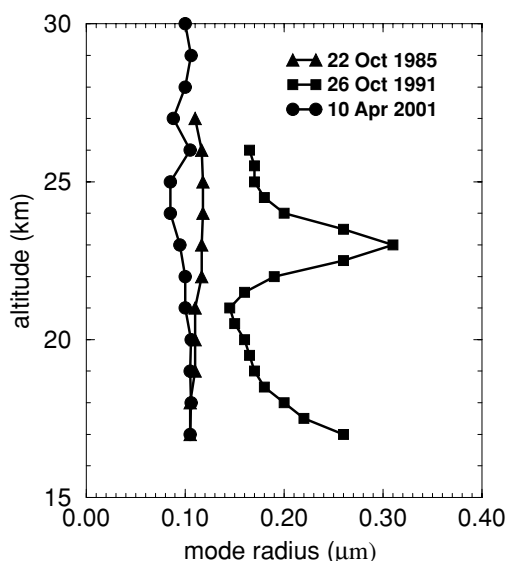


Fig. 5. Vertical profile of mode radius derived from the 10 April 2001 balloon data is compared with the mode radius profiles obtained on 26 October 1991 and 22 October 1985.

radius is above $0.3 \mu\text{m}$. For R greater than 1 the mode radius is less than $0.2 \mu\text{m}$. This indicates that when R is small the large particles dominate the aerosol size distribution, while when R is large the smaller particles dominate the size distribution. The accuracy of determining the mode radius increases with increase in R value.

The above method was successfully employed to determine the mode radius in the lower stratosphere after the Pinatubo eruption from the measurements made in October 1991. The mode radius of the aerosol layer was found to be around $0.22 \mu\text{m}$ with a prominent peak at 23 km with a value of $0.31 \mu\text{m}$ (Ramachandran et al., 1994b). The profile of mode radius obtained during April 2001 is plotted in Fig. 5. The mode radius in the lower stratosphere is found to be 0.10 ± 0.01 during April 2001. The mode radius is representative of the background aerosols in the stratosphere. The background (nonvolcanic) profile is more or less constant throughout the lower stratosphere, which is in stark contrast to the volcanically perturbed mode radius profile. The mode radius profile obtained during October 1985, a volcanically quiescent period, is also plotted for comparison. The stratospheric aerosol mode radii was found to be 0.10 ± 0.01 in 1985. This comparison indicates that the size of the background aerosols has not changed between 1985 and 2001 over

the measurement location, but the number densities have increased in 2001 when compared to 1985.

4.4. Stratospheric aerosol optical depth spectra

One of the objectives of this in situ balloon experiment was to construct spectral background and volcanic stratospheric aerosol optical depth profiles, and compare them with model estimates. Using Mie theory the aerosol extinction coefficients are calculated for the wavelength range 400–1050 nm for the mode radii measured during April 2001. The mode radii in the lower stratosphere (17–30 km) varied from a minimum of about 0.09 to a maximum of 0.11 with a mean of $0.10 \mu\text{m}$. As the stratospheric aerosols are mostly sulfate particles (75% H_2SO_4 and 25% H_2O) the refractive indices appropriate for sulfate particles are used in the calculations. A lognormal distribution with $\sigma = 1.86$ is used to determine the aerosol extinction coefficients. The aerosol extinction coefficients are then integrated for the stratospheric layer to get the stratospheric aerosol optical depth. In the measurements made after the Pinatubo eruption we derived the altitude profiles of aerosol number densities (cm^{-3}) in the 5–35 km altitude region (Ramachandran et al., 1994a). The summed up (17–30 km) aerosol number densities during October 1991 and April 1992 over Hyderabad are found to be 195 ± 37 and 107 ± 19 particles, respectively. As the aerosol number densities in the stratosphere can increase by more than an order of magnitude after a major volcanic eruption, we used an aerosol number density of about 10 particles which would represent nonvolcanic conditions and determined the stratospheric aerosol optical depths. This assumption is also based on the aerosol extinctions obtained (see Fig. 7) later during volcanically quiescent and perturbed conditions. The aerosol extinction values are about an order of magnitude lower during volcanically quiescent conditions when compared to volcanically perturbed conditions. As the aerosol extinctions are directly proportional to the number densities, this is a valid assumption. However, the number densities during volcanically quiescent periods can exhibit variations on the order of ± 10 –20%, and hence the resulting aerosol extinctions will also be uncertain by the same amount. In the recent experiment the data obtained from the sun-scanning photometer assembly in the ± 20 – 90° range with respect to the sun were found to be of poor quality, and hence the aerosol size parameter and number densities could not be estimated.

