Size-distribution of submicron aerosol particles over the Indian Ocean during IFP-99 of INDOEX

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Measurements of the size-distribution of submicron aerosol particles of diameter from 0.003 to 1 μm are made over the Indian Ocean during the IFP-99 of the Indian Ocean Experiment (INDOEX). Measurements are made during the onward journey from Goa to Port Louis, Mauritius from 20 January to 11 February 1999 onboard ORV *Sagar Kanya* and during the return journey from Port Louis to Male from 22 February to 1 March 1999 onboard *Ronald H. Brown*. Observations show large concentrations in the range of 2–6 × 10^3 particles/cm^3 over the Indian Ocean in the northern hemisphere and these drop down to about 500 particles/cm^3 in the southern hemisphere. However, the aerosol concentrations show a peak of about 3 × 10^3 particles/cm^3 at 13°S. In the northern hemisphere, the concentration of particles of diameter < 0.0749 μm increases from 14°N to 1°N and then steeply falls. On the other hand, the concentration of particles of diameter > 0.0749 μm keeps decreasing up to 6°S. Size distributions of particles at open sea in the northern hemisphere show a maximum at 0.133 μm and minimum at 0.0422 μm and are generally open-ended at the smaller size end. The size-distributions of particles are sometimes relatively flat from 0.0133 to 0.237 μm when the particle concentrations are low in the southern hemisphere. The transport and accumulation of aerosol particles in the Inter-Tropical Convergence Zone is discussed. The relative abundance of large versus small particles is examined with respect to the variation of surface atmospheric pressure along the route.

SEVERAL experiments have been conducted over the Pacific and Atlantic Oceans to study the nature and physical, chemical and optical properties of the aerosols and trace gases[1–5]. The nucleation and transport characteristics of these aerosols and their effects on the radiation budget of the atmosphere have been theoretically modelled[6–9]. However, such data on the aerosols and trace gases over the Indian Ocean are scarce and only a few studies have been made so far, to study their dynamics[10–14].

The atmospheric boundary layer over the northern Indian Ocean is mostly dominated by the northeasterly surface winds during the winter months. These prevailing winds in the region transport the natural and anthropogenic aerosols and trace gases of continental origin to the atmosphere over the Indian Ocean. This polluted air approaching from the northern hemisphere meets the pristine air of the southern hemisphere in the Inter-Tropical Convergence Zone (ITCZ) which is marked by the deep convection. To assess the extent of such land-to-ocean transfer of the atmospheric aerosols is one of the objectives of the Indian Ocean Experiment (INDOEX)[15].

The atmospheric aerosols may be of natural origin such as the wind-blown mineral dust or of anthropogenic origin such as from industry, automobiles or other human-activities in the urban areas. They may also originate from gas phase reactions of low volatile vapours in the atmosphere. Their physical, chemical and optical properties differ and have impact on the radiation budget of the atmosphere. We report here our measurements of the concentration and size-distribution of sub-micron aerosol particles made during the onward journey from Goa to Mauritius from 20 January to 11 February 1999 onboard ORV *Sagar Kanya* and during the journey from Mauritius to Male during 22 February to 1 March 1999 onboard *Ronald H. Brown* during the IFP-99 of the INDOEX. The cruise routes of the two vessels along with the surface winds observed on the ship are shown in Figure 1.

Instrumentation

The concentration and size-distribution of aerosols in the size range of 0.003 to 1 μm were measured with an Electrical Aerosol Analyser (EAA) system of TSI. The sampling tube is projected out of a wall of a cabin and its inlet was fixed on the balloon launching platform of *Sagar Kanya* at a height of 9 m above mean sea level. The EAA system is kept inside the cabin. Five aerosol size spectra were taken at an interval of every two hours throughout the cruise period. More frequent observations of spectra were taken to study some particular phenomenon. However, because of some condensation occurring in the sampling line during the rainy periods, observations could not be taken during such periods. Moreover, since the accuracy of measurements for the lowest 2 channels of the

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EAA system, i.e. for 0.003 and 0.007 μm diameter-ranges, is not sufficient due to the limitation of operating the instrument under the conditions where the rate of generation of photochemically generated aerosols is highly variable, their contributions are not considered in this analysis.

