INFLUENCE OF CHANGES IN THE PREVAILING SYNOPTIC CONDITIONS ON THE RESPONSE OF AEROSOL CHARACTERISTICS TO LAND- AND SEA-BREEZE CIRCULATIONS AT A COASTAL STATION

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Abstract. The changes in the response of near surface aerosol properties to land- and sea-breeze circulations, associated with the changes in the prevailing synoptic meteorological conditions, are examined for a tropical coastal station. Aerosol properties are nearly similar in both the breeze regimes (land and sea) during seasons of marine airmass while they are distinct during seasons of continental airmass. As the prevailing winds shift from continental to marine and the ambient weather changes from winter conditions to the humid monsoon season, the submicron mode, which dominated the aerosol mass-size distribution, is largely suppressed and the dominance of the super micron mode increases. During periods of continental air mass (winter), the aerosol loading is significantly higher in the land-breeze regime, (particularly in the submicron range) but as the winds shift to marine, the loading initially becomes insensitive to the breeze regimes and later becomes higher in the sea-breeze regime, particularly in the super micron range.

Keywords: Aerosol sampling, Aerosol size distribution, Aerosols and airmass types, Coastal aerosols, Land/sea breeze, Quartz Crystal Microbalance (QCM) Impactor.

List of Symbols

D_{pi}	Lower cut off particle diameter of the ith stage
D_{ai}	Lower cut off aerodynamic diameter of the ith stage
ρ	Particle density
ρ_0	Reference density
m_{ci}	mass concentration of aerosols in the ith stage
d_{pi}	geometric mean diameter of the ith stage = $\sqrt{D_{p(i-1)}D_{pi}}$
r_i	geometric mean radius of each size bin
Δr_i	radius-width of the ith size bin (μm) = $\frac{1}{2}(D_{p(i-1)} - D_{pi})$
$m(r)$, $\frac{dm}{dr}$	mass size distribution
M_t	Total mass concentration
M_a	Mass concentration in accumulation size range
M_c	Mass concentration in coarse size range

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v_{ci} Volume concentration of aerosols in the ith stage
a_{ci} Area concentration of aerosols in the ith stage
R_{eff} Effective radius of aerosols
R_m Mass mean radius

1. Introduction

In recent years, there has been increased recognition of the role of aerosols in the radiation budget of the earth-atmosphere system, climate and the environment (Boucher and Anderson, 1995; Satheesh and Ramanathan, 2001; Ramanathan et al., 2001; Podgorny and Ramanathan, 2001). The direct and indirect effects of atmospheric aerosols on radiative forcing and cloud physics are strongly dependent on particle size characteristics and chemical composition (Hegg et al., 1996; Haywood et al., 1997; Muller et al., 1999). Determination of the mass-size distribution is thus a fundamental and important means of characterising atmospheric aerosols (Koutsenogii et al., 1994; Nesub et al., 2000). Information on the aerosol size distribution is of importance also from the perspectives of air chemistry (origin, transport and removal of particles: Meszaros, 1968); radiative interactions (Charlson et al., 1987; Noone et al., 1992; Kaufmann, 1997); cloud microphysics and albedo (droplet number concentration and size distribution) and consequently the indirect aerosol forcing of climate (Wiedensohler et al., 1997). Size distribution of atmospheric aerosols strongly depends on the sources and sinks as well as on the meteorological processes acting during their lifetime (Choularton et al., 1982; Vakeva et al., 2001), on the type and history of the airmass, as well as on the boundary-layer circulations (like land and sea breezes that occur daily in coastal areas) (Suzuki and Tsunogai, 1988; Moorthy et al., 1991, 1993; Smirnov et al., 1994; Vignati et al., 1997). Near-surface aerosols (which are also called boundary-layer /mixing region aerosols) show larger variations on shorter scales of time and space owing to their greater dependence on source and sink processes, as well as due to the influence of both mesoscale and synoptic atmospheric processes such as land- and sea-breeze circulations, changes in prevailing winds and rainfall. Nevertheless, they contribute significantly to the total aerosol loading at any given location. In this paper we examine the response of near-surface aerosols (size distribution and mass concentration) to land- and sea-breeze activity, and the changes associated with seasonal changes in the prevailing air mass type as observed from a tropical coastal station.

2. The Aerosol Data

The base data on aerosol characteristics used in this study are the near-real-time measurements of total mass concentration (M_t) and mass size distribution ($m_c(r)$), made (at about 3 m above ground level) using a 10-stage Quartz Crystal Microbalance (QCM) Impactor (model PC-2 of California Measurements Inc.). The QCM operates at a flow rate of 240 ml min^{-1} and segregates particles into ten size bins with their 50% lower cut-off particle diameters (D_{pi}) at 25, 12.5, 6.4, 3.2, 1.6, 0.8, 0.4, 0.2, 0.1 and 0.05 μm. The typical sampling time used is about 3 to 5 min and the samples are collected at hourly intervals. The instrument was operated regularly at Thumba, Trivandrum (8.5° N, 77° E, 3 m msl), a remote coastal location situated near the south-west tip of the Indian Peninsula. The experimental site (situated within the Thumba Equatorial Rocket Launching Station) is in fairly flat coastal terrain, nearly 500 m due east of the Arabian Sea and more than 10 km to the north-west of the city of Trivandrum. More details on the sampling site are given elsewhere (Pillai and Moorthy, 2001).

