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Aerosol size distribution over the Arabian Sea during ARMEX-II

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सार – अरब सगार में ऐरोसोल के आकार के वर्गीकरण (ए. एस. डी.) का अध्ययन करने के लिए मानसून पूर्व के आरमेक्स – II में 14 स्तरीय एंडरसन निम्न दाब इम्पैक्टर (एल. पी. आई.) और 4 स्तरीय प्रकाशिक कण काउंटर (ओ. पी. सी.) परिचालित किए गए। त्रिवेंद्रम में पहले से विद्यमान ए. एस. डी. के साथ इनके आकार के वर्गीकरण की तुलना की गई। एल. पी. आई. द्वारा मापे गए और ओ.पी.सी. का उपयोग करके मापे गए ऐरोसोल के बड़े आकार के वर्गीकरण से प्राप्त हुए संख्या आकार वर्गीकरण के मध्य अच्छा सामंजस्य रहा। महाद्वीपीय स्थल त्रिवेंद्रम की तुलना में महासागरीय पर्यावरण मे इस तरह के वर्गीकरण का प्रेक्षण अपेक्षाकृत समान (आकार सूची के लिए निम्न मान सहित) रहे। ए. एस. डी. का सब–माईक्रॉन मोड महासागरीय पर्यावरण में कम स्पष्ट रहा है। महासागरीय पर्यावरण में आकार सूची का कम मान होना बड़े आकार के कणों की अधिकता की ओर संकेत करता है।

ABSTRACT. A 14-stage Anderson Low Pressure Impactor (LPI) and 4-stage Optical Particle Counter (OPC) were operated in the pre-monsoon ARMEX-II cruise to study the aerosol size distribution (ASD) over the Arabian Sea. These size distributions are compared with the prevailed ASD at Trivandrum. A fairly good agreement is observed between the number size distribution derived from LPI measured mass size distribution and those measured using the OPC. These observed size distributions in the oceanic environments are relatively flatter (with a lower value for the size index) than those obtained over the continental site Trivandrum. The sub-micron mode in ASD is less pronounced in the oceanic environment. Low value of size index in the oceanic environment is an indicator of the dominance of larger size particles.

Key words – ARMEX, Aerosols, Aerosol size distribution, Size index.