Results and discussion

From our data on the size distribution of aerosol particles, the average total number concentrations of aerosol particles for each day were calculated for the cruise period. Figure 2 shows the number, surface and volume concentrations of aerosols for the whole cruise period. Top of the bottom-plot shows the position of the ship at 0000 h on that day. One of the noteworthy features is the presence of high number concentrations in the range of 1–6×10^3 particles/cm^3 over the Indian Ocean from 14°N to the equator. Moreover, although there is no significant gradient in the number concentration from 14° to 2°N, the surface and volume concentrations do show a negative gradient which is observed to continue even up to 8°S. The lowest aerosol concentrations of only about 400 particles/cm^3 are observed at 13°S on the return journey. In contrast, however, aerosol concentrations show a maximum of 3×10^3 particles/cm^3 at the same latitude on the onward journey. Another maximum is observed in the surface and volume concentrations at 1°N on the return journey. From 13°S to 2°N the trends in concentration on both the onward and return journies were almost similar.

Although the total number concentration of aerosol particles remains almost constant from 14 to 2°N, the particles with diameter < 0.075 μm show an increasing trend and the particles with diameter > 0.075 μm show a decreasing trend towards the equator in this latitudinal belt (Figure 3). Again, while the maximum at 13°S on the onward journey is due to the increase in particle

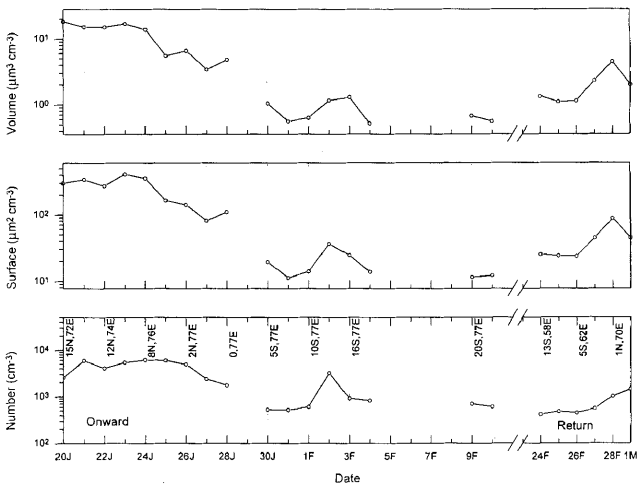

Figure 2. Latitudinal variation of the daily averaged values of the total number, surface and volume concentrations along the cruise route.

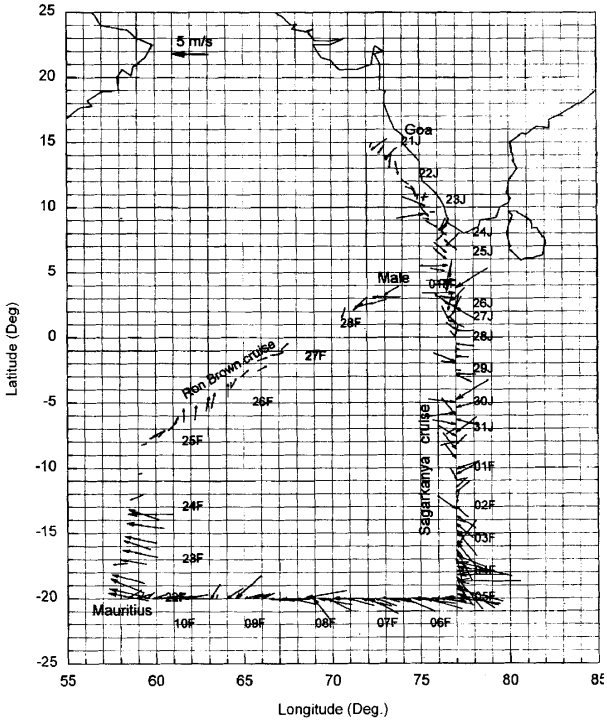

Figure 1. Cruise routes of *Sagar Kanya* from Goa to Mauritius and of *Ronald H. Brown* from Mauritius to Male. Arrows indicate the value of wind speed and direction observed on the ship.