The lower cut-off particle diameter (D_{pi}) for each stage of the QCM is estimated from the corresponding aerodynamic diameter (D_{ai}) (which is defined as the diameter of a hypothetical sphere with a reference density of 1000 kg m^{-3} that has the same settling speed as the actual particle with a density ρ) and using a value of 2000 kg m^{-3} for the particle density (ρ) in the relation

$$D_{pi} = \frac{D_{ai}}{\sqrt{\rho/\rho_0)}}, \tag{1}$$

where $\rho_0 = 1000$ kg m^{-3} (by definition).

Due to coastal proximity, the sampling site experiences land/sea-breeze circulations, almost daily, and the QCM measurements are made at regular intervals so as to cover both the breeze regimes adequately. In all, 1430 samples were made on 131 days spread nearly evenly over the period October 1998 to December 2000. Supplementary data on the wind speed and direction are obtained from anemometer measurements made at the same site (Prakash et al., 1992) and when these are not available, the data from the nearby airport meteorological station are used. The characteristics of the prevailing wind (synoptic) are obtained from monthly mean values of wind speed and direction at the 850 h Pa level, deduced from the daily radiosonde ascents from the meteorological station located in the city of Trivandrum. These are used to classify the airmass types.

3. Regional Meteorological Features

The major meteorological features of the region fall under two categories. One is the mesoscale land/sea-breeze circulation occurring almost daily and confined

within the boundary layer (<1 km) (Narayanan et al., 1967). The sea breeze (SB; onshore flow) prevails mostly during the daytime and the land breeze (LB; offshore flow) during the nighttime and early morning. The change over from LB to SB occurs generally between 0800 and 1000 local time and is accompanied by changes in air temperature and relative humidity (RH). The sea breeze is most intense in the afternoon and weakens towards evening before giving way to the land breeze between 1800 and 2100 local time (Narayanan et al., 1967; Prakash et al., 1992). Besides these, the station also experiences changes in weather and airmass type due to synoptic scale changes in the prevailing winds and rainfall associated with the Asian monsoon (Das, 1986). A summary of the average prevailing conditions is shown in Figure 1. The winds at the 850 hPa level (shown in panels 1 and 2 at the top) are basically low speed ($\approx$3 m s^{-1}) north-easterlies and directed from the land constituting a continental (C) airmass during the months of November to March. During this period the total rainfall (panel 3) is low and decreases progressively. Generally, dry land conditions prevail with relatively large diurnal variation (10 to 14 °C) in air temperature (panel 4 from top) and a comparatively low (60 to 70%) RH (bottom panel). Being a tropical station, the temperatures are rather high throughout the year; the monthly mean maximum temperature never goes below 30 °C. The prevailing winds start shifting from April and get established as an onshore flow by May and continue so till September constituting a marine (M) airmass, before reversing again in October. During this period of M airmass, the monthly mean wind speeds are higher, reaching peak values of 12 to 15 m s^{-1} in June–July months. The rainfall increases in intensity and becomes widespread and extensive. [However, 2000 was a comparatively rain deficient year for Trivandrum, with the annual rainfall being only $\sim$60% of the long-term average, the deficit occurring mainly during the south-west monsoon months.] Associated with this change in the prevailing winds, the mean air temperature increases initially before decreasing in the monsoon periods; the diurnal variation amplitude decreases ($\approx$9 °C) and the ambient RH stays at higher values ($\approx$70–80%). These changes in synoptic wind also influence the land/sea-breeze activity (Narayanan et al., 1967). As such, it is quite distinct during the months of November to March while during the months of June to September the sea breeze is superposed on the prevailing wind and at times is even masked. As both the land/sea-breeze activity as well as the change in airmass types result in advection of distinct aerosols (from the landmass or ocean) the aerosol properties would show associated changes.

