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The sub-micron aerosol size-distribution measurements during the XVI Indian Antarctica Expedition

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Abstract

Measurements of the size distribution of sub-micron aerosol particles in the range of 0.003 to 1/μm have been made with an Electrical Aerosol Analyzer over the Indian ocean and at Maitri during the sixteenth Indian expedition to Antarctica. Observations show that the high concentrations of aerosol particles observed over the Indian ocean in the northern hemisphere extend upto the southern limit of Inter Tropcial Convergence zone. Our measurements indicate the accmulation of aerosol particles in ITCZ. Beyond ITCZ in the southen hemisphere, typical low aerosol concentration of pristine air are observed. Over open ocean, the size-distribution of particles is observed to be bimodal with a maximum at 0.133μm and two minima at 0.075 and 0.42μm and aerosol concentrations are open ended at both ends. Observations also indicate production of 'large' particles in roaring forties and enhancement of aerosol concentrations associated with the low pressure systems circulating around the continent of Antarctica. Gradients of aerosol concentrations as one goes away or approaches the landmasses of India and Durban, are observed.

At Maitri, the aerosol concentrations in the observed size range are of the order of 10^2 - 10^3 cm^{-3} during this season. Some peaks, possibly associated with low pressure systems circulating around the continent of Antarctica, reaching upto 10^4 particles cm^{-3} are observed. In general, aerosol concentrations remain quite constant for several hours and have bimodal size distributions not much different in shape from those observed over open ocean. The results indicate that the particles in nucleation mode generated by the gas-to-particle conversion process are transported close to the ground under subsidence of the midtropospheric air.

Introduction

The atmospheric aerosol particles of size ranging from 0.001 to 100 μm exist in the free atmosphere and play important roles in atmospheric processes such as cloud formation, ice nucleation, radiative transfer, chemical reactions, wet and dry removal and so forth. Over oceans, part of the marine aerosols originates directly from ocean, but a large fraction of it could be advected from the continents or be formed within the atmosphere. The submicron size aerosols

play important role in the formation of cloud droplets in the precipitation process and are of vital importance in assessing the background air pollution over oceans. The number, size distribution and composition of marine aerosol particles vary with time and space in response to a number of different processes such as the transport of continental air, the bursting of whitecaps and the oxidation of gases emitted by the oceans etc. There are several experiments conducted in the Pacific and Atlantic oceans to study the physical, chemical and optical properties of submicron aerosols (e.g. Hoppel et al. 1985, 1990, Hoppel and Frick, 1990. Colvert, et. al. 1996) However, inspite of some preliminary measurements reported by Lal and Kapoor (1992) and Jayraman et al. (1998), detailed data on the spatial and temporal distributions of the aerosol concentrations over the Indian ocean is lacking.

The vast ice sheet of Antarctica is located in the southern hemisphere with no or very little human activity and has only about 2% bare land area exposed to the atmosphere. Thus the local production of aerosols from the surface over this continent is almost negligible. Further, the air circulation over Antarctica is isolated from the lower latitudes to transport air at the lower tropospheric heights. In spite of these facts, the measurements of submicron-size aerosols made over Antarctica indicate that their transport takes place over global scale distances. There fore, Antarctica provides an ideal site, free from anthropogeanic pollution, to study the background aerosols. Some of the earlier investigations of the properties and behaviour of aerosols at Antarctica are reported by shaw (1979, 1988), Lal and Kapoor (1989), Ito (1993), Hogan and Gow (1993) etc.

Here, we report our measurements of the submicron aerosol size-distribution made along the cruise track onboard Polar Bird and at the Indian station, Maitri (70° 45' 52" S, 18° 44', 03"E, 117 m above msl), during the XVI Indian Antarctic Expedition in the summer of 1996-97. The results are interpreted in terms of the local meteorological and synoptic conditions.

Instrumentation and sampling

The aerosol measurements were carried out with a TSI 3030 Electrical Aerosol Analyser (EAA) System. It measures the size distribution of particles of diameter 0.003 to 1μm in 10 different size ranges. The accuracy of contributions from the lowest 2 channels i.e. for 0.003 and 0.007 μm diameter-ranges is not sufficient due to the limitation of operating the instrument where the rate of generation of photochemically generated aerosols is highly variable, and hence, as discussed later in this report, the contributions from these channels are not considered in this analysis. The EAA system was operated after every 3 hours to collect 5 size-distribution samples and the data was in a PC. In the present analysis, we use the averge of 5 samples collected after every 3 hours. The inlet of the EAA system was cleaned frequently to avoid accumulation of sea salt, snow or dust at the inlet.

The air was sampled at a height of 12 m above mean sea level through a 1-cm diameter stainless steel tube of 1-m length which was projected outward form a cabin and was properly grounded. The EAA system was installed inside the cabin. The position of the inlet to collect the air sample was so slelected that the exhaust of the chimney of the ship does not pollute the aerosol measurements.

At Maitiri, the aerosol measurements were carried out in a summer hut, namely Tirumala. The inlet of the air sampling stainless steel tube was projected out at height of 2 m above ground through a window such that most of the time, it faces the persistent wind direction. The single storeyed building of Maitri station and other structures such as generator huts, gas plant, incinerator were about 150-200 m away in the southwest direction from the site of the measurements so that any pollutant released from them had little or no chance of reaching the site of measurements.

The aerosol size-distribution measurements were carried out onboard Polar Bird from December 15, 1996 to January 4, 1997 during the onward journey and from March 9 to April 4, 1997 during the return journey. At Maitri, the measurements were conducted form January 11 to February 24, 1997.

Aerosol concentrations over the Indian ocean

Fig. 1 Shows the total aerosol concentration in the size-range ($0.01 < d < 1.0$ μm) obtained by adding particle concentrations in each of the 8 channels of EAA system along the onward and return cruise tracks. No data could be obtained between 32° S and 46° S on the onward journey due to very rough sea conditions. The number at the top of the x-axis represent the latitude at 1200 GMT of each day. Lower two sections in Figure, (a) and (b) are for onward journey and sections. (c), (d) and (e) are for return journey. The cruise routes are given in Fig. 1 of author's accompanying article in this report.