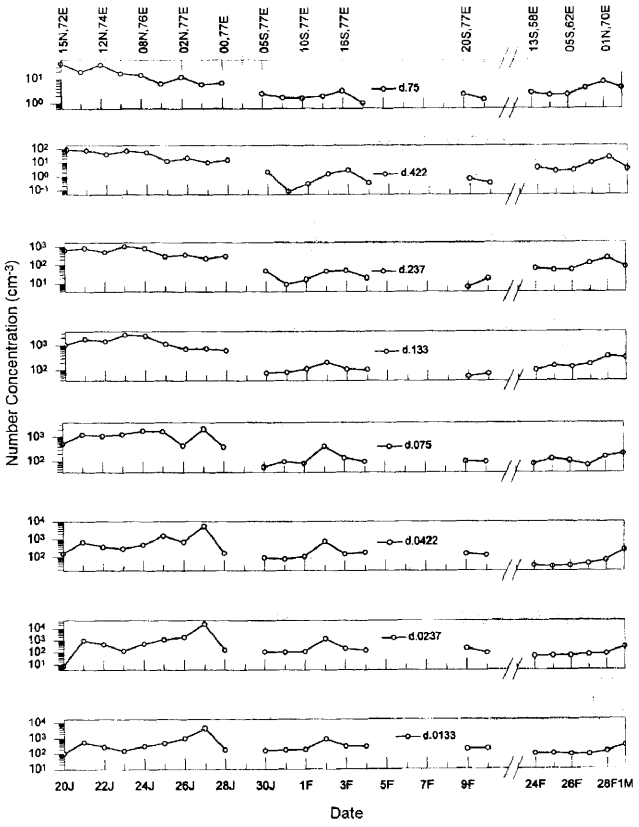

Figure 3. Latitudinal variation of the daily averaged values of the number concentrations of particles of different sizes along the cruise route.

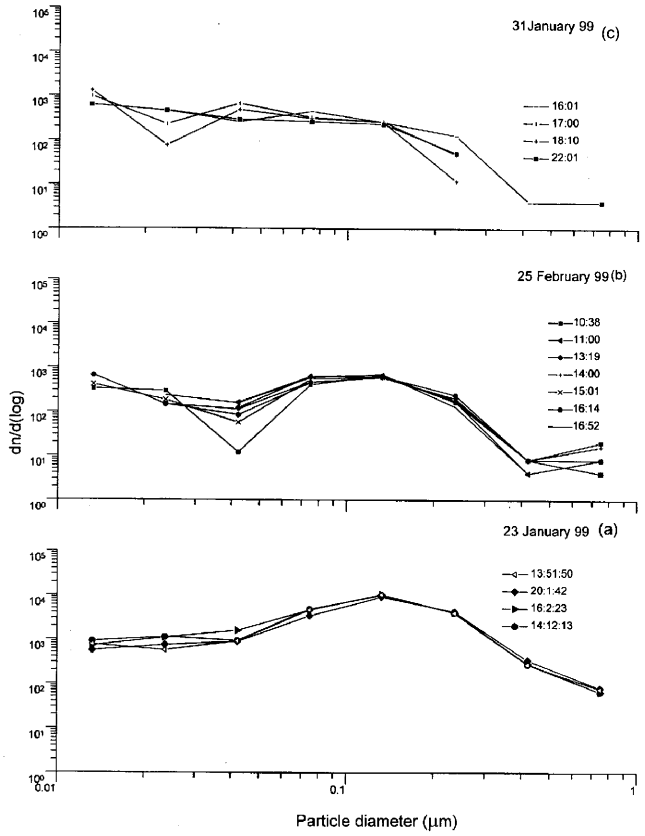

Figure 4. Particle size distributions of three types frequently observed during the cruise (***a***) monomodal, (***b***) bimodal, open distribution, (***c***) flat, low particle concentrations.

concentrations of all sizes in this size-range, the maximum at 1°N on the return journey is mainly because of the increase in particle concentrations of size > 0.133 μm.

During the cruise period, generally three types of the aerosol size distributions, as shown in Figure 4, were observed. The first type was observed near the coastline and was similar to the distributions observed over continents. In these distributions the aerosol concentrations are of the order of $10^4/cm^3$ or higher and the size distribution is monomodal with the maxima at 0.133 μm. The second type was observed as one moves away from the coastline. Here, the concentration of large particles decreases and the size-distributions become bimodal with the maximum at 0.133 μm and minimum at 0.0422 μm and are open-ended at the small-size end. It indicates large concentrations or even a maximum in the concentration of ultrafine particles smaller than 0.0133 μm. Such a size distribution indicates relatively recently formed particles which had less time to grow by coagulation or vapour condensation. As suggested by Covert *et al.*[5], this type of distribution can be associated with either large-scale subsidence or post-frontal subsidence and advection of anticyclonic systems. However, in contrast to the observations[5] over the Pacific Ocean where the aerosol size distributions do not show the presence of ultrafine particles in tropics, the concentrations of ultrafine particles over the tropical Indian Ocean are comparatively very large. The third type was sometimes observed over open ocean when the aerosol concentrations remain lower than $10^3/cm^3$ or so. In