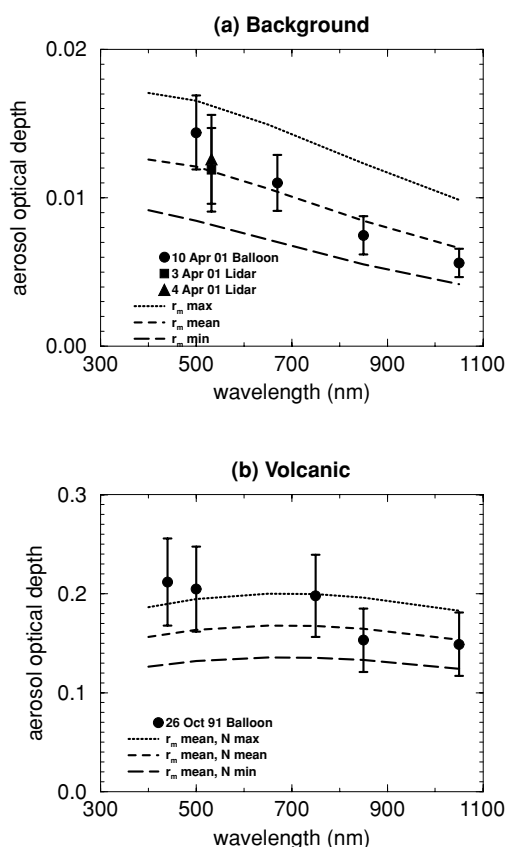


Fig. 6. (a) Stratospheric aerosol optical depth spectra calculated from the aerosol extinction coefficients obtained on 10 April 2001. The lidar-derived aerosol optical depths are also plotted. (b) Stratospheric aerosol optical depth spectra for the volcanically perturbed stratosphere after the Pinatubo eruption. The lines show the estimated optical depths obtained by using the derived mode radii (Fig. 5) for the stratosphere. See text for details.

The stratospheric aerosol optical depths determined for the maximum, mean and the minimum mode radii measured in the stratosphere are shown in Fig. 6a. We see that as the mode radius increases from 0.09 to 0.11 μm the aerosol optical depth at 500 nm can almost double from a value of 0.008 to 0.016. The aerosol optical depths measured at 500, 670, 850 and 1050 nm by the balloon-borne experiment in April 2001 are also shown. We see that the measured values are within the range of estimated aerosol optical depths, though they are more closer to the mean mode radii profile. Vertical bars in the aerosol optical depth represent $\pm 1\sigma$ from the mean, and this arises due to the uncertainties in

deriving the vertical profiles of aerosol extinction. The lidar-derived aerosol optical depths at 532 nm on 3 and 4 April 2001 are also plotted. The mean aerosol optical depths are close to the 0.10 μm mode radius profile. The vertical bar represents $\pm 1\sigma$ and arises from the three values of B_a used.

In Fig. 6b the aerosol optical depth spectra obtained when the stratosphere was volcanically perturbed after the Mt. Pinatubo eruption is plotted. These balloon measurements were made 4 months after the eruption and hence represent fully grown aerosol particles. The volcanically perturbed aerosol optical depths at all wavelengths are about an order of magnitude higher when compared to the nonvolcanic spectra in the stratosphere. The stratospheric aerosol optical depths are calculated for the mean mode radius of about 0.20 μm for the mean, minimum and maximum number densities obtained on 26 October 1991. We see that the aerosol optical depths calculated using r_m and measured aerosol number densities agree well with the measured optical depths.

Thomason et al. (1997) using the SAM and SAGE data compared the stratospheric aerosol optical depths during 1979 and 1989–1991. They found that in 1979 the minimum 1.02 μm zonal mean aerosol optical depth derived from SAGE data in the tropics (10°S – 10°N) was 1.1×10^{-3} ; in the northern subtropics (15°N – 25°N) the optical depth was 9.7×10^{-4} , while in the northern midlatitudes it was 9.6×10^{-4} . They estimated that depending on latitude the 1.02 μm stratospheric aerosol optical depth in 1989–1991 was 10–30% higher than that observed in 1979. The mean stratospheric aerosol optical depth at 1.05 μm derived from the present balloon measurement is about 0.005. This is about five times more than the average value in the tropics of about 0.001 measured in 1979.

In Fig. 7 the stratospheric (17–30 km integrated aerosol extinction at 525 nm) aerosol optical depth measured by SAGE II over the Indian subcontinent (5°S – 35°N , 60°E – 100°E) is plotted for the period 1985–2000. The decay of the aerosol optical depths following El Chichon, Nevado del Ruiz volcanic eruptions is clearly seen until mid-1991, when the aerosol optical depths are in the range 0.002–0.005. Immediately after the Pinatubo eruption in June 1991 the aerosol optical depths show a very large increase, and the optical depths are >0.06 . The data from June 1991 to December 1992 do not extend to 17 km because of the dense Pinatubo cloud. The optical depths during this period are integrated from the altitude range of 19–23 to 30 km. Nevertheless the aerosol optical depths are

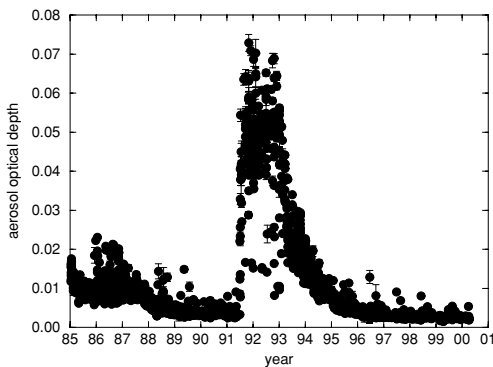


Fig. 7. SAGE II measured stratospheric aerosol optical depth (17–30 km integrated aerosol extinction coefficient) at 525 nm over the Indian region (5–35°N latitude and 60–100°E longitude) from 1985 to 2000.