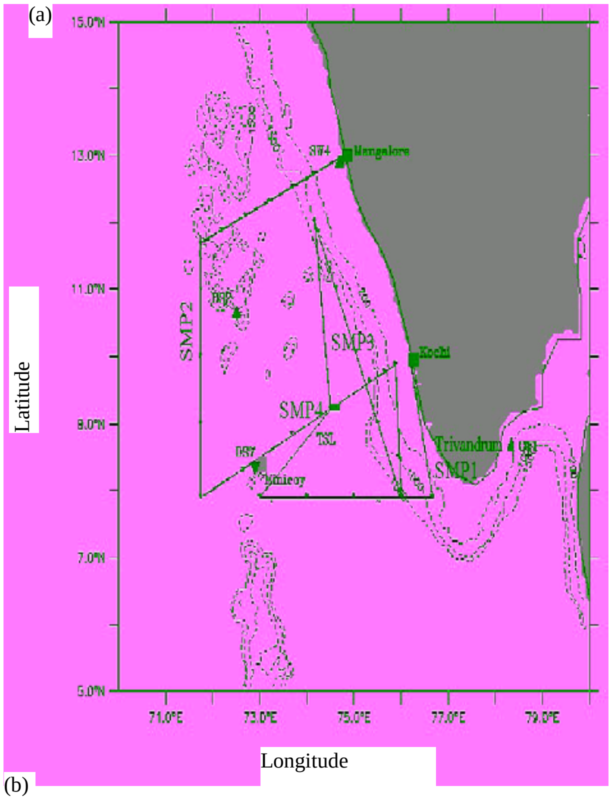
1. Introduction

The spatial distribution of aerosols in the oceanic environment influences the regional pattern of radiative forcing (Rajeev and Ramanathan, 2000; Tahnk and Coakley, 2002). As the monsoon circulation is mainly driven by the land-sea temperature contrast, the modification of spatial pattern of radiative forcing on a regional scale by atmospheric aerosols can significantly influence the monsoon pattern. Atmospheric aerosol can significantly influence the climate and weather system on a regional scale and also can influence the monsoon rainfall pattern (Lal *et al*., 1995) in the Indian context. The study of aerosol characteristics in the oceanic environment and its spatial distribution thus becomes important in monsoon related studies. The aerosol characteristics at a particular location depend on the prevailing meteorological conditions. In the far oceanic environments, major contribution to atmospheric aerosols comes from the *in situ* production of sea-spray aerosols. But, near the land-mass significant contribution comes through the advection of continental aerosols. Once advected over the oceanic environments, these aerosols undergo various transformation processes. Study of aerosol characteristics in the coastal oceanic environment thus becomes important in assessing the influence of continental advection.

2. Instrumentation

The 14-stage Anderson Low Pressure Impactor (LPI) size-segregates the aerosols in the aerodynamic size (diameter) range 0.04 to 35 μm in 12 size bins depending on the cut off size for each stage and collects the particles on glass substrate. The physical size range (stokes diameter) of particles collected in each size bin is estimated applying correction for particle density (Parameswaran and Vijayakumar, 1994). The mass of particles (m) collected in each size bin is estimated by taking the difference in mass of the collection substrate before (tare weight) and after sample collection. As the impactor size-segregates particles according to their sizes in the prevailing atmospheric relative humidity (RH) condition, appropriate corrections (Hanel, 1976) for the hygroscopic growth of particles with RH are to be applied. From these the mass-size distribution of aerosols is

(a)



(b)

Figs. 1(a&b). (a) The cruise track of the pre-monsoon ARMEX-II cruise from 14 March 2003 to 10 April 2003. The locations from which the aerosol samples are collected are also indicated and (b) A photograph of LPI during its sampling on the ARMEX-II cruise

obtained, the details of which are described in detail in earlier publications (Parameswaran, 1996; Parameswaran *et al.*, 1999). The sampling rate of LPI is 3 liters/minute and the instrument is operated for about 60-80 hrs depending on the aerosol environment to collect measurable deposits on the collection substrates. Weighing of the substrates is performed using a high sensitive microbalance (Metller model AT20) with sensitivity, 2 μg. The LPI measurement thus provides an average mass-size distribution for the duration of sample collection.

The Optical Particle Counter counts total number of particles in five different size bins (diameter); above 0.3 μm, above 0.5 μm, above 1 μm, above 3 μm and above 5 μm. The counter is battery-operated (Ni-Cd) instrument with a microprocessor controlling all instrument functions. Isokinetic probe is connected to the counter inlet tube (on top of counter) through which the air sample is sucked in using a pump. The sampling rate of this pump is 0.1 cfm. A diode laser is used as the light source for particle detection. Particles scatter light energy from solid-state laser diode onto the collection optics. The collection optics focus the light on a photo diode that converts the bursts of light into electrical pulses. The pulse height is proportional to particle size. Pulses are counted and their height is measured for sizing. From the cumulative number concentration in different bins, the differential counts are obtained by taking the difference of counts in successive bins. From this the number-size distribution is obtained.

3. Measurements

During the Second Phase of Arabian Sea Monsoon Experiment (ARMEX-II) from 14 March 2003 to 10 April 2003 (the pre-monsoon cruise) the 14-stage Low Pressure Impactor (LPI) and hand held Optical Particle Counter (OPC) are carried on the cruise along with other instruments for making measurements on aerosol concentration and size distribution in the oceanic environment over the Arabian Sea. Fig. 1(a) shows the cruise track. The cruise started from Mangalore on 14 March 2003 and reached the "Time Series Location" (TSL) taking a rectangular path. The ship then sailed towards the coast and made a southward excursion up to 8° N followed by a northward journey up to 12° N and returned to the TSL location. After TSL the ship sailed towards Minicoy before docking at Cochin port on 10 April 2003.