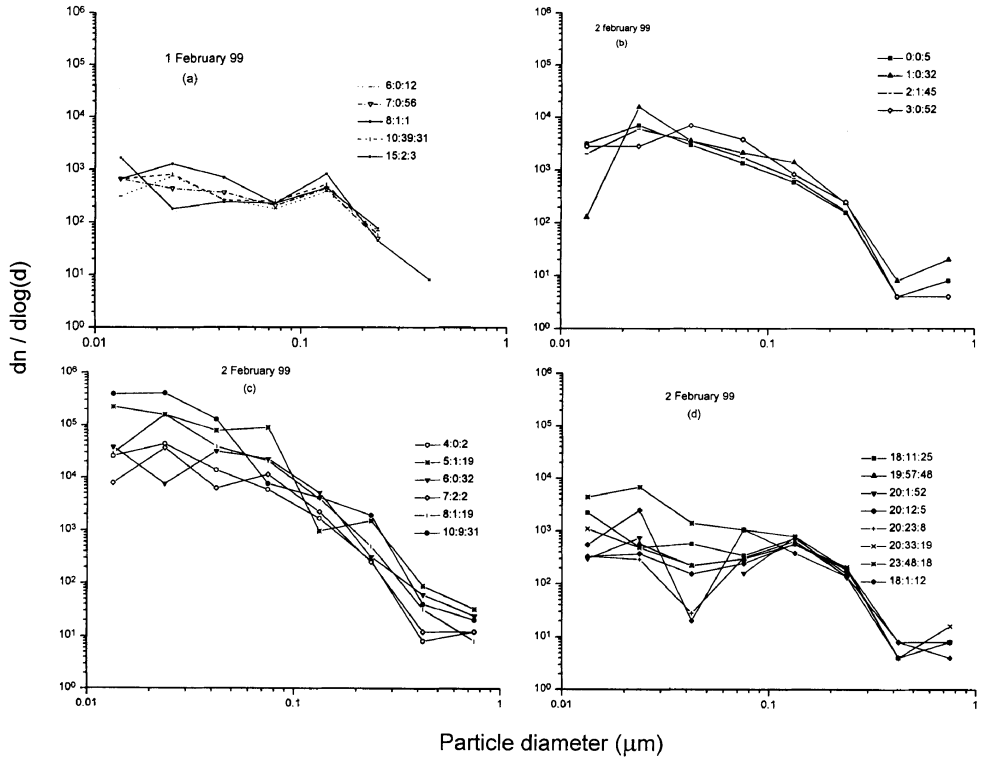

Figure 5. Changes in the particle size distributions associated with the maximum observed in the total particle concentration at 13°S on 2 February 1999 during the onward journey.

such distributions, the concentrations of large particles become very low and no pronounced maxima was seen. The size distributions mostly remain flat and the concentrations of particles of sizes between 0.0133 and 0.133 μm do not show any significant difference.

We have examined the size distributions prevailing before, during and after the maxima observed at 13°S and 1°N on 2 and 22 February 1999 respectively. The size distributions remain bimodal before and after the maxima and the height of the maxima is generally always below 10^3 particles/cm^3. The increase in particle concentrations during the maximum observed at 13°S on 2 February 1999 occurs mainly due to the increase in concentrations of smaller particle (Figure 5). The size distribution of particles, in this case, tends to change from bimodal to Junge's power-law type. On the other hand, the increase in particle concentrations during the maximum observed at 1°N on 28 February 1999 was due to the increase in comparatively larger particle concentrations and, in this case, the height of the maximum increases (Figure 6). The size distribution in this case, however, continues to be bimodal.

The positions of the aerosol concentration maxima observed at 13°S and 2°N in our measurements roughly lie at the southern and northern limits of the ITCZ during the onward and return journies of the cruise. Figure 7 shows a satellite image of the cloudiness associated with ITCZ on 28 February 1999 with the position of the ship marked on it.