very large when compared to the pre-eruption value. The time evolution of stratospheric aerosol optical depths show a decrease in 2000 when compared to 1990. The balloon-derived aerosol optical depths have shown an increase when compared to the SAGE value in 1979 and also with respect to the balloon-measured aerosol extinction coefficient in October 1985 (Jayaraman, 1991). During volcanically quiescent conditions we estimate a maximum uncertainty of 10% in the derived aerosol extinctions at lower altitudes, which increases to about 30% at the stratospheric altitude levels. These uncertainties will become less during volcanically perturbed conditions, as there are more aerosol particles, resulting in a higher extinction and hence a better signal-to-noise ratio. During nonvolcanic (background) conditions it is a challenging task to determine the aerosol extinction coefficients in the stratosphere, as there are only a few aerosol particles present. The aerosol scattering is much less, so that the total scattering is almost same as Rayleigh scattering. This might increase the errors in the estimation of aerosol extinction coefficients at these altitudes. When compared to the SAGE II value for the stratospheric aerosol optical depth of 0.007 ± 0.0005 at 525 nm during October 1985 the balloon-derived optical depth is less and has a value of 0.001 ± 0.0002 at 500 nm. In April 2001 the balloon-derived aerosol optical depth is 0.014 ± 0.0025 at 500 nm, while the SAGE II value during April 2000 at 525 nm is found to be 0.0021 ± 0.0004 . Differences between the aerosol characteristics measured in situ and SAGE II data in the stratosphere have been observed, especially under nonvolcanic conditions (Hervig and Deshler, 2002). While

we measured the aerosol extinctions directly, Hervig and Deshler (2002) derived aerosol extinction from size distributions, and it was found that size distribution errors propagate into extinction uncertainties on the order of 30–40%. We suspect that there was an underestimation of the aerosol extinction coefficients in 1985 while in 2001 there was an overestimation or, as stated earlier, the errors could be larger. Hence, for the stratospheric data we refrain from making any trend estimates.

Hervig and Deshler (2002) compared the University of Wyoming balloon-borne optical particle counter (OPC) data with the SAGE II and the Halogen Occultation Experiment (HALOE) data on aerosol extinctions for the period 1982–2000. They found that when the aerosol amount was low, SAGE II and HALOE extinctions were higher than OPC estimates. Also, variations in agreement over altitude and time seem to be related to the changes in aerosol loading (due to the appearance and decay of volcanic aerosols), with better agreement found under conditions of high aerosol loading. A very good agreement was seen post-Pinatubo between the balloon-borne aerosol extinctions obtained over Hyderabad during October 1991 and April 1992 and the SAGE II extinctions (Ramachandran et al., 1994a). The agreement in general was good among the three data sets at 16 and 22 km, with greater variability and poorer agreement at 28 km (Hervig and Deshler, 2002). During the background periods post-El Chichon and Pinatubo the SAGE II extinctions at 16 and 22 km were higher than the OPC values.

In section 4.6 we discuss several atmospheric processes such as the Inter Tropical Convergence Zone (ITCZ), troposphere–stratosphere exchange, the tropical reservoir and the influence of anthropogenic emissions that could have resulted in the increases in the upper tropospheric/stratospheric aerosol optical depths, particularly over the tropics in the past 20 yr.

4.5. Ångström wavelength exponent

With an attempt to gain further insight into the aerosol size variation with respect to altitude, a parameter α is defined such that

$$\tau = \beta \lambda^{-\alpha} \quad (6)$$

where τ is the aerosol optical depth and β is the Ångström coefficient. The vertical profile of wavelength exponent obtained in 2001 is plotted in Fig. 8. The horizontal bars indicate $\pm 1\sigma$ error in fitting the

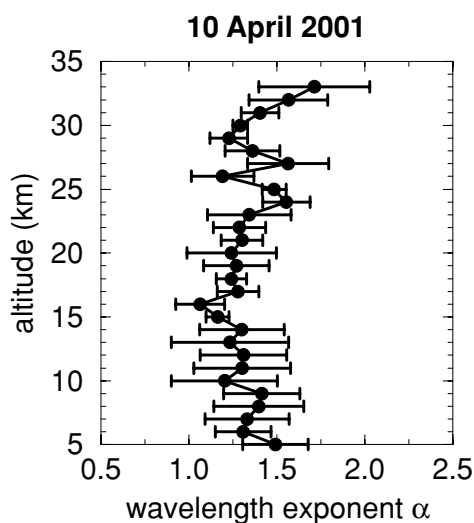


Fig. 8. Profile of wavelength exponent α derived from the aerosol extinction coefficient spectra at each altitude. Horizontal bars indicate $\pm 1\sigma$ error in fitting α .

exponent. The wavelength exponent is about 1.5 at 5 km, decreases with height, and shows a value of about 1.2 at 15 km. The mean exponent in the troposphere is 1.32 ± 0.09 . Above the tropopause, in the lower stratosphere the wavelength exponent is about 1.3 at 17 km, which then increases to about 1.7 at 33 km. The mean wavelength exponent in the 17–30 km altitude region is 1.33 ± 0.12 . While for Rayleigh scattering (scattering by air molecules) $\alpha = 4$, for aerosol scattering the wavelength exponent decreases with increasing particle size and vice versa.