The total aerosol concentration increases as the ship approaches the landmasses of Africa or India. Similarly, a decrease in the total aerosol concentration can be noticed as the ship goes away from Durban. Such increases or decreases in aerosol concentration near the landmasses are generally observed and are due to the transportation of the aerosols from the continents (see e.g. Deshpande and Kamra, 1995 and Kamra and Deshpande, 1995). The influence of the closeness of landmass can also be observed in comparatively lower values of aerosol concentrations in 20° - 30° S belt observed on the onward cruise as compared to those observed on the return cruise when the ship berthed at Durban. The aerosol concentration shows some peaks near the Antarctica coast in both the onward and return cruises. Such enhancements in aerosol concentrations are most likely associated with the low pressure areas which form a "wall of storms" around the continent of Antarctica.

On the onward journey, three peaks of comparatively high aerosol concentrations are observed between the equator and 8°S. The positions of these peaks are within the Inter Tropical Convergence Zone during these periods.

On the onward journey, comparatively low and nearly constant values of aerosol concentrations are observed from 8°S to 30°S and to some extent also from 46° S to 65° S. On the return journery also such low and nearly constant values are observed from 56° S to 40° S. Such values of aerosol concentration are typical of the prestine air in the southern hemisphere. At about 41°S on the return cruise, the surface wind direction suddenly changes by 180° form westerly to easterly or north-easterly direction. North of 41° S, the aerosol concentrations remain comparatively high and variable.

Aerosol size **distributions over the Indian Ocean**

From the large number of size distributions obtained during the cruise, we present in Fig. 2, some typical size distribution curves observed in different latitudinal belts during the onward and return cruises. On the onward cruise (Fig.2) the size-distribution curves for all latitudinal belts except Lhat for the 40° - 60°S belt, show a maximum at 0.133 um and two minima at 0.07 and 0.42 µm. The size distribution curves are open ended and the aerosol concentrations keep increasing on both sides of the minima. In the 40 - 60° S belt, the maximum occurs at 0.042μm and the minima at 0.024 μm and 0.42 μm. The concentration of large particles in accumulation mode is the least in the 40 - 60° S belt, show a maximum at 0.133 μm and two minima at 0.07 and 0.42 Aim and the minima at 0.024 μm and 0.42 μm. The concentration of large particles in accumulation mode is the least in the 40 - 60° S belt and the highest in the 5° N - equator belt. This indicates the transportation of the continental aerosols with the prevailing northeastern winds in the equator to 15° N region and the local generation of large particles by breaking waves on the sea surface due to strong winds in the roaring forties. Actually, the contribution of aerosols in accumulation mode are higher in the 5° N-equator belt than in all other belts in the southern hemisphere. Also, the aerosol concentration of the 0.013μm is higher in the 5° N to 18° S region as compared to the region south of 20° S. In our observations, the aerosol size distribution in the southern hemisphere remains constant over several hours. Both, the decrease in the concentration of particles from 0.133 to 0.422 µm in size and the increase in the concentration of particles from 0.422 µm to 0.75 µm in size are much steeper in the southern hemisphere than in the northern hemisphere.

Fig. 2b shows the aerosol size-distributions for different latitudinal belts during the return cruise from March 9 to April 4, 1997. The Size distributions observed in the 70 - 60° S and 60 - 40° S belts are similar in shape to those observed in these belts during the onward journey. In the 40-30° S and 10° - 5° S belts the size distributions are monomodal and open ended at both ends. In the 10° -15° N belt particle concentrations in all sizes are higher by about an order of magnitude than in all other latitudinal belts, especially south of the ITCZ and the size distribution show a maximum at 0.133 µm and a minimum at 0.42µm. However, it need be emphasized that unlike in all other size distribution curves in the onward and return cruises, the particle concentration in the accumulation mode in 10 - 15° N belt is relatively higher. Further, instead of showing a minimum at

0.42μm the particle concentration keeps decreasing in the 10° - 15° N and 40 - 30° S belts. Once again, this indicates either the transportation of the continental aerosols with the prevailing northeastern winds in the 10 - 15° N belt and the local generation of large particles by breaking waves on the sea surface due to strong winds in the 40° - 30° S belt Further, it needs to be emphasized that while the aerosol size distribution in the belt from 20° -10° S is of typical bimodal type observed over oceans, it changes to monomodal type in the 10 - 5° S belt upto which the ITCZ extends during this period. Moreover, there is significant increase in the aerosol number concentration of each size category in ITCZ. In the region north of the equator where winds are northeasterly or northwesterly, our observations show significant variations in the aerosol size distribution within a period of few hours

Extension of continental aerosols over ocean

Extension of continental aerosols to a distance of a few hundreds of kilometers over oceans has often been reported near landmasses. (e.g. Deshpande and Kamra, 1995). During our expedition, the ship berthed at Mauritius and South Africa, The extension of continental aerosols, while approaching the landmasses from the open sea or departing from them, is very well signatured in our measurements.

Fig. 3 Shows the change in the aerosol size distribution when the ship, on her return cruise, berthed at Durban from 0530 to 2205 GMT on March 20, 1997. The aerosol size distributions at 0000 and 0300 GMT, obtained at open ocean while approaching Durban, are bimodal which is typical of the aerosol size distributions over oceans. Also, the concentration of aerosols in accumulation mode ($0.13 < d < 1.0\mu m$) is comparable with that in the nucleation mode ($0.013 < d < 0.13\ \mu m$). The size distributions at 0600, 0900, 1200, 2100 GMT, obtained at Durban harbour, are monomodal and show comparatively higher concentrations of the particles in each size category. After departure from Durban, the aerosol concentrations decrease and their size distributions systematically attain the

oceanic bimodal nature. Similar reuslts obtained when approaching or departing from Port Louis, Mauritius.

Effect of fog

There was on occasion of the occurrence of fog during the onward cruise when the ship was idle in the pack ice at 69° 40'S, 11° 55' E near to the Antarctica coast. The fog set-in at 2100 GMT on January 5, 1997 and dissipated at 0300 GMT of January 6, 1997. Being within the Antarctic circle, there was almost 24 hours sunlight to observe the fog conditions even at midnight. Fig 4 shows the size distributions of aerosol particles before, during and after the fog situation. A few hours before the onset of fog and after the dissipation of fog, the particle size distributions show a maximum at 0.113 μm and two minima at 0.075 and 0.42 μm with the particle concentration increasing on both ends of the size-distribution. A few hours before the onset of fog, the peak shifts between 0.042 and

0.075 μm size-range and the minima in accumulation mode disappears. After the onset of fog, the particle concentrations in each size range decrease by about and order of magnitude.

