

Results on direct radiative forcing of aerosols obtained over the tropical Indian Ocean

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Aerosols reduce the surface reaching solar flux (negative radiative forcing) by scattering the incoming solar radiation out to space. Various model studies on climate change suggest that surface cooling induced by aerosol scattering is the largest source of uncertainty in predicting the future climate. Measurements made by *ORV Sagar Kanya* over the tropical Indian Ocean, as part of the pre-INDOEX campaign, in January to March of 1996, 1997 and 1998 consistently present aerosol characteristics with large spatial and temporal variations. The columnar aerosol optical depths measured at different spectral bands using a sun-photometer reveal a systematic latitude gradient in aerosol amount decreasing from north to south, until the Inter-Tropical Convergence Zone (ITCZ). Independent surface level aerosol size distribution measurement complement the optical depth data. The submicron size particles of radius less than $0.5 \mu\text{m}$ are responsible for the high aerosol optical depths obtained north of the ITCZ. Surface reaching direct

solar and global (solar plus sky) radiation intensities are measured using standard pyrheliometers, pyranometers and photodiode radiometers. Data on aerosol optical depth and radiation intensities available simultaneously on cloud-free days are correlated to estimate the direct radiative forcing by aerosols. Combining the 1996 and 1997 cruise data, the diurnal averaged radiative forcing for the tropical Indian Ocean is obtained as -5.2 W m^{-2} , which is the reduction in the global flux for every 0.1 increase in aerosol optical depth. This is the first ever direct experimental evidence on the aerosol radiative forcing which applies to the aerosols found over the tropical Indian Ocean during January to March. This value is higher than the computed global average value, which is about -2 W m^{-2} . The excess forcing could only be explained by an excess absorption by aerosols or by other atmospheric constituents which are hitherto unaccounted for in the model calculations.

AEROSOLS are derived from many different sources and are distributed in the atmosphere through turbulent mixing and transport of air masses. Their residence time in the atmosphere differs by type, but mainly depends on the size of the particles and altitude at which they are present. Along with other constituents of the atmosphere, such as molecular gases and clouds, aerosols determine what fraction of the solar radiation incident at the top of the atmosphere reaches the earth's surface and what fraction of the thermal radiation emitted from the earth escapes to space. These two processes essentially determine the earth's climate. Though the short wave radiative forcing caused by aerosols is much less (about 1 to 2%) than that caused by cloud cover, both are not negligible, both kinds of forcing are also almost equal (but opposite in sign) to the longwave radiative forcing caused by an increase in several radiatively active 'greenhouse' gases^{1,2}. The anthropogenic component of the aerosols is estimated to have a direct negative radiative forcing (resulting in cooling) of about 2 W m^{-2} in the shortwave. Most of the anthropogenic aerosols are sulfate or smoke particles which reside in the troposphere for about a week, contrary to greenhouse gases which last from decades to a century. Thus the greenhouse warming balanced by the aerosol-

induced cooling is not uniform around the globe but concentrated in packets where these aerosols are produced in maximum. These regional variations in the chemical and physical properties, consequently in the radiative properties of aerosols, make it difficult to assess aerosol radiative forcing on a global scale³.

One of the important objectives of the Indian Ocean Experiment (INDOEX) is to assess the climate forcing caused by both the natural and anthropogenic aerosols. INDOEX focuses its study on the tropical Indian Ocean, where both natural and anthropogenic aerosols are transported from the surrounding continents. Currently, we know very little about these aerosols and their radiative forcing effects on the surface reaching solar flux. The ideal time to examine this aerosol radiation interaction problem is January to March; then the low level winds are predominantly from the north-east direction, favourable for transporting aerosols and precursor gases from continents to the pristine ocean surface. A series of pre-INDOEX cruises on ships-of-opportunity were conducted before the main field phase campaign⁴ which is to occur in early 1999. The first of these was on the *RV Malcom Balridge* cruise conducted in March through April 1995. Ozone, trace gases and aerosols measurements exhibited large temporal and spatial variations which were explained in terms of changes in the observed air mass characteristics⁵.