The Ångström wavelength exponent α and the Junge aerosol size parameter ν , defined as the slope of the Junge power law curve size distribution, can be approximately related as $\alpha = \nu - 2$ (Bullrich, 1964). All though a lognormal size distribution may give a better description of the aerosol size distribution, for the optically effective particles in the size range $0.05\text{--}10\text{ }\mu\text{m}$ the Junge power law can be taken as a good representation in the upper troposphere and lower stratosphere (Volz and Sheehan, 1971; Bigg, 1976). As the Junge size parameter decreases, the number of larger particles increases compared to the number of smaller particles and vice versa. Therefore the exponent and Junge parameter profiles in principle should correlate with each other qualitatively, because as the relative size of the particle increases, the slope of the power law curve Junge parameter decreases, as does the ex-

ponent, indicating that aerosol scattering is less dependent on wavelength λ . A value of 3 is found typical for the Junge parameter which represents background aerosol particles.

The Junge parameter profile obtained during October 1991 (Ramachandran et al., 1994a) shows a marked decrease in the region between 17 and 23 km, with a value of around 1.8. This indicates the presence of larger aerosol particles formed 4 months after the Pinatubo eruption. The Junge parameter values in the lower stratosphere over Hyderabad in April 1984, about 2 yr after the El Chichon eruption were in the range 2.8–3.5 (Jayaraman and Subbaraya, 1988). In the troposphere (5–15 km) the Junge parameter was found to be about 3 in April 1984, 2.72 ± 0.20 in October 1991 and 2.45 ± 0.35 in April 1992. In the lower stratosphere the Junge parameter was 3.06 ± 0.24 in April 1984, 2.18 ± 0.22 in October 1991 and 1.99 ± 0.21 in April 1992. Junge parameter values in the range of about 2 indicate the dominance of larger particles in the size distribution when compared to the smaller-sized aerosols. The wavelength exponent values in the present experiment in the entire altitude region of 5–33 km are in the range of 1.1 (minimum) and 1.7 (maximum). Junge parameter values obtained from the 22 October 1985 balloon experiment conducted during a volcanically quiescent period are 3.06 ± 0.05 in the 5–15 km altitude region and it was about 3.05 ± 0.16 in the 17–30 km altitude region (Jayaraman and Subbaraya, 1988). In the troposphere the Junge parameter values exhibit much smaller variations varying from 3 (minimum) to 3.1 (maximum). In the lower stratosphere the Junge parameter is found to vary from a low of 2.9 to 3.4 in the altitude region of 17 to 30 km. A wavelength exponent value of 1.5 translates into a Junge parameter of about 3.5, representing smaller aerosols and hence background aerosol particles. The exponent values obtained in 2001 exhibit a close correspondence with the 1985 Junge parameter values, indicating that the aerosol size distributions between 1985 and 2001 have not changed and substantiating the results obtained on mode radii.

4.6. Possible reasons for the increase in upper tropospheric/stratospheric aerosols over tropical India

In the upper troposphere the aerosols exhibit quite significant geographical-scale characteristics and are seasonally dependent. For example, it has been found in the northern hemisphere that the maximum lifting

of material occurs in the spring (Kent et al., 1995). The low-latitude aerosol enhancements seem to have the characteristics of aerosols derived from arid surface regions, while the higher-latitude aerosol appears more likely due to anthropogenic sources. There exist certain major uncertainties concerning the stratospheric aerosol distribution during nonvolcanic conditions. One question is whether the background (i.e. nonvolcanic) aerosol can be usefully defined through natural and anthropogenic sulfur emissions (Schwartz et al., 1995). The SO₂ emitted from fossil fuel combustion and biomass burning are the major sources of sulfate aerosols in the troposphere.

The deep-convective cirrus cloud systems of the ITCZ contribute significantly to the vertical exchange of aerosols and trace gases between the surface and the upper troposphere, possibly including the lower stratosphere (Ramanathan et al., 2001). Another important aspect that needs mention is the aerosol–cloud interaction and its effects. The aerosol transport in the horizontal direction is controlled by winds and is a mesoscale process. The vertical transport of aerosols and their scavenging is determined by convective motions and so has convective timescales. The aerosol particles trapped in the convective updrafts serve as cloud condensation nuclei (CCN) and help in cloud droplet formation (Jayaraman, 2001). The efficiency of an aerosol particle to serve as CCN is dependent on its size and the amount of water adsorbing material in it. It has been observed that an increase in amount of aerosol can suppress precipitation at least from low clouds. It should be noted that the probability of cloud-processed aerosol existing in the free troposphere is high.