Three sets of aerosol measurements were carried out using LPI in the ARMEX cruise at different regions over the Arabian Sea. Fig. 1(b) shows the aerosol instruments installed on Sagar Kanya. The LPI was installed in the bow side of the ship which ~20 m above the water level. Aerosol Samplings were made only when the ship is cruising or when wind is towards the bow side (when ship is stationary). The LPI is operated almost continuously for 3-4 days (amounting 60-80 hrs) depending on the environment condition, to collect measurable amount of aerosol samples. Before taking on to the ARMEX cruise the LPI was operated at Trivandrum (8.5° N, 77° E) during the period 27 February 2003 to 05 March 2003 to

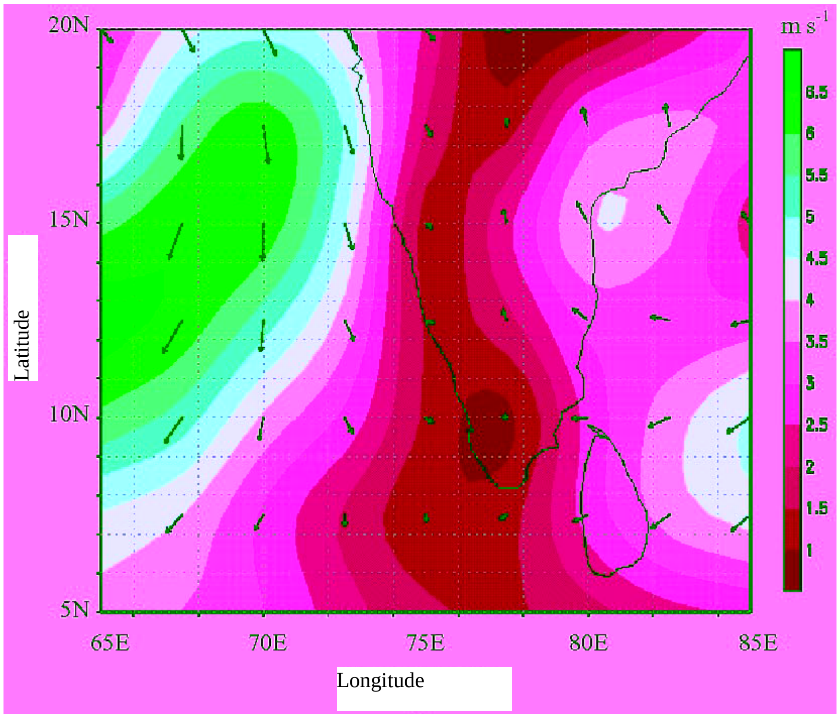


Fig. 2. Mean isotachs (m s^{-1}) at 1000 hPa level near the Indian Peninsula for March 2002 obtained from NCEP/NCAR reanalysis (website: http://www.cdc.noaa.gov)

monitor the prevailing aerosol concentration and size distribution. This sample will be referred to as SMP1 for the following discussions. Other three samples designated as SMP2, SMP3 and SMP4 respectively are collected over the Arabian Sea at different oceanic locations. The first sample (SMP2) on cruise was collected during its starting phase from 15 March 2003 to 19 March 2003 in the latitude-longitude region 8°-12° N, 72°-75° E. The second sample (SMP3) was collected from the latitude-longitude region 8°-11° N, 74°-75° E during the period 20 March 2003 to 23 March 2003. During this period ship made an up and down voyage in the latitude region 8°-11° N around 74.5° E. The third sample (SMP4) was collected at the TSL (around 9° 13′ N, 74° 30′ E) when the ship remained stationary for 15 days. Sufficient care is taken to make the sampling when the wind is blowing towards the bow side of the ship such that any contamination from ship exhaust is not affecting the samples. The regions from which the samples are collected are marked in Fig. 1(a).