4. Analysis of the QCM Data

For our studies, the QCM data have been considered separately for the Land-Breeze Regime (LBR) and Sea-Breeze Regime (SBR) by segregating the data in terms of surface wind directions and grouping the data collected during periods of onshore flow to SBR and offshore flow to LBR. Referring to the local coastline geometry,

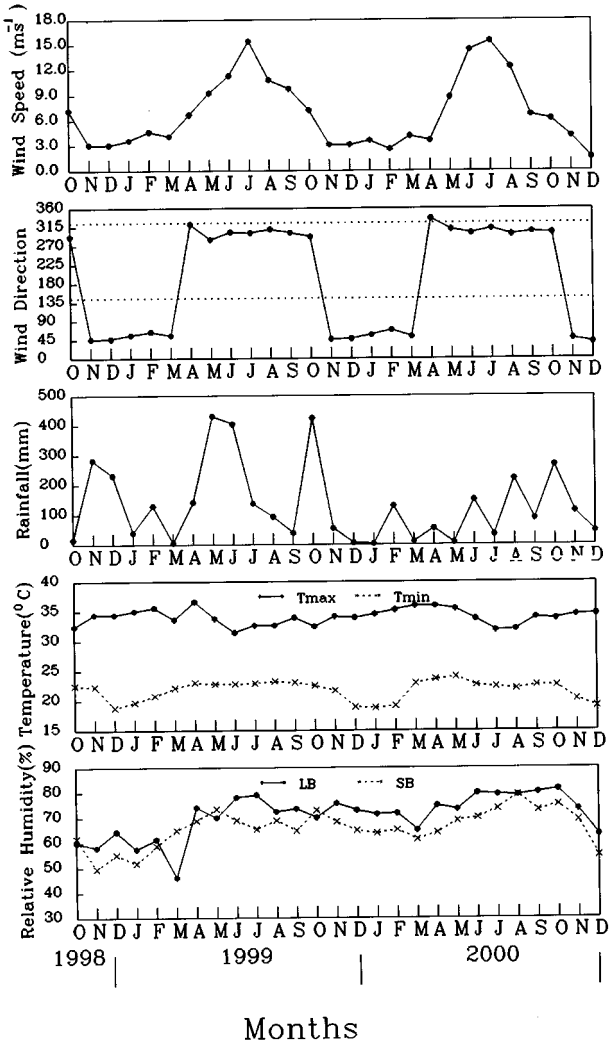

Figure 1. Variation of the prevailing wind field over Trivandrum at 850 h Pa level (panels 1 and 2 from top), total rainfall (panel 3) during the study period, maximum and minimum temperatures (panel 4) and RH (panel 5) during observation period. Wind directions (in panel 2) lying between the dashed lines signify marine airmass.

breezes arriving at angles between 145° and 325° (measured clockwise) would have a marine component and are considered to constitute the SB while breezes arriving from other directions do not have any marine component and hence are taken as land breezes. Data collected during periods of breeze transitions (when the wind direction was shifting between the two regimes, prior to the reversal) were not considered.

The QCM provides the total mass concentration M_t and mass concentration m_{ci} for the ith size bin as a function of size (channel no) such that

$$M_t = \sum_{i=1}^{10} m_{ci}. \tag{2}$$

The cumulative mass size distribution $m(r)$ is deduced by dividing the mass concentration at each size bin by the width Δr_i of that bin so that

$$\frac{dm}{dr} = \frac{(m_{ci})}{\Delta r_i}, \tag{3}$$

where $\Delta r_i = \frac{1}{2}(D_{p(i-1)} - D_{pi})$.

While estimating mass size distribution, the 2 to 10 stages of the QCM only are considered, as the upper cut-off of the first stage is not well defined. Both M_t and $m(r)$ are segregated in terms of breeze regime and are averaged over each month to obtain the mean values representative for that month. (The number of samples considered per month lay between 30 to 75 in the LBR and 30 to 90 in the SBR; offering a statistically significant sample size).

5. Error Budget

As the physical mass collected by the QCM during any sampling is very small and the mass concentration in each size bin is proportional to the shift in the beat frequency between the sensing and reference oscillators of that stage (details are given in Pillai and Moorthy, 2001) the accuracy in the estimated mass depends on the frequency shift itself, such that stages registering higher shift (collecting more mass) have a better signal to noise ratio. The overall uncertainty in m_{ci} in each measurement varies between 5% to 20%, with higher errors for lower m_{ci}. In absolute terms the errors are typically 1 μg m^{-3} for stages where $m_{ci} \leq 10$ μg m^{-3} (Pillai and Moorthy, 2001).

6. Results and Discussion

First we examine the response of $m(r)$ and M_t to the breeze regimes. In Figure 2 the month-to-month changes in the mean mass size distribution are shown separately for the two breeze regimes; the solid lines indicating the LBR and the dashed line indicating the SBR. These are obtained by averaging the individual sample mass size distributions, obtained over identical months, but separated in accordance with the breeze regimes. The vertical bars are the standard errors ($= \sigma/\sqrt{(n-1)}$); σ, being the corresponding standard deviations. There are three main features noticeable in Figure 2: (i) The aerosol mass concentration under LBR is generally

higher than the corresponding values in the SBR during the period October to March when the continental airmass prevails (Figure 1). (ii) This difference is well marked in the submicron size range ($d_I < 1\ \mu$m); but is less significant at larger sizes ($d_I > 1\ \mu$m) where the values are more or less comparable in both the breeze regimes (except in November). (iii) The above patterns start changing from April. During the period from May to September when a marine airmass prevails (Figure 1) the mass concentration in the SBR is generally comparable to and in some months (e.g., August and September) even higher than the values in the LBR in most of the size bins, particularly in the super micron regime. Along with this, the peak in the aerosol mass concentration in the submicron regime starts decreasing significantly and the super micron mode becomes conspicuous, thereby clearly indicating a change in the mass size distribution with a shift in the airmass type.