Transport of mass between the stratosphere and troposphere is a key process in atmospheric physics and chemistry. The vertical mixing timescale is the parameter that distinguishes the troposphere from the stratosphere; vertical transport of air and chemical species throughout the troposphere can occur on timescales of a few hours via strong updrafts associated with large cumulus formation (like in the vicinity of ITCZ), whereas vertical transport over a similar altitude range in the stratosphere can take months to years or even more (Seinfeld and Pandis, 1998). Tropics are the location in which the largest net upward transport into the stratosphere occurs which directly can influence the composition of the global middle atmosphere. Hadley cell circulation injects significant amounts of the tropical tropospheric air through its upward-flowing branch into the stratosphere (Asnani, 1993).

While analysing the global aerosol SAGE I and II data Trepte et al. (1994) found zonal band structures in stratospheric aerosol optical depths. The extinction distributions are found to be compatible with Brewer–Dobson circulation, which has a rising motion in the tropics, subsidence at high latitudes and a net poleward flow in mid-latitudes (Trepte et al., 1994). This upward transfer from the troposphere into the stratosphere occurs predominantly in the tropics (Andrews et al., 1987).

4.6.1. Anthropogenic emissions: India. Charlson et al. (1992) showed that anthropogenic sulfate aerosols play a crucial role in regional and global climate change. It is now known that sulfate aerosols and organic matter will cool the Earth's atmosphere while black carbon (soot) aerosols can warm the atmosphere. In contrast to greenhouse gases, aerosols have short atmospheric residence times of the order of a week and would be concentrated in the source regions and hence will exhibit strong spatial and temporal variations in the resulting climatic effects. It has been found that approximately 35% of the particles entering the troposphere is airborne sulfate from oxidation of SO₂ emissions (Wolf and Hidy, 1997). Fossil fuel combustion, particularly coal and biomass burning dominated the worldwide emissions. The emissions were projected to grow by about two times in 2040 largely from fossil fuel combustion (Wolf and Hidy, 1997). This growth was expected to be greatest in the developing countries, especially India and China.

India is one of the fastest growing economies in the Asian continent. It ranked sixth in the world in total consumption of commercial energy during 1999 (CMIE, 2001). Very recently, Reddy and Venkataraman (2002a,b) came out with emission inventories for aerosols and sulfur dioxide emissions over the Indian continent on a spatially resolved (0.25° × 0.25°) grid. They estimated the aerosol (PM_{2.5}, black carbon and organic matter) and sulfur dioxide emissions for fossil fuel and biomass combustion.

Coal, petrol and diesel oil are the major fossil fuels used in India for electric power generation and for road and rail transportation, in addition to furnace (fuel) oil and kerosene (Reddy and Venkataraman, 2002a). These fuels are used in a variety of industries and processes such as electrical power generation, iron and steel plants, coke ovens, fertilisers, refineries and petrochemicals, pulp and paper making, mining, brick making, the domestic sector and for transportation. In addition these fuels are used, for example, in the textile, chemical, glass and ceramics, food and

plantation industries. SO_2 emission factors (g kg^{-1}) (emfacs) for fuel combustion are dependent on the fuel sulfur content, combustion technology and pollution control equipment employed. As the emission factors are dependent on the factors mentioned above and also on the processes, they exhibit a wide range. For example, for a sulfur content of 0.45 (wt %) the emfac for SO_2 is 8.78 g kg^{-1} . The emfacs of SO_2 for the iron and steel industry is 8.84, and for brick kilns, domestic usage and railways it is 7.08. The emfacs for petrol and diesel [high speed diesel oil (HSDO)] are on an average about 4 and 15 g kg^{-1} , respectively. The low-speed diesel oil (LSDO), fuel oil (FO) and kerosene have emfacs of 36, 80 and 4, respectively, while for natural gas it is 0.01.

Particulate matter (PM) of sizes ranging from sub-micron to several orders of microns are emitted during fossil fuel and biomass combustion. However, as larger particles settle within a short period of time, only particles $<2.5 \mu\text{m}$ with typical lifetime of about a week would travel several hundred kilometres and produce climatic effects (Krishnamurti et al., 1998; Prospero, 1979). The PM emissions are made up of a carbonaceous fraction, inorganic mineral ash and trace elements. The carbonaceous aerosols are further divided into organic matter (OM) and black carbon (BC). PM emissions for example depend on the fuel characteristics and combustion technology, such as coal ash content, boiler type, vehicle type etc., while the carbonaceous aerosol fraction depends on the combustion technology/characteristics. The emission factors for $\text{PM}_{2.5}$, OM and BC also exhibit a wide range.