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The present work focuses on the results obtained from the three pre-INDOEX cruises made in 1996, 1997 and 1998 by the *ORV Sagar Kanya* over the Arabian Sea and the tropical Indian Ocean. Table 1 gives the cruise period and the range of latitude and longitude covered in each cruise. The list of measurements and the equipment relevant to this paper's results appears in Table 2. The same instruments were used in all three cruises to avoid any instrumental bias in the measured values.

Experiment

The aerosol optical depth is measured from the ship deck using a hand-held filter sun-photometer operated in 5 different spectral bands (Table 2). Developed in-house the photometer has a baffle, interference filter assembly, peak detector electronics and a sun-pointer. The hand-held sun-photometer is preferable to an automatic sun-tracking assembly because of operational convenience. The interference filters used in the experiment are regularly calibrated in the laboratory before and after each cruise for spectral transmission characteristics; the results are also

used to calculate Rayleigh scattering and absorption due to ozone for correcting the measured total optical depth. The sun-photometer is also calibrated regularly, using the standard Langley plot technique, at Mt. Abu, a hill station about 1680 m above mean sea level. The instrument constant for each wavelength band obtained from the calibration experiment helps derive the optical depth from the radiation intensity measured on-board the ship. Optical depth measurements were made with a typical time interval of 15 to 30 min on all clear days along the ship cruise track. The derived aerosol optical depth is uncertain because of measurement error and the use of model values for columnar air and ozone concentrations for correcting the total optical depth. The latter possesses only an error of less than 5%. However, the relatively large field-of-view of the photometer, which is 8° in the present case, enables measurement of the forward scattered radiation along with the transmitted direct radiation. The forward scattering effect is greater in the short wavelengths than compared to long wavelengths. Approximately, the error introduced in the derived aerosol optical depth from neglecting the forward scattering effect is

Table 1. Details of pre-INDOEX cruises made by *ORV Sagar Kanya*

Cruise no.	Period	Latitude covered	Longitude covered
cr109	5 January–4 February 1996	18°N to 5°S	60° to 76°E
cr120	27 December 1996–1 February 1997	18°N to 15°S	71° to 79°E
cr132	17 February–30 March 1998	18°N to 21°S	59° to 74°E

Table 2. Observations made on board *ORV Sagar Kanya* during the three pre-INDOEX cruises

Parameters measured	Instrument employed	Institute
Aerosol optical depth 399 nm (19 nm FWHM) 497 nm (13 nm FWHM) 667 nm (13 nm FWHM) 848 nm (14 nm FWHM) 1051 nm (25 nm FWHM)	Hand-held sun-photometer (developed in-house)	Physical Research Laboratory, Ahmedabad
Aerosol mass concentration and size distribution in 10 size ranges from 0.05 to 25 µm diameter	Quartz crystal microbalance (QCM) cascade impactor, California Meas. Inc., USA	Physical Research Laboratory, Ahmedabad
Direct solar flux, W m ⁻² Broadband (280–2800 nm) Near IR (780–2800 nm)	Pyrheliometer Eppley Laboratory Inc., USA	Physical Research Laboratory, Ahmedabad
Global flux, W m ⁻² Broadband (280–2800 nm) Near IR (780–2800 nm)	Pyranometers (2 nos.) Eppley Laboratory Inc., USA	Center for Clouds, Chemistry and Climate, Scripps Institute of Oceanography, San Diego, USA
Spectral global flux, W m ⁻² 305 nm (10 nm FWHM) 320 nm (FWHM) 380 nm (FWHM) PAR (400–700 nm)	Photodiode Radiometer Biospherical Institute Inc., USA	

about 8% in the case of 399 nm and less than 4% for the 1051 nm. More details on the calibration and data reduction appear in Jayaraman *et al.*⁶

A quartz crystal microbalance (QCM) cascade impactor system measured the aerosol mass concentration ($\mu\text{g m}^{-3}$) and aerosol size distribution. Air sample was drawn at the rate of 240 ml/min and the particles are segregated into 10 size bins with 50% efficiency cut off diameter at 25, 12.5, 6.4, 3.2, 1.6, 0.8, 0.4, 0.2, 0.1 and 0.05 μm . A pair of sampling and reference quartz crystal wafers were used in each stage to provide real time mass data for the sampling period, which is typically 10 min. The impactor and other trace gases monitoring equipment in the ship drew air from a plenum to ensure that all equipment sample the same air. Air reached the plenum through a duct at a reasonably high flow rate collected from about 11 m above the sea level. About 4 to 6 measurements were made every day along the cruise track. Measurements were avoided when the ship was docked, for example at Male Port and Port Luis during the last cruise (cr132), to avoid contamination from ship exhaust. In the absence of quoted uncertainty by the manufacturer, we estimated the maximum uncertainty in the mass concentration measurement by operating simultaneously with an Anderson Impactor System, as 25% for all 10 stages.