6.1. AEROSOL MASS CONCENTRATION

The change in the response of the total mass concentration, M_t, is examined in Figure 3, where the ratio of the monthly mean value of M_t in LBR to M_t in SBR is plotted as a function of the months. Ordinate values greater than 1 indicate higher mass concentrations in the land breeze regime. It can be noticed that the ratio is high (>1) from October to April (being very high in November and March about 1.7 and 1.9 respectively) when the synoptic airmass is continental in nature. It decreases from May onwards and reaches its minimum of about 0.6 in August which means the mass concentration in SBR is now higher than the mean M_t in the LBR by a factor of ≈1.6. As seen from Figure 1 this period corresponds to the marine airmass conditions, when the synoptic flow is from the ocean. Consequently, the mass size distribution changes with the airmass types as well as with the breeze regime as seen in Figure 2. The marine aerosols have a more significant supermicron (sea salt) component, while the continental aerosols have a large amount of fine/accumulation aerosols with diameter <1 μm. However, the accumulation aerosols are of greater concern owing to their potential role in environmental and health impacts (Berg et al., 2000). Moreover, due to their longer residence times (Jaenicke, 1984) accumulation aerosols are also prone to long range transport. On the other hand the coarse aerosols (which are mostly mechanically generated), are rather short lived and hence are more of local importance. As such, we have estimated the mass concentrations separately for the accumulation ($d \leq 1\ \mu$m) and coarse ($d \geq 1\ \mu$m) aerosols from the QCM measured mass-size distribution by summing up the masses collected in the individual bins over the size range of relevance. Thus for the accumulation aerosols we have the mass concentration M_a as