From the CMIE report (2001) we estimated emissions in Tg yr^{-1} ($10^{12} \text{ g per year}$) of SO_2 , $\text{PM}_{2.5}$, OM and BC over India for a 20-yr period beginning from 1980–81 to 1999–00. We used offtake of coal as a proxy to coal consumption; for petrol (motor gasoline) we used statewide consumption given for the 19 major states of India. Information for the smaller states of Sikkim, Meghalaya, Arunachal Pradesh, Nagaland, Manipur, Mizoram and Tripura are not available and hence not used in this estimate. For diesel oil (HSDO and LSDO), FO and kerosene we took for all India the consumption given under various categories, which include transport, road transport, railways, food/plantation, power utilities, industry, mining and quarrying, iron and steel, textile, engineering and other industries. The consumer-wise consumption of all the fuels are given in tonnes. From the range of emfacs available for SO_2 , $\text{PM}_{2.5}$, OM and BC (Reddy

Table 1. *Emission factors for sulfur dioxide, $\text{PM}_{2.5}$, organic matter and black carbon in g kg^{-1} used in the present study (Reddy and Venkataraman, 2002a)*

Fuel	SO_2	$\text{PM}_{2.5}$	OM	BC
Coal	8.00	8.10	0.33	0.077
High speed diesel oil	15.00	3.38	0.28	0.29
Low speed diesel oil	36.00	3.38	0.28	0.29
Petrol (motor gasoline)	4.00	3.59	0.65	0.07
Fuel (furnace) oil	80.00	2.47	0.29	0.06
Kerosene	4.00	3.33	0.29	0.29

and Venkataraman, 2002a) we calculated the mean emission factors which are given in Table 1 and used in this study.

The emfacs of SO_2 for the fuels show an order of magnitude variation from 4 for petrol and kerosene to 80 for fuel oil (Table 1). The $\text{PM}_{2.5}$ emfacs vary from 2.47 to 8.10 for these fuels. The variation in OM emfacs is less, where the factors vary from a minimum of 0.28 to a maximum of 0.65. For black carbon the emfacs show a wide range of variability from 0.06 for fuel oil to 0.29 for HSDO, LSDO and kerosene. It should be noted that the emfacs of SO_2 , $\text{PM}_{2.5}$, OM and BC for coal that is used in power generation are for a 50% control scenario. It has been estimated that India's power sector is primarily growing on coal, with its total coal consumption almost doubling between 1989–90 and 1997–98 (Reddy and Venkataraman, 2002a). This increase in coal consumption would therefore result in a proportionate increase in SO_2 emissions.

Figure 9 shows the estimated emissions in Tg yr^{-1} of sulfur dioxide and black carbon for the different fuels and their total. The emissions are projected from aggregate national energy use assuming constant emission factors (Table 1) for the 20-yr period. In general coal and HSDO dominate the emissions. In the figure for SO_2 emissions the furnace oil curve also contributes more or less the same as HSDO, though in the initial periods of the 1980s the furnace oil emissions are higher than HSDO emissions. The total SO_2 emissions add up to $\sim 6 \text{ Tg yr}^{-1}$ in 1999–2000. The total $\text{PM}_{2.5}$ is about half of the SO_2 emissions in the 20-yr period (not shown). The emissions from OM (not shown) and BC are about one–two orders of magnitude less than SO_2 emissions. The percentage increase in the SO_2 , $\text{PM}_{2.5}$, OM and BC emissions from coal is about $10\% \text{ yr}^{-1}$ in the 20-yr period. The maximum percentage increases have occurred in petrol (14%)

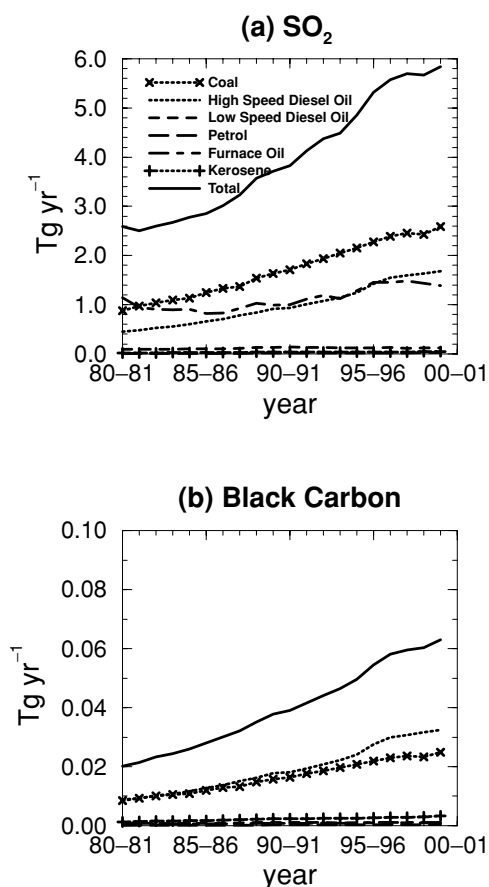


Fig. 9. SO_2 and black carbon emissions in Tg yr^{-1} for India over a 20-yr period from 1980–81 to 1999–2000 for the fossil fuels coal, petrol, high speed diesel oil, low speed diesel oil, furnace oil and kerosene, and their total.