We have calculated the value of $N = \mathrm{d}n/\mathrm{dlog}(0.0422) - \mathrm{d}n/\mathrm{dlog}(0.0237)$ from our measurements of the number concentrations as a parameter related to age of the aerosol. The concentration at 0.0422 μm was selected for comparison as it is relatively stable since it is less affected by rapid changes due to removal by the scavenging processes or by the production of new particles by nucleation or condensational growth. Figure 8 shows the variation of N and sea level pressure with latitude throughout our cruise. The negative values of N indicate a dominant ultrafine mode or nucleation mode with a mean diameter smaller than 0.0422 μm and thus had a relatively shorter aging time in the marine boundary layer. On the other hand, the positive values of N indicate that aerosol had a longer aging time and was isolated from sources of new

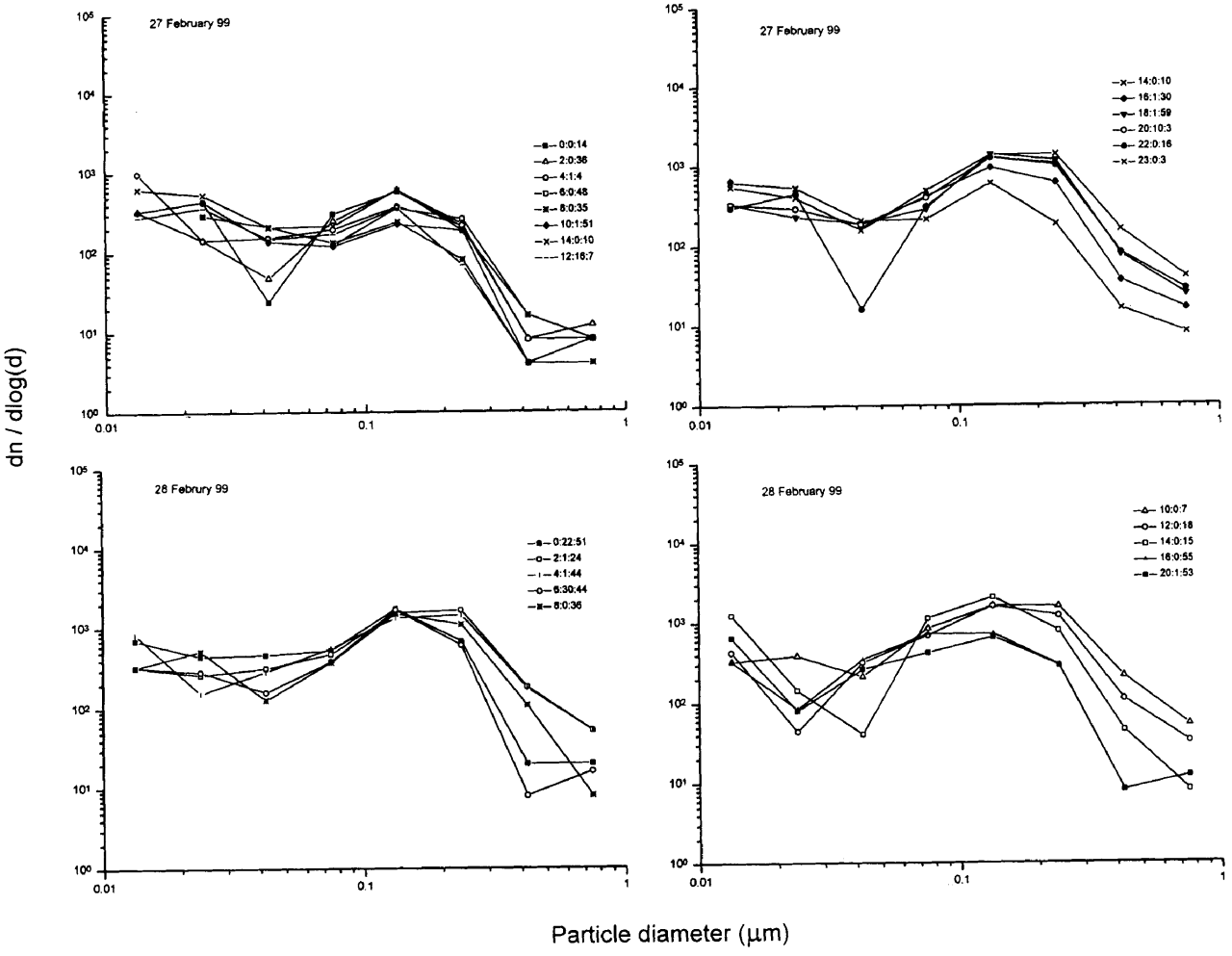

Figure 6. Changes in the particle size distributions associated with the maximum observed in the total particle concentration at 1°N on 28 February 1999 during the return journey.