Aerosol Concentrations at Maitri

Fig. 5 shows the 3-hourly values of the total aerosol concentration obtained by adding the number concentrations in each of the eight size categories from 0.013 to 0.75 μm, from January 10 to Febuary 24, 1997 at Maitri. The total aerosol concentration is generally less than 1000 particles/cm^3 in the absence of any meteorological phenomena. During summer months hs, concentrations of condensatinon unclei of the same order have been reported at different Antarctic costal stations, e.g,by Ito(1993)at Syowa(69 °S, 39.6° E), by Gras and Adriasseson (1985)at Mawson (67.6° S, 62.9° E). shaw (1986) at Ross Island in summer months. Earlier measurements at Maitri by lal and Kapoor (1989) also show similar concentrations of Aitken nuclei in the months of January and February. The measurements of Voskresenskii (1968) at Mirny (66.6° S, 93° E), however, show condensation nuclei concentrations of only upto a few hundreds of particles cm^{-3}. Most of these observers report a decrease in nuclei concentration by an order or two in winter months,

Our measurements, especially for the month of February show some peaks of very high aerosol concentrations. Such peaks in aerosol concentrations have also been observed at other Antarctic coastal stations such as Mirny (Voskresensii, 1968), Siple (Hogan 1975), Syowa (Ito, 1980) and even at the South Pole (Hogan, 1979). These peaks are most likely associated with the passage over the station of circulating cyclonic storm systems around the continent of Antarctica. The 3-hourly observations of surface pressure and cloudiness made at Maitri and plotted in Fig. 7, support such correlation.

Size distribution of aerosols at Maitri

In Fig. 6, are plotted the obcrvations of the aerosol size distributions for February 6, 1997 which is a typical fair-weather day without any low pressure system or high wind conditions over the station. Except at little higher values of aerosol concentration of 0.1 to 0.4 μm particles from 2100 to 0600 hours, all size-distributions are almost similar to each other and there is no typical diurnal pattern in the size distribution or concentrations of aerosols. All size distributions are bimodal with a peak at 0.133 μm and minima at 0.075 and 0.42 μm and is open ended at both ends. Our observations show that the total aerosol concentrations and their size-distribution at Maitri remain unchanged for several hours provided there is no substantial change in the meteorological conditions at the station. Earlier observations of size distribution by Ito (1993) at Syowa station show a peak at about 0.06 μm diameter in December in the size-range common to that of ours. Also, Harvey et. al. (1991) in their observations in Ross sea region did not observe a minimum at 0.42 μm such as is observed at Maitri.

Changes in the aerosol concentration and size distribution associated with the low pressure systems The air circulation over Antarctica occurs via direct transport of air orginating at lower latitudes through the lower troposphere. The walls of storms' formed by polar lows around the Antarctica Convergence Zone makes stong barrier for aerosol transport. Under the influence of depression moving east-southeastward between latitudes 65° - 70° S, the low pressure systems occur over Maitri. There was a sequence of rise and fall in surface pressure over Maitri during the months of January and February. As shown in Fig. 5, a dip in suface pressure is most of the time proceeded by a peak in aerosol concentration.

Fig. 7 shows the change in the total aerosol concentration associated with a frontal weather system. The atmospheric pressure starts decreasing from 0800 hours on February 18, 1997 and attains its minimum value at 1800 hours on February 20, 1997. Two peaks in aerosol concentration, the first one showing an increase of about an order of magnitude and the second one of 2-4 times, in aerosol concentration are observed during this period which is followed by a period of cloudiness. Each peak in aerosol concentration lasts for about 12 hours. This is one of the few occurrences, observed at Maitri during this period, of the increase in total aerosol concentration associated with a frontal weather system. Similar observations are also reported by Lal and Kapoor (1989) at Maitri, Voskresenskii (1968) at Mirny station and by Hogan (1975) at Siple station in the Antractic continent. The aerosol size distributions on 18-19 February, 1997, shown in Fig. 8, show that it remains bimodal throughout these days. However, during the period of peaks from 0000 to 0600 hours, the concentration of particles of diameter > 0.4° μm remains almost unchanged.

Discussion

Persistent northeast winds in the northern Indian ocean from the months of January to April, bring with them the polluted air from the landmasses of India and Arabia. A North South gradient of aerosol Concentration observed in our observations in this region strongly supports such transport of aerosols. This polluted air of the northern hemisphere meets and mixes with the presitne air of the southern hemisphere in the Inter Tropical Convergence Zone (ITCZ).Since in this region, ITCZ is located south of equator during this season, the polluted air from the northern hemisphere penetrates well into the southern hemisphere and carries its pollutants upto the southern limit of the ITCZ. These pollutants may be convected up with the deep convective activity in the region of ITCZ and get distributed over larger area longitudianlly at higher altitudes. During the periods of no or weak convective activity in the ITCZ, the pollutants carried by winds from the northern hemisphere, may accumulate in ITCZ at lower altitudes. Since the air circulation in the ITCZ is characterized with eddies, the accumulation of air pollutants in this region may result in forming some pockets of high aerosol concentration. Observation of some peaks in aerosol concentrations in ITCZ in our measurements supports the formation of such pockets of very high aerosol concentrations.

In this season, the northeastern winds over the Indian ocean transport with them not only the aerosols, but also the trace gases the landmasses of India and Arabia. In addition, some trace gases are generated over the sea. With the abundance of solar radiation available in the tropics, production of the aerosol particles by the gas-to-particle conversion processes expected to be quite large in this area. The presence of very large concentrations of small particles in nucleation mode in our observations in this area supports such hypothesis.