The optical particle counter (OPC) was operated regularly on everyday during ARMEX cruise from the bow side (where the LPI is operated) of the ship by holding the instrument in the hand. The aerosols particles are counted for about one minute for each size range. Thus the OPC sampling is made within a time duration ~5 minutes. On an average 2-3 observations are taken in each hour (mostly during the day). From the particle counts (converted to number density) obtained in five size ranges, the aerosol number concentration in four radius bins 0.15-0.25 μm, 0.25-0.5 μm, 0.5-1.5 μm and 1.5-2.5 μm (corresponding to 0.2 μm, 0.375 μm, 1 μm and 2 μm mean radius respectively) are estimated. The aerosol size spectra obtained in terms of particle radius (r) from LPI and OPC are used for further analysis.

4. Meteorological features

The meteorological parameters such as wind speed, wind direction, relative humidity and temperature at the deck level (of the ship) are obtained from the meteorological instruments associated with Sagar Kanya. The mean prevailing wind pattern at 1000 hPa level (near the surface) for March 2003 in the ARMEX region obtained from NCEP/NCAR reanalysis (Kalnay *et al.*, 1996) is shown in Fig. 2. The prevailing wind is north-northeasterly. The maximum wind speed was always less than 6.5 ms^{-1}. Relatively high wind speeds between 4 & 6.5 ms^{-1} were observed in the longitude region 65°-70° E. In the other region, the wind speeds were always less than 4 ms^{-1}. A significant high in wind speed is observed in the mid Arabian Sea followed by a convergence along the

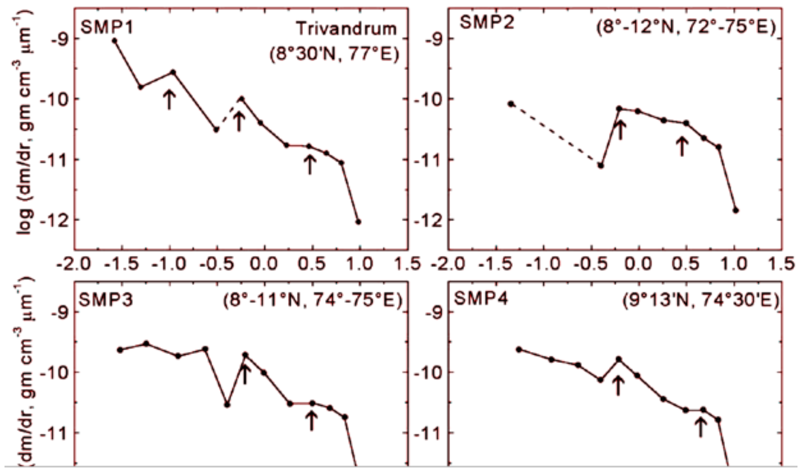


Fig. 3. The mass-size distribution of aerosols at different locations obtained from LPI