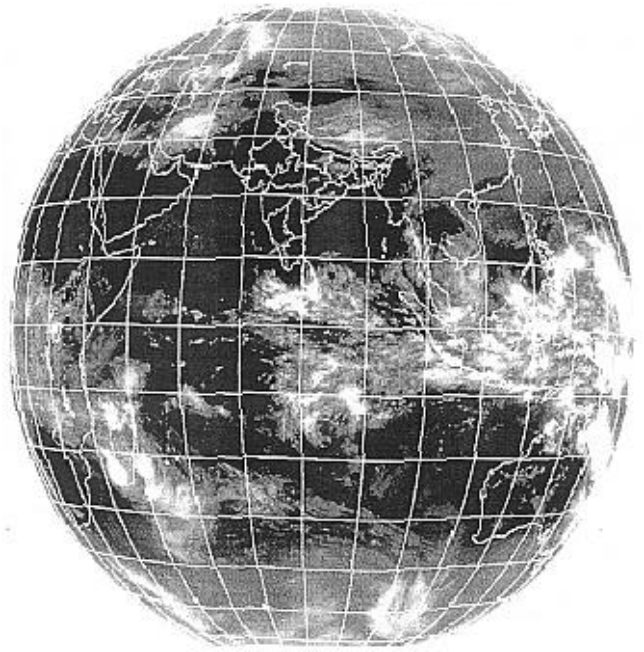

Figure 7. INSAT picture for 0000 h on 28 February 1999. Position of the ship shown by X.

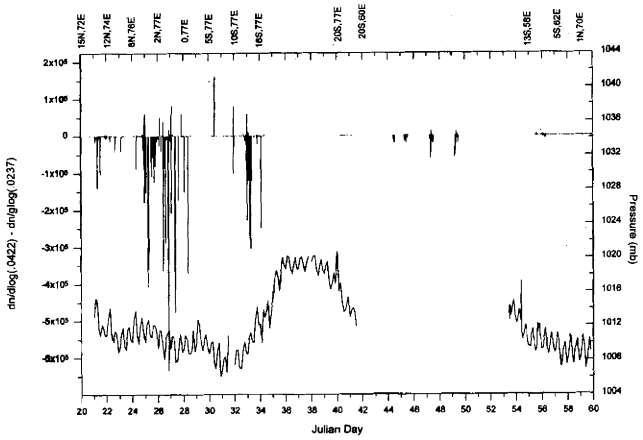

Figure 8. Latitudinal cross section plots of $N = dn/d\log(0.0422) - dn/d\log(0.0237)$, the difference in the particle concentration at 0.0422 and 0.0237 μm and the atmospheric sea level pressure.

particles. Figure 8 shows extensive latitudinal bands in which N is near zero or predominantly negative and occasionally positive. N is near zero or negative from 15°N to 7°N and near 20°S but predominantly negative and only occasionally positive from 7°N to the equator and from 10°S to 16°S. In contrast to the onward journey, positive or negative values of N were not observed on the return journey and N was mostly near zero from 17°S to 2°N. This result is in contrast to the observations of Covert *et al.*[5] in the Pacific Ocean where they report that N is predominantly positive in the tropics. So, small particles predominate in the high values of the total aerosol concentrations observed at these latitudes over the Indian Ocean in this season.

Conclusions

Large concentrations of submicron aerosols observed over the northern Indian Ocean in our measurements in winter months indicate the transport of large quantities of aerosols from the continents to the oceanic atmosphere with the prevailing northeasterly winds in this region. As the aerosols are being advected equatorwards, larger particles keep settling with the passage of time and smaller particles seem to be generated or advected to this region. The size distribution of particles observed over northern Indian Ocean indicates very large concentrations of ultrafine particles that are probably formed locally or have a short age. Polluted air approaching from the northern hemisphere meets the pristine air of the southern hemisphere in the ITCZ. The two maxima in aerosol concentration observed at the southern and northern limits of the ITCZ indicate the formation of some pockets of high aerosol concentrations in the ITCZ. Formation of such pockets of high aerosol concentration is likely to be associated with the eddy structure of airflow in the ITCZ.

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