The transportation of continental aerosols and trace gases beyond the southern limit of ITCZ is almost negligible. As a consequence, the size distribution curve for the 20° - 25° S belt in Fig. 2 a shows comparatively small concentration of the particles in nucleation mode. Stong surace winds in the roaring forties generate large number of particles by the breaking waves on the sea surface. The curve for the 30° - 40° S belt in Fig. 2b, shows not only the large concentrations of particles in each size-range but also the change in their size-distributions from bimodal to monomodal with comparatively much higher concentrations of 'large' particles. The region of the 40° - 60° S belt is almost free from the continental air and the wind direction is persistent for several days. Thus the total aerosol concentration is mostly steady and their size distribution is bimodal. Further south, in the Antarctic water (South of 60° S) the region is often frequented by low pressure systems circulating around the Antarctic continent associated with the Antarctic Convergence Zone. The peaks in the aerosol concentrations observed in the region may be due to the gas-to-particle convesion of precursor gases under the strong westerly winds. Also the generation of sea-salt particles under conditons of disturbed sea due to high winds during the weather fronts contribute for the increase in the 'large' aerosol concentrations. The observations made at the coastal stations are more difficult to interpret than those at the higher polar plateau because of greater intermixing of the effects of strong continental drainage flows due to sloped inversions and katabatic winds and the baroclinic disturbances generated over the southern ocean regions surrounding the Antarctica continent. The gas-to-particle conversion of trace gases and the breaking of waves on the surroudning ocean surfaces are the two possible sources of aerosol generation at Antarctica. As the Indian station Maitri is located on the bare land of Schimachar Oasis, there is also a third source of aerosol generation due to the exposed rocks. The total aerosols concentration observed at Maitri in our measurements is the typical of the Antarctica continent (Gras and Adriansen, 1985 and Ito, 1993). The processes responsible for these two modes are different. The smaller size particles in nucleation mode are mainly due to the nucleation of trace gases and they have only a few days of life time. Observations of Big (1980), Big et al. (1984), Cadle et al. (1968) and others show that about 95% of condensation nuclei are sulphate particles, mostly in the form of droplets of sulphuric acid. The stong seasonal sulphate particles, mostly in the form of droplets of sulphuric acid. The strong seasonal variation of such particles observed by Hogan (1975) and Shaw (1979) and Ito (1985) may directly relate them to sunlight intensity which indicates to their production by the photochemically

dervied gas-to-particle conversion. The production of these particles by the gas-to-particle conversion processes is also supported by the fact that excess or non-sea-salt sulfur is found all over the ice sheet at coastal and interior locations and is present mostly as Aitken particles which, as copared to accumulation mode particles, have much smaller residence time in the atmosphere.

The process of coagulation of small particles to large particles contributes to the creation of the accumulation mode aerosol particles. However, contribution of coagulation process is small around Antarctica since it varies with the square of the particle concentration which is small. Other possible sources for large particles on Antarctica may be the weathering products from Earth's curst, sea-salt particles from bubble breaking at the surrounding sea surface, extraterrestrial debris, and volcanic emissions (Shaw, 1988).

The peaks in the total aerosol concentration before the approaching of a low pressure system as observed on few occasions in our measurements may be due to the subsidence of mid-tropospheric air when the radiative inversion weakens. Our observation that the aerosols during these episodes are very small particles in nuclealion mode, supports such transport of aerosols. Also, greater vertical mixing associated with weakening of temperature inversion and moderate breeze may enhance the lifting of very fine crustal particles from the bare land area around Maitri.

Acknowledgements

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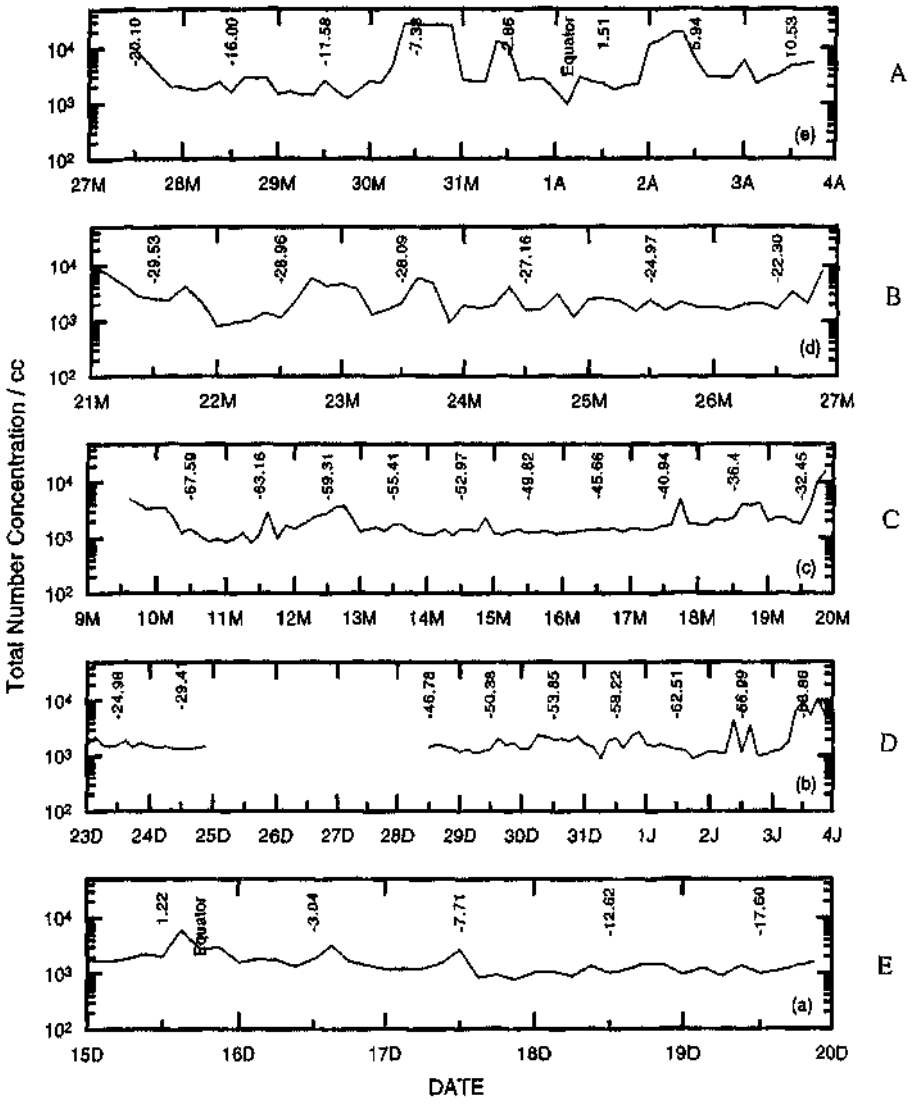