followed by HSDO (13%). Emissions due to kerosene have increased by about $8\% \text{ yr}^{-1}$. The percentage increases in LSDO and furnace oil emissions over the 20-yr period are the lowest at around 1%. It is quite clear that over the 20-yr period the percentage increases in emissions due to coal, petrol, HSDO and kerosene are in league with the increase in population, industries, automobiles etc. in India. The percentage increases for different fuels are the same for SO_2 , $\text{PM}_{2.5}$, OM and BC emissions because we are essentially looking at the increase in the consumption of the fuels over the 20-yr period. However the percentage contribution due to these fuels to the total emissions are different as it is dependent on the consumption during each year and on emission factors. In SO_2 emissions coal on an

average contributes about 42% to the total, furnace oil and HSDO contribute about 29 and 24%, respectively. In $\text{PM}_{2.5}$ emissions, coal on average contributes about 85%, followed by HSDO ($\sim 11\%$) and furnace oil and kerosene contributing about 1.5% each. Coal contributes about 72% and HSDO contributes about 18% in the OM emissions, the other fuels contributing the rest. In BC emissions, the HSDO contribution dominates at around 47%, while the contribution by coal is not far behind at 42% and kerosene contributes about 6% to the total.

In the year 1996–97 biofuels accounted for 93% of the total biomass consumption, with forest fires contributing only 7% (Reddy and Venkataraman, 2002b). The national average biofuels mix was found to be 56:21:23% of fuelwood, crop waste and dung-cake, respectively. In India, biomass combustion mainly arises from fuel used for domestic cooking, forest fires and burning of crop waste and harvest. Wood, crop waste and dung-cake are the biofuels used in rural India, while in urban areas wood is reportedly the biofuel used. Reddy and Venkataraman (2002b) estimated that for 1996–97 the biomass combustion in India resulted in about 2 Tg yr^{-1} of $\text{PM}_{2.5}$ emissions, which was equal to the emissions from fossil fuel combustion.

Population of India has been steadily increasing at an average rate of about 23% per decade over the past two decades. The rural and urban mix on average is about 74 and 26%, respectively. Currently the Indian population stands at an excess of 1 billion. Though we have not estimated the SO_2 and aerosol emissions from biofuels for India, it is quite clear that all the biofuels will also contribute to the emissions significantly in addition to the fossil fuel emissions, and these emissions would have only increased in the 20-yr period with the increase in the population (out of which 74% is a rural population which predominantly uses biofuels for cooking). The increases in emissions from various fossil and biofuels are important, as it is found that non-sulfate aerosol particles are present in the lower troposphere which can then be transported and participate in the troposphere–stratosphere exchange. As the non-sulfate particles are also likely to affect the upper troposphere and the lower stratospheric AODs, it is worth mentioning that 50% PM emission is from biomass burning; this should be addressed in future emission trends analysis.

It is clear that if we take into account the dynamic atmospheric phenomena such as ITCZ, which is active over the Indian subcontinent during summer, the troposphere–stratosphere exchange which is very

active over the tropics, and the increase in anthropogenic emissions due to fossil fuel and biomass combustion over India, it is possible to explain the observed increase in the upper tropospheric and the background stratospheric aerosols over the past two decades. It is quite possible that the upper tropospheric aerosol increase seen over India is region-specific and happened because of the combined effect of both the natural and anthropogenic phenomena. In addition to the region-specific features over the tropics there are other processes that could be responsible for the upper tropospheric/stratospheric aerosol increase seen in the last 2-decades.

4.6.2. Other possible causes. There have been many investigations to determine the sulfur flux estimates required to sustain the stratospheric background aerosols. The sulfur flux estimates have ranged from a minimum of 0.043 Tg yr^{-1} (Crutzen, 1976) to a maximum of 0.17 Tg yr^{-1} (Hofmann et al., 1976). Hofmann (1991) estimated that a sulfur source of 0.063 Tg yr^{-1} is required to sustain the background sulfate aerosol levels. In the present work it is shown (Fig. 9a) that the SO_2 emissions are about 2.6 Tg yr^{-1} in 1980–81, which increases to about 6 Tg yr^{-1} in 1999–2000. The steady increase in SO_2 emissions over the 20-yr period can possibly explain the increase in the upper tropospheric aerosol optical depths over India. Even if we assume that 10% of the SO_2 emissions can reach the stratosphere, it is more than enough to sustain the background sulfate aerosol layer.

As carbonyl sulfide (OCS) is believed to be one of the sources for the stratospheric background aerosols, it was suggested that OCS could be responsible for the enhanced aerosol loading, since biogenic emission of OCS may increase in response to global warming (Turco et al., 1980). Chin and Davis (1995) ruled out the possibility that OCS could be one of the potential sources for this increase, as they found that the sources and concentrations of OCS have remained constant in the atmosphere in the 1980s and 1990s. Hofmann (1991) also arrived at a similar conclusion. A global three-dimensional climate model calculation shows that the contribution from OCS to stratospheric sulfate formation is only about 10%; instead the sulfate formation seems to be dominated by oxidation of SO_2 injected from the troposphere (Kjellström, 1998).