$$M_a = \sum_{I=7}^{10} m_{ci} \tag{4}$$

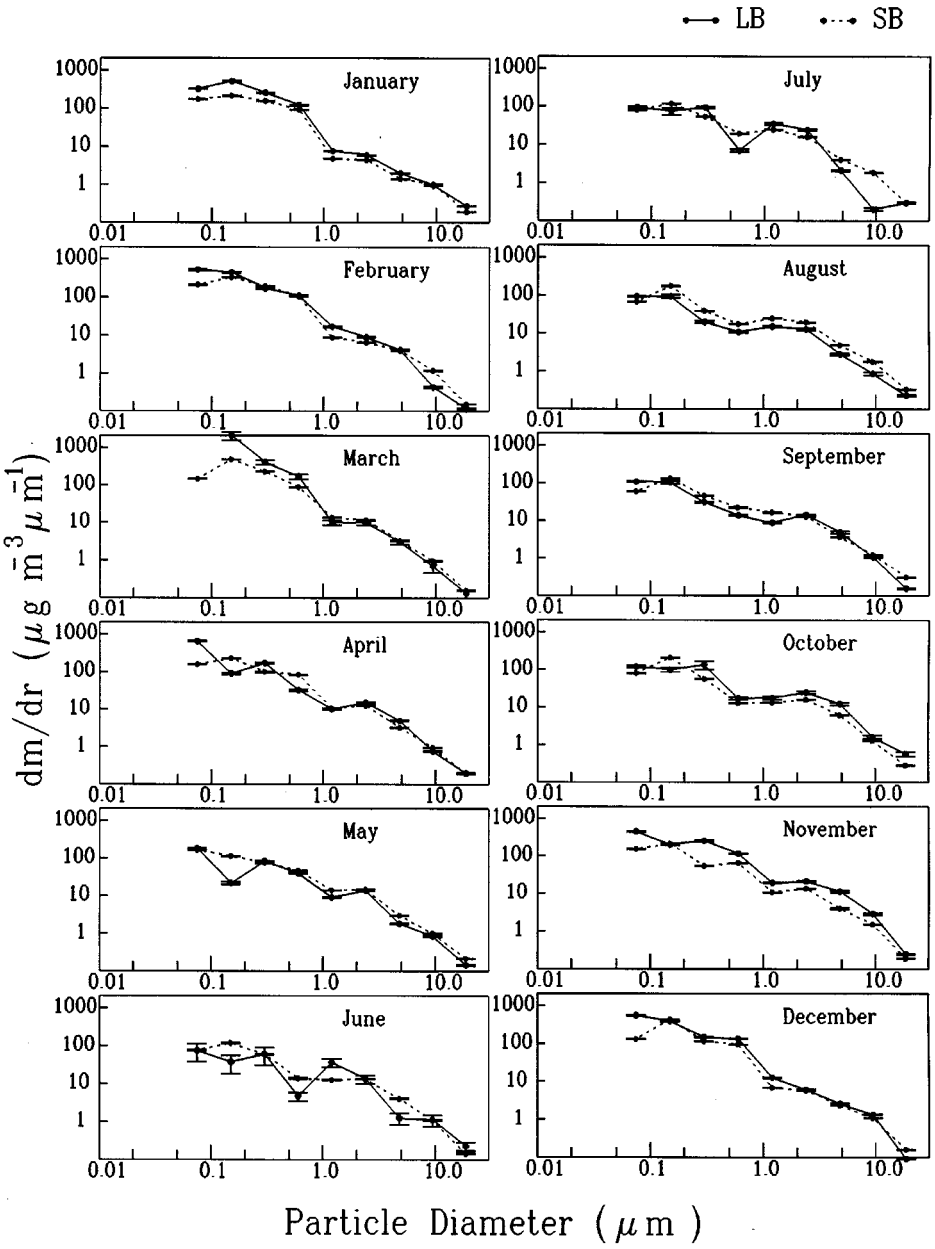

Figure 2. Change in the response of mass size distribution to SB (dashed line) and LB (continuous line) circulations with change in airmass type. The individual points are the monthly mean values of mass size distribution (Equation (3)) and the vertical bars through these are the standard errors.

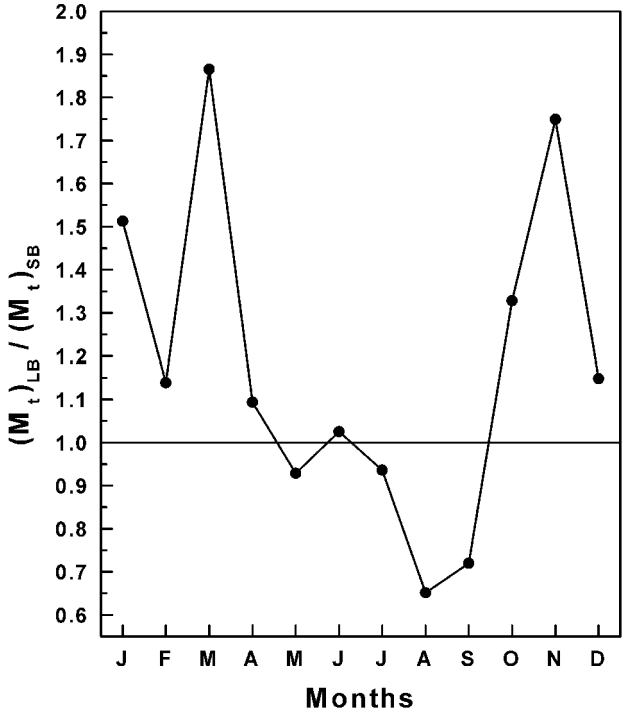

Figure 3. Annual variation of the ratio of total mass concentration under land breeze to that under sea breeze. Values less than 1 represent higher mass concentration during SB activity.

and for the coarse aerosols

$$M_c = M_t - M_a. \tag{5}$$

Both M_a and M_c are separated in terms of the breeze regimes and averaged over identical months and their variation over the year are shown in Figure 4 with coarse aerosols on the top and accumulation mode in the bottom panels. Similar to the earlier figure, the continuous lines depict the variation during the LBR and dashed lines that during the SBR.

It is readily seen that the accumulation mode mass concentration (M_a) is significantly higher under the LBR; but only during the months of November to April when the airmass, in general, is of continental type. After April, M_a under LBR falls rapidly and becomes almost independent of the breeze regimes till November (i.e., during the entire period of marine airmass). This clearly attributes the enhancement of M_a under the LBR (occurring during the *C* airmass period) to sources of continental origin. These become insignificant during the *M* airmass periods due primarily to the extensive rainfall, which plays the dual role of removal of aerosols by wash out and also of weakening the continental features conducive for aerosol