Fig. 1: The total aerosol concentration along the onward and return cruise traks (a) Goa to Mauritius, (b) Mauritius to Antrarctica, (c) Antractica to Durban, (d) Durban Mauritius (e) Mauritius to Goa

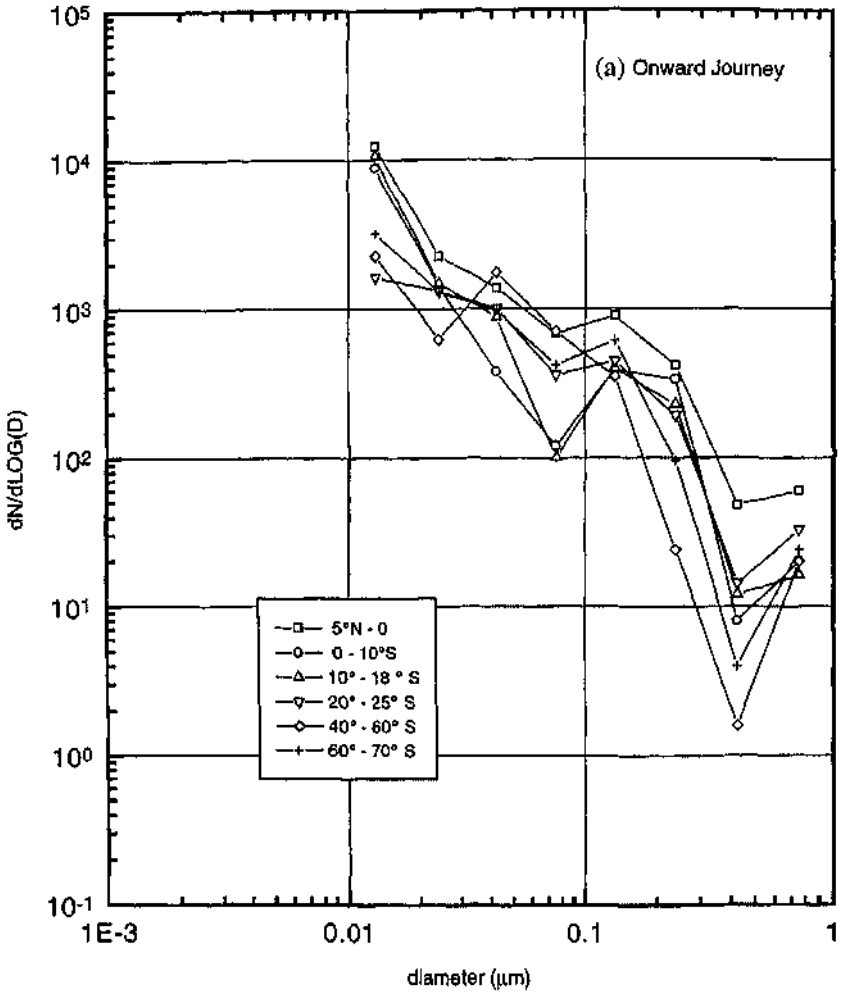
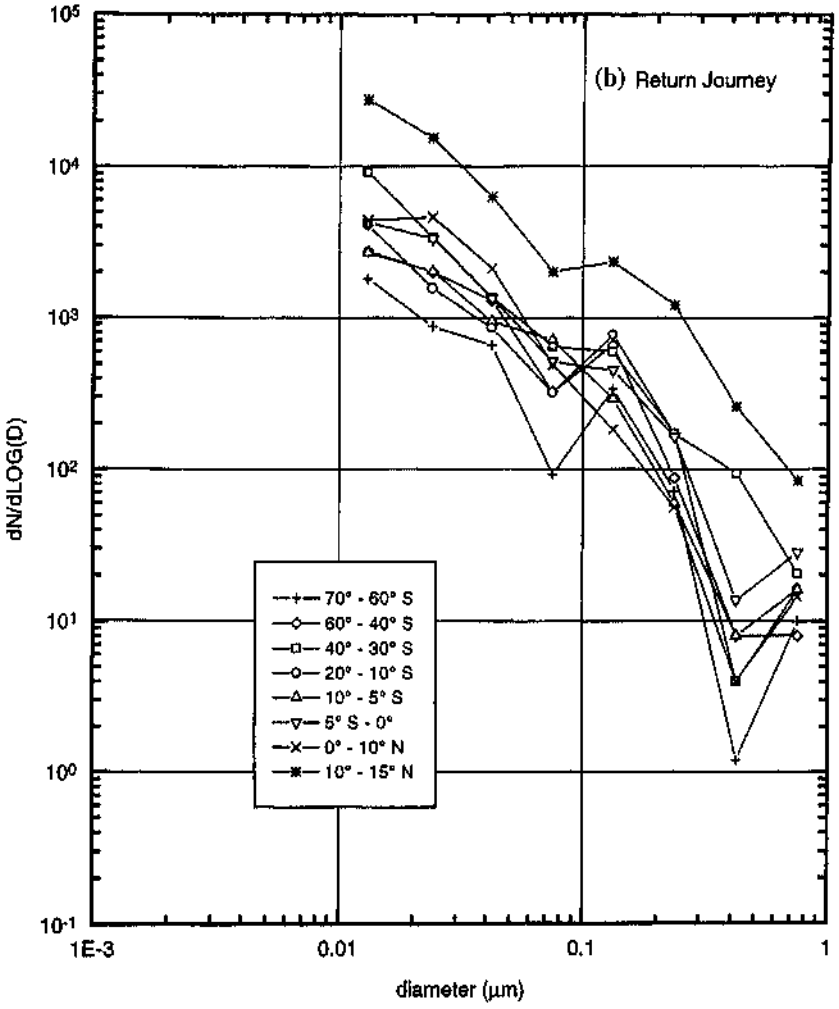


Fig. 2 : Typical aerosol size-distribution curves in different latitudinal belts on the (a) onwards, and (b) return cruises.