Direct solar radiation intensities were measured on board using a pyrheliometer, pointed manually towards the Sun. Measurements were made in two different wavelength bands, viz. 280–2800 nm and 780–2800 nm, selected using a filter wheel. The difference between the two measurements is referred to as the 'visible' radiation (280–780 nm). For global (direct solar plus scattered sky) radiation measurements, two different instruments were used: a multichannel radiometer measuring radiation intensities in 4 spectral bands (Table 2). A pair of pyranometers, each measuring the broadband flux

(280–2800 nm) and the near-IR (780–2800 nm) flux. Data corresponding to PAR channel (400–700 nm) are only used in the present work. All the radiation flux measurements have an uncertainty of less than 3%.

Results and discussions

Figure 1 shows the distribution of aerosol optical depth (AOD) as a function of latitude obtained from the three cruises. For clarity, only results pertaining to 497 nm are shown. A large hemispherical difference is seen in AOD, with very high values north of the equator, supporting the argument that large amount of aerosols are transported from the landmass surrounding the Arabian Sea. In addition, AOD measured at a shorter wavelength region (399 nm) shows a larger variation (not shown in the figure) with values ranging anywhere from 0.02 in the pristine region to about 0.8 within the turbid northern region. In contrast, AOD measured at 1050 nm does not show any appreciable variation with latitude, and the values are found to be 0.2 and less. It is known that smaller particles of submicron size contribute more to the AOD at lower wavelengths compared to at higher wavelengths. Figure 2 shows sample data on aerosol optical depth spectrum selected for different latitudes. Generally, the AOD spectrum does not show appreciable wavelength dependence in the pristine air condition, and the optical depth values are below about 0.1 at all wavelengths. In contrast, in the Arabian Sea and near the coastal region, a steep increase in AOD is observed particularly at lower wavelengths.

The aerosol particles from the continents have different origins and hence can have different chemical compositions. The most common particles are mineral dust, carbonaceous particles and sulphates. Because of their varying chemical compositions, their refractive index differ and therefore, they display different scattering and absorption efficiencies at various wavelengths. Though it