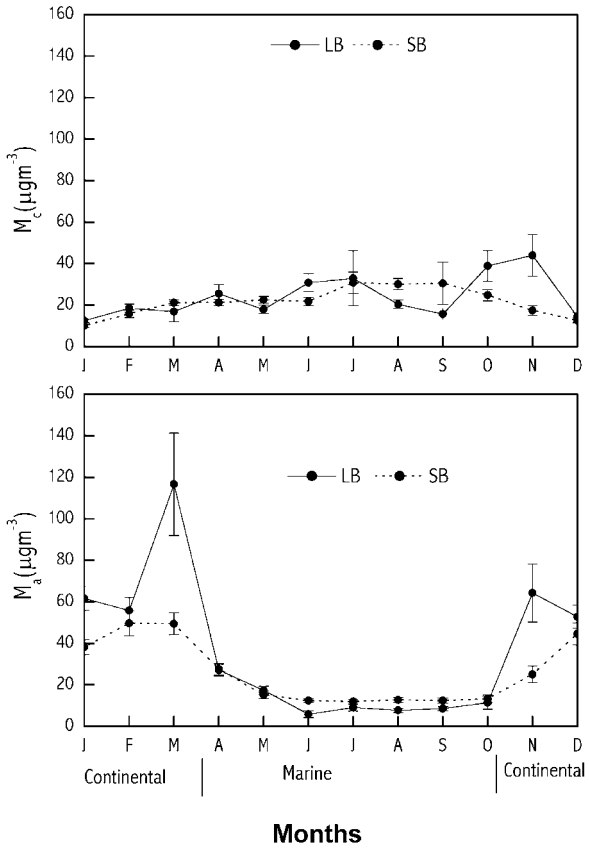

Figure 4. Annual variation of the mass concentration under LB (continuous line) and SB (dashed line) regimes for coarse (top panel) and accumulation aerosols (bottom panel). The vertical bars are standard errors.

generation, by the shift in airmass type. In contrast, the coarse aerosol concentration (M_c) increases gradually from the low values during *C* airmass period (under both the breeze regimes) to higher values, reaching a peak during the marine air mass period ($\approx$15 to 20 μg m^{-3}). However, the values of M_c in any month are not distinctively different in either the breeze regimes. The enhancement in M_c during the marine air mass period (from that in the continental air mass period) is small in absolute terms; nevertheless is quite significant (>200%). Further it is more fluctuating in nature (compared to M_a) and also is of comparable magnitude (within the error bars) in both the breeze regimes. This suggests that because the coarse aerosols are more nascent (closer to their sources) and rather short-lived, they respond faster to the changes in source strengths. Further, they appear to be more marine in origin as their increase in abundance is highly associated with the marine nature of the air mass. In this context it is useful to recall that winds over the sea surface are known to produce sea spray, the abundance increasing exponen-

tially with wind speed (Lovett, 1978; Fitzgerald, 1991; O'Dowd and Smith, 1993; Salzen et al., 1997). The favourable prevailing wind direction would advect these aerosols to the land to produce their signatures in the mass size distribution and spectral optical depths as observed by Moorthy et al. (1991). Vignati (1997) has reported from measurements conducted at the coastal site of Vindeby, Denmark that when the local wind is from the sea, the major constituents of the aerosols are sodium and chlorine and when it is from the land continental constituents like NHx and sulphate reach the highest concentrations. Vakeva et al. (2001) based on his observations at Helsinki (60° N, 41° E), reported that wind direction is the most important parameter controlling aerosol distributions at any given location. From observation at a remote island Minicoy (8.3° N, 73.04° E) in the Arabian Sea, Moorthy and Satheesh (2000) have reported an enhancement in aerosol optical depth at longer wavelengths during the periods when south-westerly winds are prevailing. Suzuki and Tsunogai (1988) reported, from observations made at the west coast of Okushiri island (42° N, 139° E), an increase in the concentration of sea salt due to transport of large particles from the Japan Sea by the strong north-west monsoon winds. Khemani et al. (1982), from observations made at Poona (18° N, 73° E), reported that average concentrations of chloride and sodium are significantly higher during the monsoon months than during the winter and attributed it to the maritime influence. They also found that the percentage contribution of coarse size aerosol was higher in monsoon periods whereas that of fine size aerosols was higher in winter. Examined in the light of all the above, the increase in M_c during monsoon season is suggestive of the increased impact of sea-spray aerosols.

Summing up, we note that during the period when the continental airmass prevails (November to March), $(M_a)_{LB} > (M_a)_{SB}$. As the airmass changes to marine, M_a decreases and also becomes insensitive to the breeze regime while $(M_c)_{SB} > (M_c)_{LB}$. The overall reduction in M_t is caused by the extensive rainfall during this period ($\approx$70% of the annual), which would remove both coarse and fine aerosols by wet scavenging processes (Pruppacher and Klett, 1978). However, sea-spray aerosol production, by the strong winds over the ocean, appears to replenish and often overcompensate the loss of coarse aerosols leading to an increase in M_c. But no such replenishment occurs in the accumulation mode. Consequently the mode in the size distribution shifts to larger sizes, even though the overall mass loading decreases. The low levels of coarse aerosols prevailing during the C airmass period (Figure 4) will also include continental materials such as wind blown dust, pollens, and leaf fragments and crustal materials (besides the sea salt aerosol) recycled by the land/sea-breeze cell. Similarly the recycled continental aerosols would also contribute to the M_a values during marine airmass periods.