Hofmann (1991) estimated that the jet fuel consumption increased at a rate of $5\% \text{ yr}^{-1}$ during the 1980s, similar to the background sulfate levels. He estimated that about 65% of the required source strength of sulfur comes from the jet fleet and hence is a significant

source of stratospheric sulfate. One sixth of the NH air traffic takes place directly in the stratosphere. Hofmann estimated that worldwide airline traffic is expected to double by year 2005 in a doubling time of 15 yr.

Thomason et al. (1997) listed many possible causes for the increase in the stratospheric aerosol optical depths during 1989–91 when compared to 1979. This enhancement seemed to be a characteristic of an extended recovery from the volcanic perturbations associated with El Chichon and Nevado del Ruiz. They concluded that the slow decrease of aerosol loading in 1989 and 1990 is likely due to a combination of several processes, such as continuing loss of aerosol from the stratosphere following these volcanic eruptions, variations in the transport of aerosol from the tropical reservoir associated with the phase of quasi-biennial oscillation (QBO), and possibly due to the influence of small volcanic events in 1990. It should be noted that in this time period (between 1979 and 2001) there were two major tropical volcanic eruptions, viz. El Chichon (17.3°N) in April 1982, Mt. Pinatubo (15.4°N) in June 1991, and about five or six less major eruptions, viz. starting from Sierra Negra (0.8°S) in November 1979, Mt. St. Helens (46.2°N) in May 1980, Nevado del Ruiz (4.9°N) in November 1985, Kelut (8°S) in February 1990 and Mount Cerro Hudson (45°S) in August 1991. Bekki and Pyle (1992) suggested that industrial emissions of SO_2 could be one of the sources for the increase in the background stratospheric levels.

5. Conclusions

A high-altitude balloon carrying sun-scanning/tracking multichannel photometer systems was launched from Hyderabad (17.5°N , 78.6°E) on 10 April 2001 to study the aerosol characteristics in the upper troposphere and stratosphere. By estimating the attenuation of the incoming solar radiation at each altitude and after correcting for molecular scattering and gaseous absorption, aerosol extinctions are obtained. The aerosol extinction coefficient is about 10^{-2} km^{-1} in the upper troposphere and is in the range 10^{-3} – 10^{-4} km^{-1} in the stratosphere. The aerosol extinction coefficient values are about two orders of magnitude less compared to those obtained after the Pinatubo eruption over the same location. The aerosol extinctions derived from independent lidar measurements conducted on 3 and 4 April 2001 over

Mt. Abu (24.6°N) are found to be in good agreement with the balloon results in the stratosphere.

The integrated aerosol extinction in the upper troposphere (5–15 km) at 500 nm wavelength shows an average increase of about $11 \pm 1\%$ yr^{-1} over Hyderabad during the period 1985–2001. This increase is consistent with the increase in columnar aerosol optical depths increase from long-term data available over a few stations in India. The mean mode radius of the aerosols in the stratosphere is found to be $0.10 \mu\text{m}$, which represents the background (nonvolcanic) aerosol particles, and is comparable to the value found during the October 1985 balloon experiment over the same site. Similar values of mode radii indicate that between 1985 and 2001 the sizes of aerosols in the stratospheric altitudes have not changed over the measurement site. The background stratospheric aerosol optical depth at $1.05 \mu\text{m}$ is found to be 0.005; this is about five times larger than the SAGE-derived aerosol optical depth value of 0.001 for the tropics in 1979. We derive the background and volcanic stratospheric aerosol optical depth spectra from the mode radii and, using the aerosol number densities, compare them with the measured optical depths. A good agreement is seen between the estimated and the balloon, lidar-measured aerosol optical depths during both volcanic and quiescent conditions. The mean Ångström wavelength exponent in the stratosphere is derived as 1.33 ± 0.12 . The close correspondence between the wavelength exponent values obtained in 2001 and the Junge size parameter values obtained in 1985 indicates that the stratospheric aerosol size distribution has not changed in these years, corroborating the results obtained on mode radii.

We have discussed various atmospheric dynamical processes that are important in the tropics, in particular and the influence of the anthropogenic emissions that could have resulted in the differences in the upper tropospheric/stratospheric optical depths observed between 1985 and 2001. The SO_2 , $\text{PM}_{2.5}$, organic matter

and black carbon emissions for the 20-yr period starting from 1980 over India are estimated. The variations in emissions of these aerosol species due to six fossil fuels, namely, coal, petrol, high-speed and low-speed diesel oils, furnace oil and kerosene, are calculated. The sulfur dioxide and aerosol emissions from coal have increased by about 10% yr^{-1} over India in the 20-yr period. The emissions from high speed diesel oil and petrol are higher and are in the range of $13\text{--}14\%$ yr^{-1} over India. The emissions from kerosene have gone up by about 8% yr^{-1} over the 20-yr period from 1980–81 to 1999–2000. These increases in emissions over the 20-yr period in India could possibly explain the increase in upper tropospheric aerosol optical depth. Estimates possibly indicate that even if about 10% of SO_2 emissions from the surface reach the stratosphere, due to cumulus convection in the vicinity of ITCZ and due to troposphere–stratosphere exchange, that is enough to sustain the background stratospheric aerosols over tropical India.

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