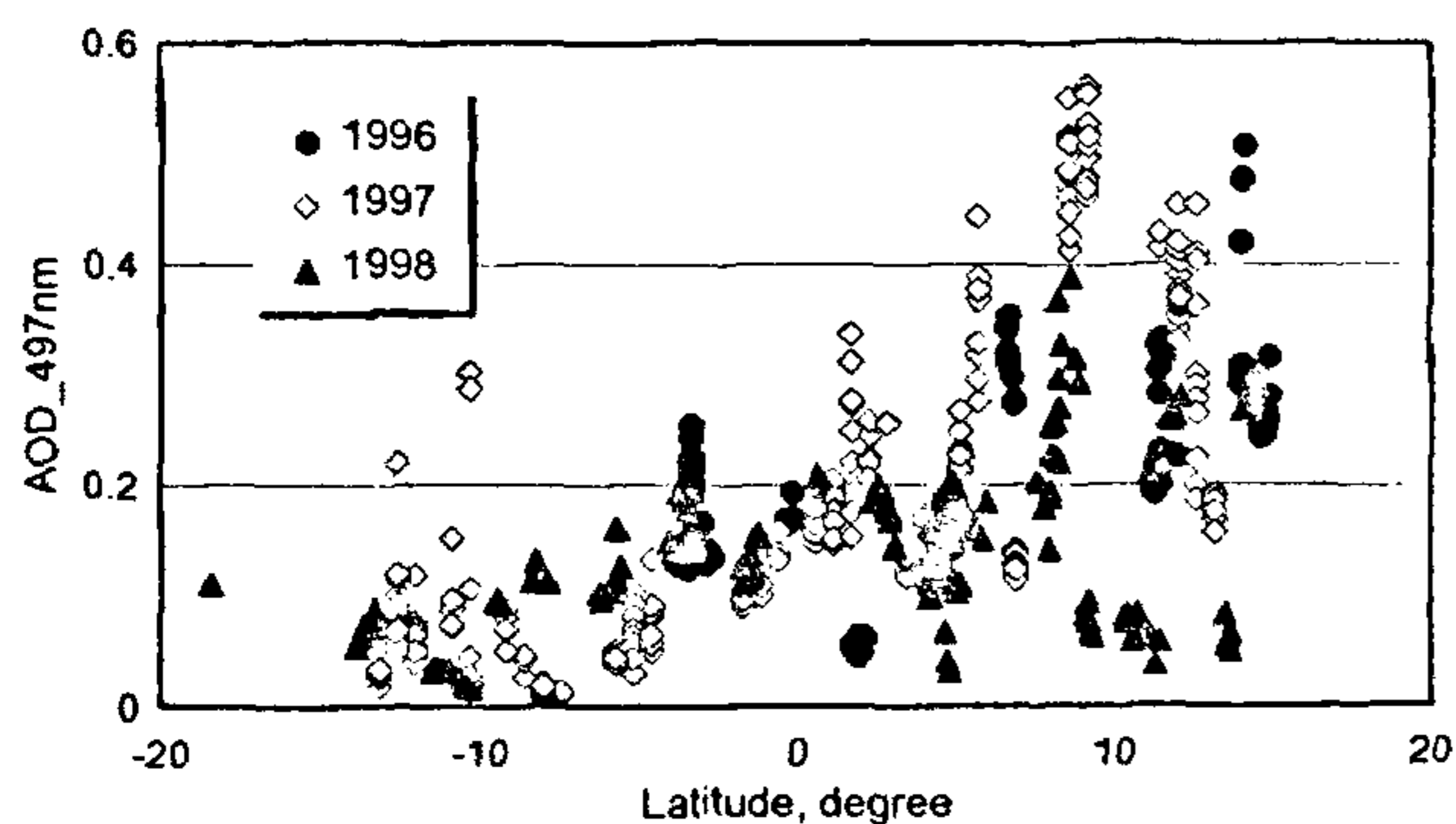


Figure 1. Columnar aerosol optical depth (AOD) as a function of latitude obtained from the three cruises conducted in January to March in 1996, 1997 and 1998. For clarity, only results pertaining to 497 nm channel are shown. Large hemispherical difference is seen in AOD, with very high values in the north of the equator.