(b) Return Journey
dN/dLOG(D)
diameter (μm)
1E-3
0.01
0.1
1
70° - 60° S
60° - 40° S
40° - 30° S
20° - 10° S
10° - 5° S
5° S - 0°
0° - 10° N
10° - 15° N

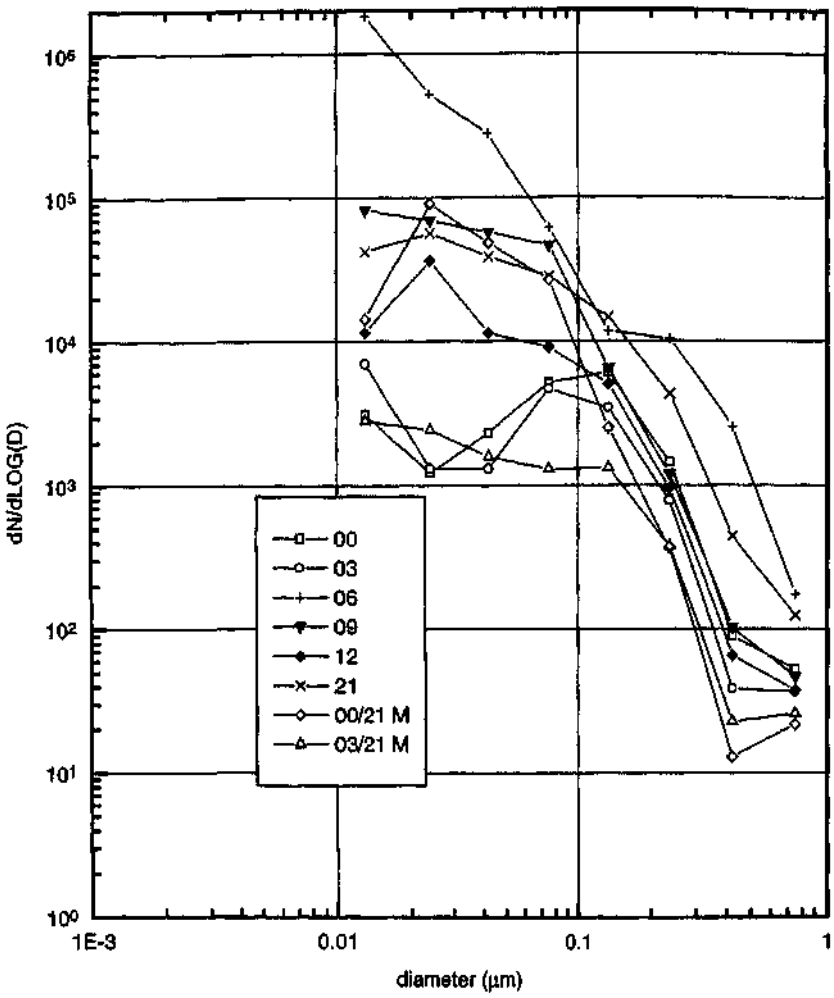


Fig. 3 : The aerosol size-distribution curves while approaching and departing from Durban.

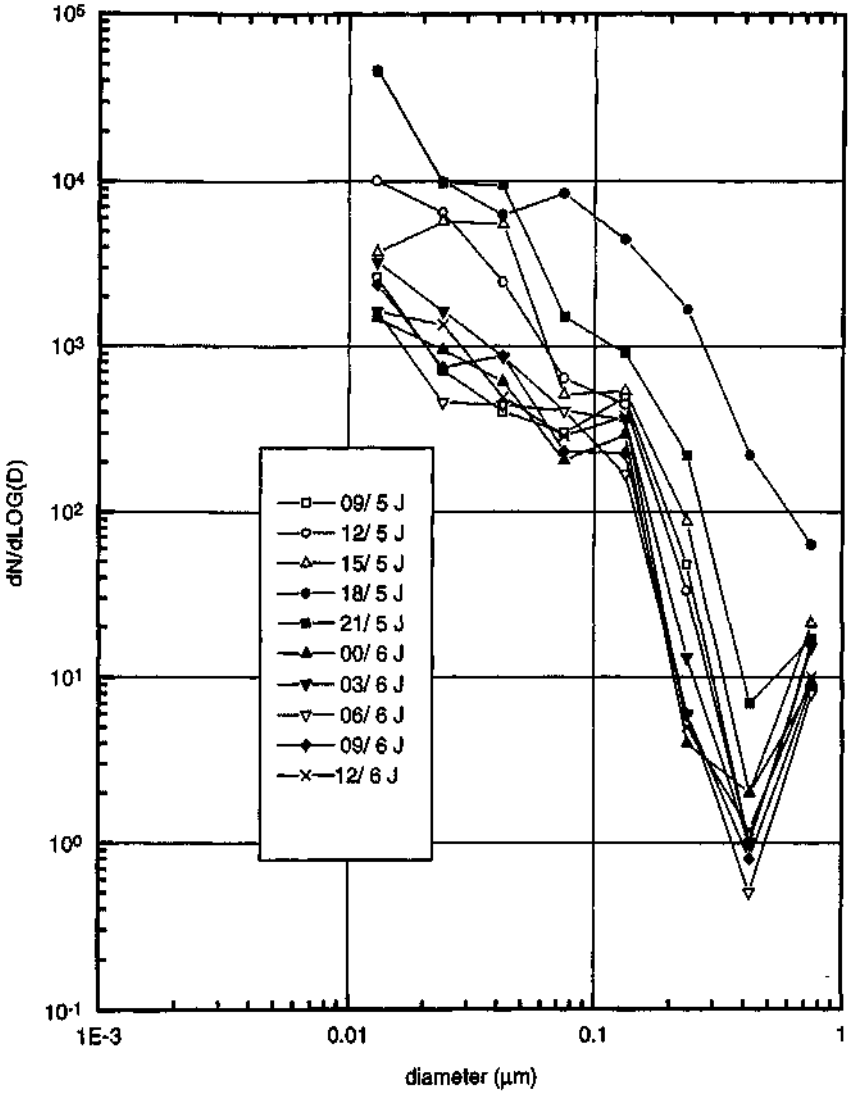


Fig. 4 : The aerosol size-distribution curves before, during and after the fog.