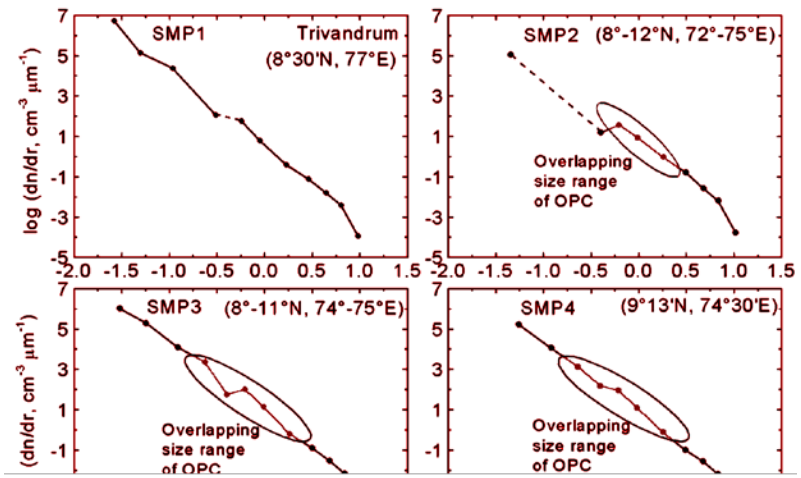


Fig. 4. The number-size distributions derived from the mass-size distributions shown in Fig. 3 (LPI measurements)

west coast. It may also be noted that the wind vectors over the Arabian Sea and Bay of Bengal are oppositely directed favouring a convergence in the peninsular region resulting a very weak northerly flow. The same feature is revealed from the measurement carried out using the meteorological instruments located in Sagar Kanya also. The atmospheric RH in the cruise region was between 60 & 75%.

5. Results and discussion

The data obtained from LPI measurement are used to derive the mass-size distribution of aerosols at different locations under prevailing atmospheric conditions. Fig. 3 shows these size distributions for Trivandrum (SMP1) and over the three locations over the Arabian Sea (SMP2, SMP3 and SMP4). In SMP2 taken during the first phase

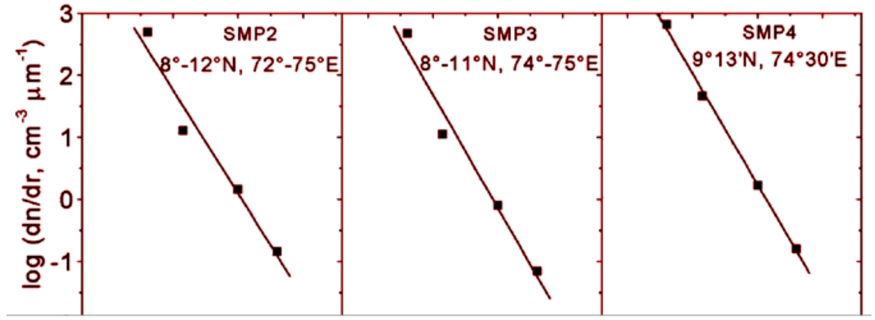


Fig. 5. The mean size distribution at the three oceanic regions (SMP2, SMP3 and SMP4) obtained from OPC observation. The best-fit straight lines obtained through regression analysis are also shown

of the cruise, the mass of aerosols collected in the sub-micron size regime is too small to be detected through gravimetric analysis. However, there were measurable sample in the higher size bins. In all the other three samples measurable quantity of aerosols could be collected in almost all the size bins.

Fig. 3 shows that in general the aerosol mass concentration decreases with increase in size. There is a sharp decrease in mass concentration in the last size bin. This rate of decrease is much larger than the average gradient below 6.5 μm radius. The size spectra of the oceanic samples (SMP2, SMP3 and SMP4) are relatively flatter, than that at Trivandrum (SMP1) indicating a relative dominance of larger size particles in the oceanic samples. Considering the mass concentration aerosol particles below 6.5 μm radius, the size index of aerosol size distribution (Junge, 1963) for Trivandrum (SMP1) is –0.74 ± 0.1. The size indices for the other three regions (oceanic) are –0.28 ± 0.19, –0.50 ± 0.11 and –0.55±0.07 respectively. These values of size index for SMP2, SMP3 and SMP4 are significantly lower than that of SMP1 taken at Trivandrum. Considering all the data points in Fig. 3, the mean size index for Trivandrum is –0.80 ± 0.2 and that for the other three samples are respectively –0.43 ± 0.2, –0.7 ± 0.15 and –0.80 ± 0.2. The number-size-distributions derived from these mass distributions are presented in Fig. 4. The size index for the number size distribution is –3.74 ± 0.1 for Trivandrum (considering data below 6.5 μm) and that for other samples are respectively –3.28 ± 0.19, –3.50 ± 0.11 and –3.55 ± 0.07 (3 more than the size index for mass distribution).