Thus the share of accumulation and coarse aerosols to the total aerosol mass concentration varies with the season, as shown in Figure 5, where the mean share of M_a and M_c to M_t is shown for each season. Accumulation aerosols have the highest share in winter ($\approx$77%); it decreases gradually and the share is lowest in monsoon (31%) and rebuilds in post-monsoon (49%). On the contrary the share of

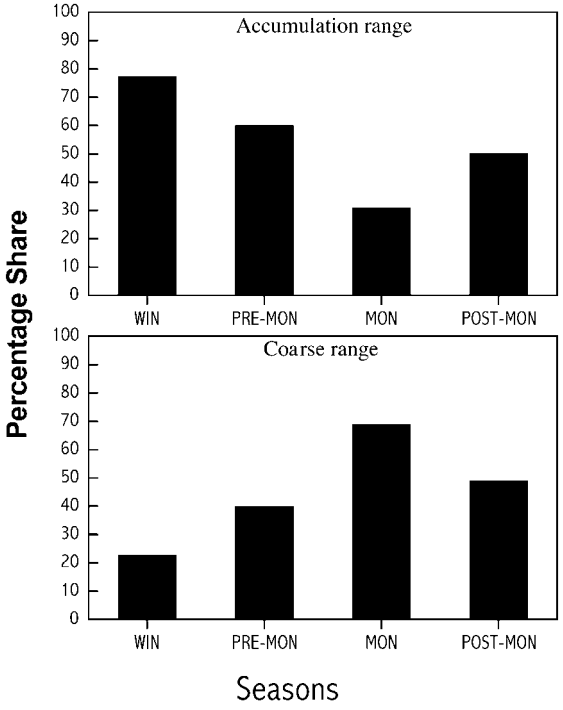

Figure 5. Seasonal changes in the percentage share of accumulation and coarse mass aerosol to total aerosol mass concentration.

coarse concentration has its maximum in monsoon (69%), and minimum in winter (23%). Thus in general continental environment prevails during north-easterly wind regime and a marine environment prevails during southwesterly wind regime.

6.2. Retrieved parameters

The mass size distribution measurements are used to retrieve other useful parameters of aerosols such as the effective radius (R_{eff}) and mass mean radius (R_m). Besides representing the physical characteristics of aerosols, these parameters are useful to model the optical properties of aerosols. The effective radius of a polydispersive aerosol system is equivalent to the radius required for a monodisperse aerosol to exhibit the same total scattering characteristics as the polydispersion (Hansen et al., 1974; McCartney, 1976). It is defined as the ratio of the third moment to the second moment of the aerosol size distribution (or the ratio of the total volume to total area).

From the mass concentration m_{ci} measured at each size bin the corresponding volume v_{ci} and area a_{ci} are estimated (Pillai and Moorthy, 2001)

$$v_{ci} = \frac{m_{ci}}{\rho} \tag{6}$$

and

$$a_{ci} = \frac{v_{ci}}{r_i}, \tag{7}$$

where r_i is the geometric mean radius of each size bin estimated as

$$r_i = \frac{1}{2}\sqrt{D_{p(i-1)} D_{pi}}\,. \tag{8}$$

Then the effective radius is estimated as

$$R_{eff} = \frac{\sum_{i=2}^{10} v_{ci}}{\sum_{i=2}^{10} a_{ci}}, \tag{9}$$

and the mass weighted mean radius is estimated as

$$R_m = \frac{1}{2}\frac{\sum_{i=2}^{10} d_{pi} m_{ci}}{\sum_{i=2}^{10} m_{ci}}, \tag{10}$$

where d_{pi} is the geometric mean diameter of each stage (= $2r_i$).

In estimating R_{eff} and R_m using Equations (9) and (10) the summation is carried over only from stages 2 to 10 because the first stage collects all particles with size exceeding 25 μm; and its r_i could not be determined. But while estimating M_t, the mass collected on the first stage is also added to the rest as in Equation (2). However, the mass concentration of the first stage is usually between 3 to 5% of the total mass on the rest of the bins.

The annual variations of the monthly mean values of R_{eff} and R_m (considering the entire data set) are shown in Figure 6 separately for the LBR and the SBR, the vertical bars representing the standard errors. In both the panels, points joined by the continuous lines correspond to the LBR, while those with the dashed line correspond to the SBR. Examining the figures, the following features are evident.

(i) The nature of variation of R_{eff} and R_m are quite similar. Both these parameters exhibit distinct responses to the breeze regimes as well as a distinct annual variation.

(ii) Both R_{eff} and R_m increase from low values in January to March (when C airmass prevails) to reach a broad peak during June to September (when M airmass prevails), followed by a gradual decrease towards December. The overall increase is nearly twice the value during January–March and resembles that of M_c in Figure 4. This is generally true for both the breeze regimes.

(iii) R_{eff} is generally higher during SBR irrespective of airmass type (except in June and November) indicating significant influence of coarse aerosols of marine origin in the SBR. R_m also shows a similar picture during the C airmass period. However during marine airmass conditions, it is rather insensitive to breeze regime.