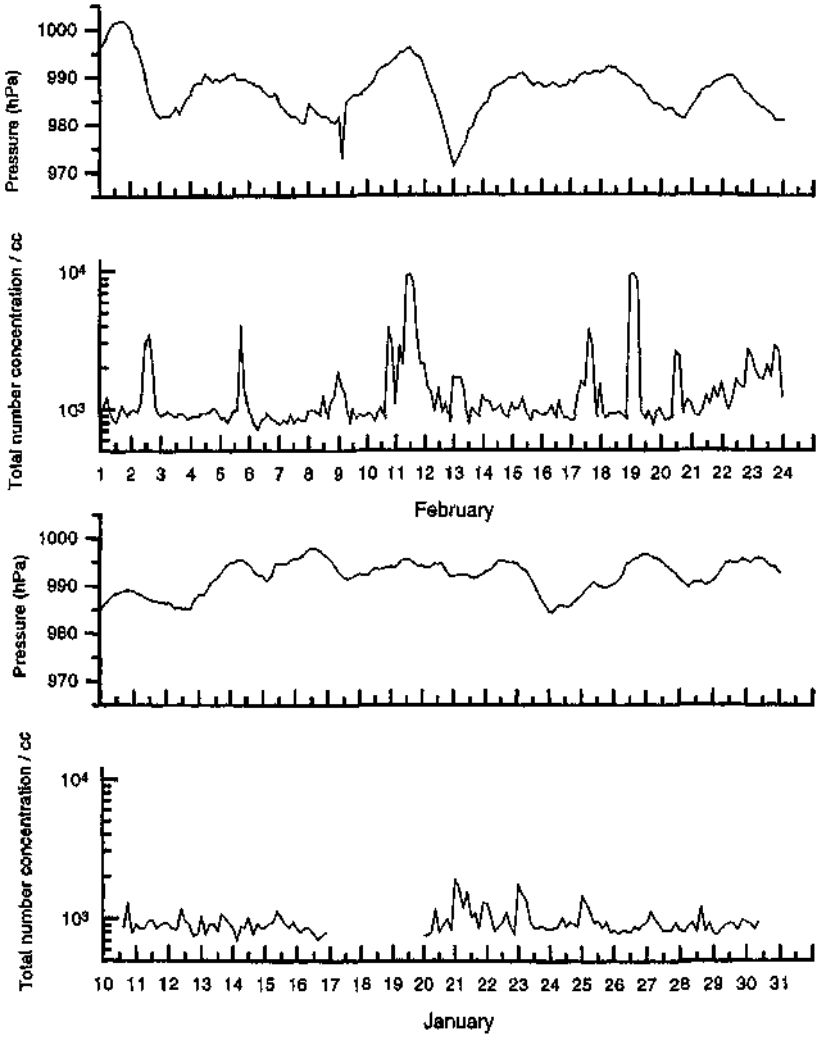


Fig. 5 : The variation of the total aerosol concentration and suface atmospheric pressure at Maitri from January 10 to February 24, 1997,