In addition to the overall decrease in concentration (*n*) with particle size, Figs. 3 & 4 shows small peaks at two/three values of *r*. These fine structure peaks are due to the presence of different modes in composite size distribution. This essentially indicates that the aerosol size distribution is not essentially power law type but a combination of two or three lognormal distribution with characteristics values for the 'mode radius' and 'half width'. Each of these size distributions corresponds to distinct aerosol sources, even though the composite model can be approximated to a power law. Examining the size distribution for Trivandrum it can be seen that there are three prominent modes with radii around 0.11 μm, 0.57 μm and 2.9 μm. The oceanic samples however indicate presence of two prominent modes only (indicated in Fig. 3 by vertical arrows). For SMP2 the modes are around 0.62 μm and 3.14 μm radii and for SMP3 they are respectively at 0.62 μm and 3.2 μm. For SMP4 the mode radii are around 0.61 μm and 4.7 μm. The sub-micron mode around $r \approx 0.11$ μm prominently seen in Trivandrum (SMP1) is less pronounced in the oceanic samples. The mode radius for second mode is larger in oceanic samples. This can mainly be attributed to long-lived accumulation mode sea-spray particles originating from sea-surface. The third mode in the coarse size regime can be attributed to nascent sea-spray aerosols. The value of this mode radius is also large for oceanic samples.

The dynamic size range of LPI for the prevailing RH is 0.02 μm –11 μm (mean radius). The observed dry aerosol mass-concentration (adjusted to RH→0) in the entire size range (of LPI) is 155 μg m^{-3} for Trivandrum, 189 μg m^{-3} for SMP2, 257 μg m^{-3} for SMP3, and 235 μg m^{-3} for SMP4. Aerosol mass concentrations over the ocean were larger than that over the continent. There is an increase in particle concentration near the coast. As seen from Fig. 2, there is convergence zone near the west coast of Indian Peninsula, which is favourable for accumulation of aerosols. The observed high in aerosol concentration near the coast could be attributed to this feature associated with synoptic wind near the surface.

The Optical Particle Counter (OPC) gives the instantaneous number concentrations in different size bins. In order to have fruitful comparison of size distribution obtained from LPI, the number-size distributions from OPC during the LPI sampling period are averaged to obtain the mean number-size distribution. The mean number-size distribution thus obtained from OPC corresponding to SMP2, SMP3 and SMP4 regions are presented in Fig. 5.

The mean number concentrations in different size bins observed by OPC compares favourably with those observed in the respective size ranges of LPI. The mean slopes (size index) of these distributions are compared with the slopes corresponding to the distributions in Fig. 3 in the overlapping size range 0.15 μm to 2.5 μm. The mean size indices are obtained through regression analysis. The size index for SMP2, SMP3 and SMP4 obtained from OPC are respectively -3.33 ± 0.46, -3.65 ± 0.42, -3.57 ± 0.12. The size index for the same size regime from LPI measurements shown in Fig. 3 (enclosed by an ellipse in these figures) are respectively -3.41 ± 0.09, -3.64 ± 0.63, -3.50 ± 0.31. This shows that the aerosol concentration and size distribution derived from these two different types of measurements are in good agreement.

6. Conclusions

Compared to that over the continent, the size distribution is relatively flat in the oceanic region. This indicates a relative dominance of larger size particles over the ocean. The ASD shows the presence of three distinct modes in size spectra; a fine mode dominated by the continental component, an accumulation mode dominated by long-lived oceanic aerosols and a coarse mode dominated by nascent oceanic aerosols. Number-size distribution obtained from LPI and OPC compares favourably in the over lapping size range.

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