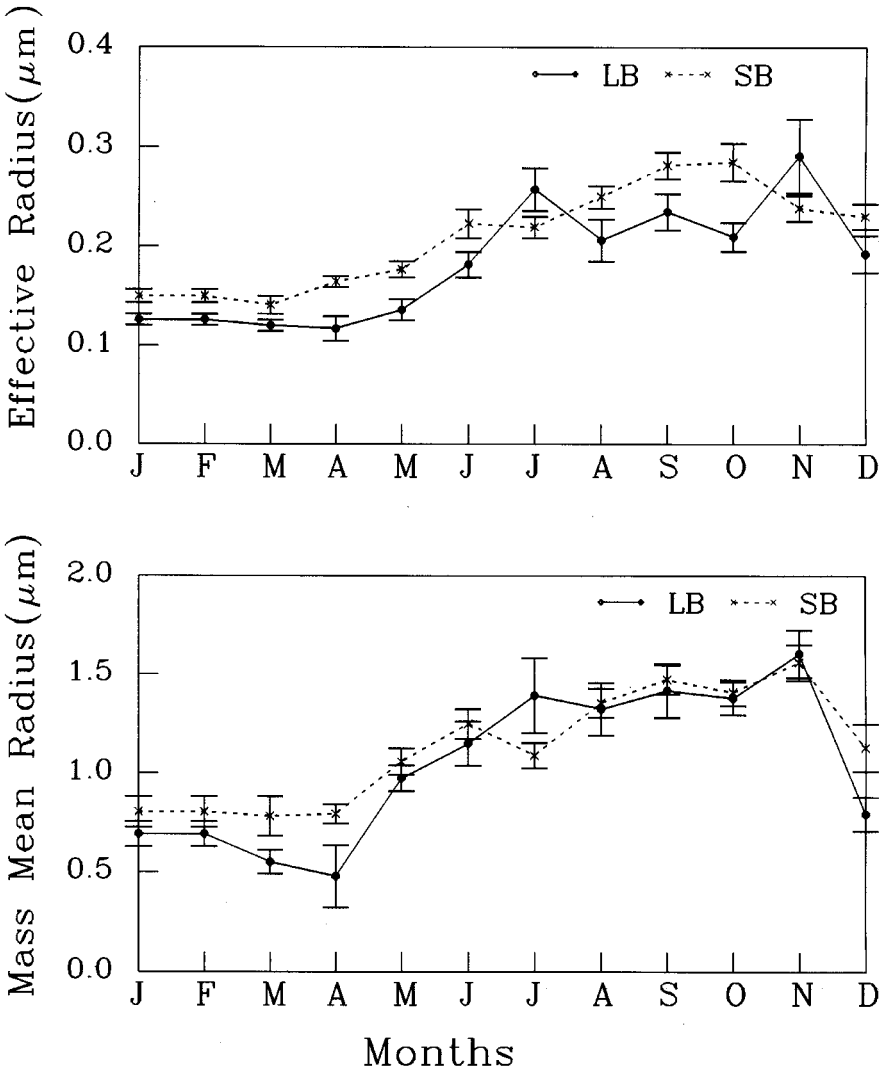

Figure 6. Annual variation of R_{eff} (top panel) and R_m (bottom panel). Vertical bars are the standards errors. Points joined by the continuous lines are for the LBR and those joined by the dashed lines are for the SBR.

The above observations imply that

(i) In general, coarse aerosols influence the aerosol size distribution more during the sea-breeze period (as both R_{eff} and R_m are more dependent on the coarse aerosols).

(ii) This effect increases during periods of marine airmass leading to an overall increase in the relative abundance of coarse aerosols; irrespective of the fact that there is a considerable wet scavenging during this period.

Thus, though the monsoon rains cause a significant decrease in the overall mass concentration due to wash out, the decrease is more extensive in the submicron/fine size range. As pointed out earlier, this is due to the weakening of local continental sources, and convective activity, and the opposing nature of the prevailing air mass. The larger particles are replenished by the advection of sea-spray aerosols (produced by the winds which become stronger) by the favourable air mass.

7. Summary

1. Near surface aerosols at the coastal station respond distinctively to land/sea-breeze circulations with higher mass concentration in the LBR during periods when a continental airmass prevails; This is attributed to continental source processes, which are quite strong in the accumulation size ranges ($d < 1\ \mu m$) and during the dry months, when a *C* airmass prevails. During months of marine airmass higher mass concentrations are seen in the SBR.
2. As the airmass changes to marine, the dominant mode in the mass size distribution shifts towards larger sizes. The accompanying rainfall though depletes the aerosol loading substantially, the effect is seen only in the accumulation mode. Coarse aerosols, in fact, increase in concentration, being replenished by the sea-spray activity and are mainly responsible for variations of aerosol loading during this season. During this period the aerosol properties are generally insensitive to land/sea-breeze activity.
3. The effective radius and mass mean radius are generally higher during SBR and also during marine airmass periods. The increase in these is more due to the change in dominant aerosol types.

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