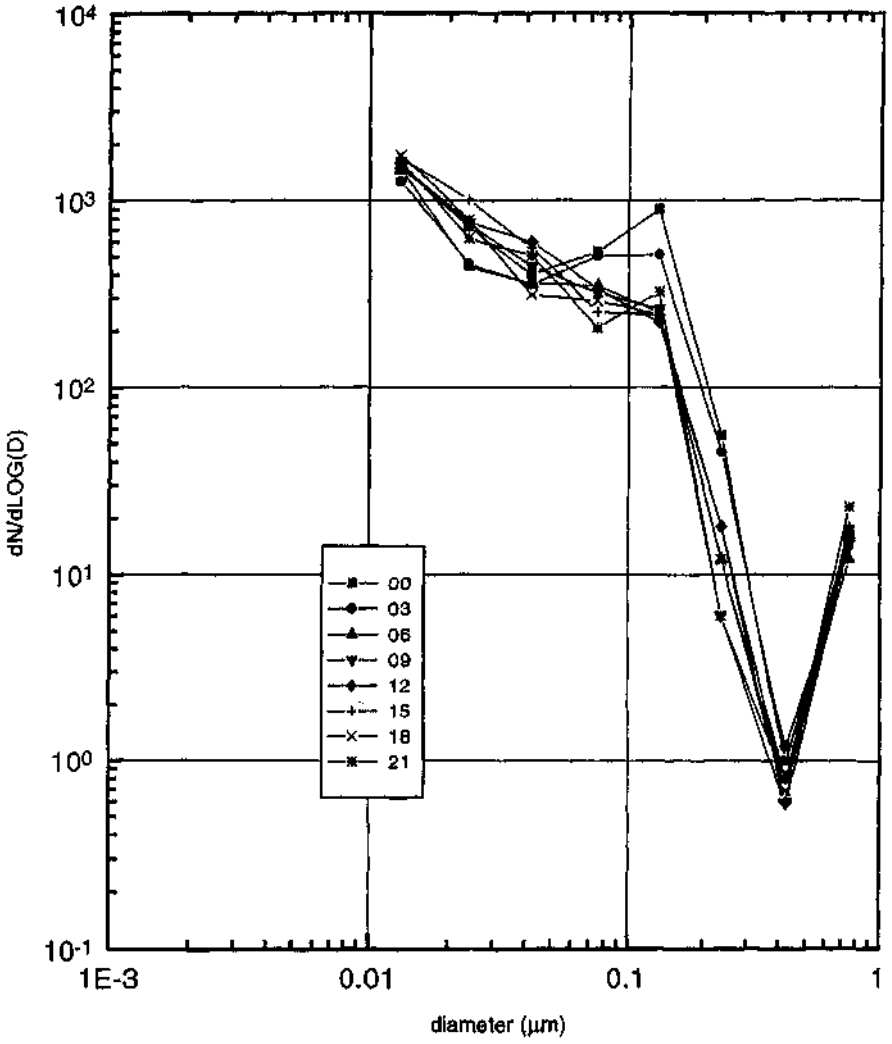


Fig. 6 : The aerosol size-distribution curves at Maitri on February 6, 1997,

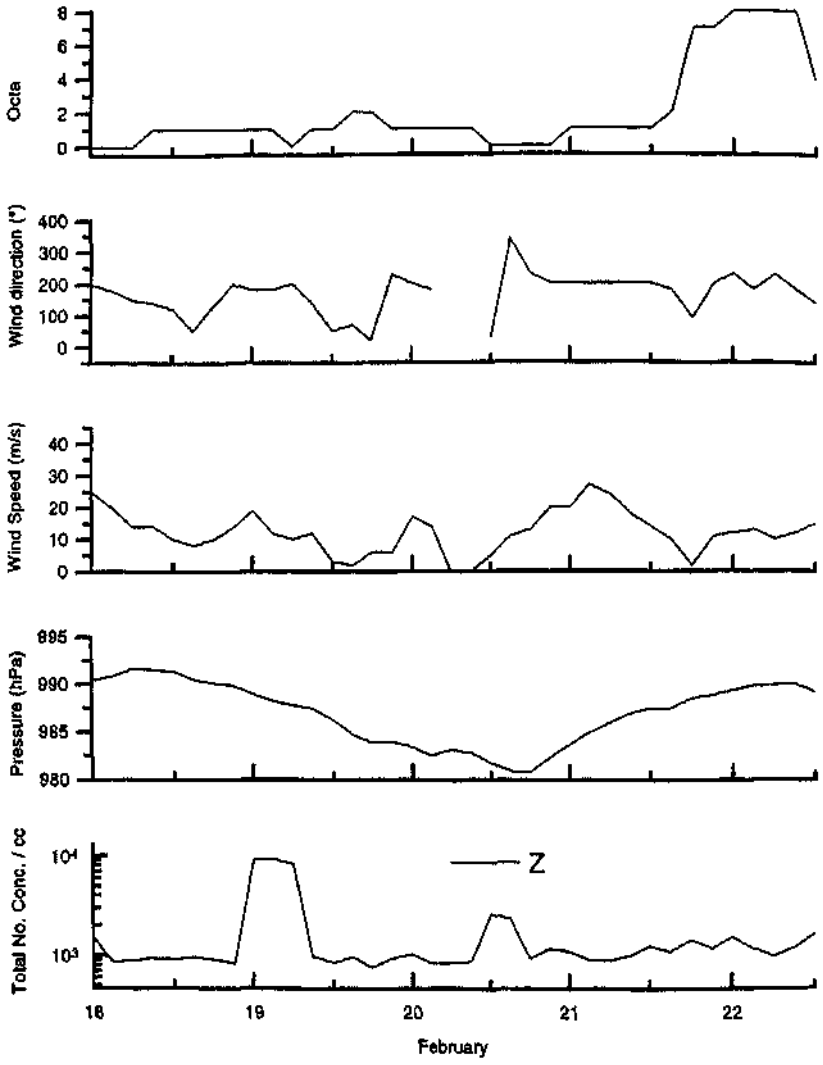


Fig. 7: The total aerosol concentration alongwith the surface atmospheric pressure, wind speed, wind direction and cloud coverage during the passage of a low pressure system over Maitri.

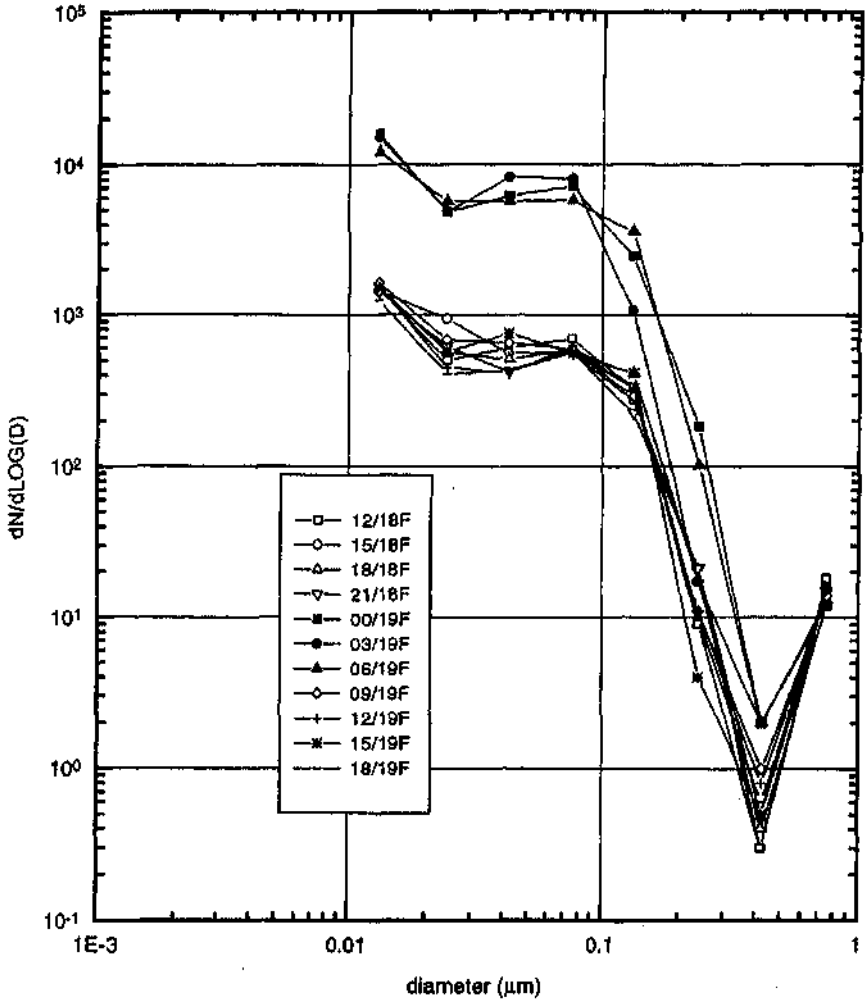


Fig 8. : The aerosol size - distribution curves before, during and after the aerosol peak associated with a low pressure system.