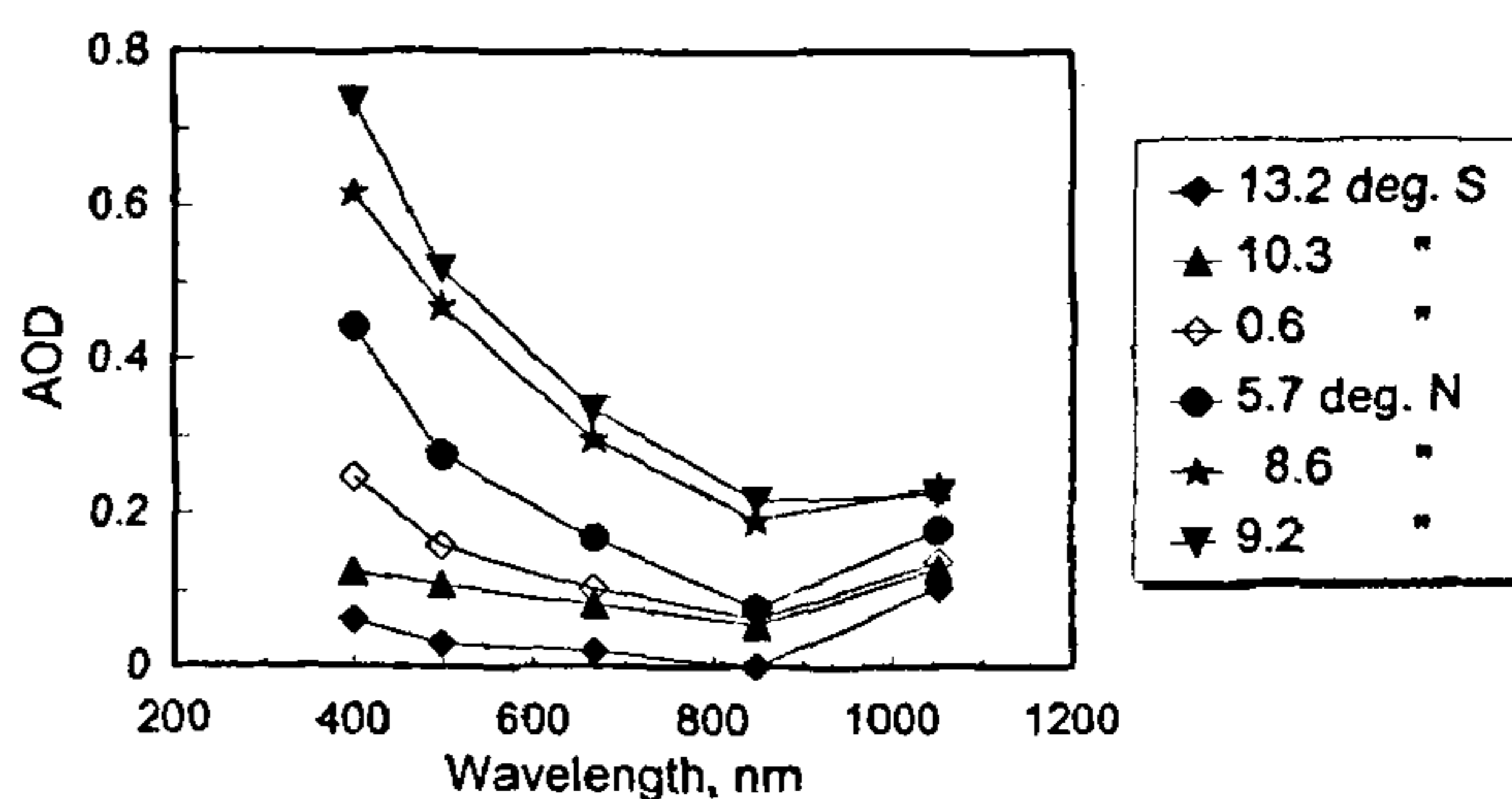


Figure 2. Sample data on aerosol optical depth spectrum measured at different latitudes. The AOD spectrum does not show appreciable wavelength dependence in the pristine atmospheric condition south of the equator.

is difficult to pinpoint the individual contribution from these different aerosol types to the measured AOD, the larger variation seen in the AOD at the shorter wavelengths indicates that submicron size particles, in the size range 0.1–0.4 micron radius, are responsible for the observed large values of AOD in the Arabian Sea. Surface level wind flow patterns play a dominant role in transporting these particles from the continents and are responsible for the observed spatial variation within the region. Krishnamurti *et al.*⁷ have linked the observed spatial variation in the measured AOD from one of the cruises (SK109) to the winter monsoonal flow using a high resolution global reanalysis. Evidently, the low level wind flow can transport aerosols within about 6 to 7 days from the landmass to the Inter-Tropical Convergence Zone (ITCZ) where the air from the northern hemisphere meets the relatively pristine air from the southern hemisphere.

Figure 3 shows the measured aerosol mass concentrations in the study area. Values corresponding to only the submicron particles (sum of the mass values obtained from the last five channels of the QCM, corresponding to particle diameter less than 0.8 μm) are only shown in the figure. The submicron particles are only transported to long distances compared to coarse particles (greater than 1 μm diameter) which are effectively removed by gravitational settling near to their source region. A systematic latitude gradient in the concentration occurs from 15°S to about 22°N, similar to the behaviour observed in the AOD. Also a systematic decrease in the total (sum of all size ranges) mass is seen (not shown in the figure) as the ship sailed away from the coast towards the interior ocean region. The total mass concentration was in the range of 40 to 80 $\mu\text{g m}^{-3}$ near coastal India, 20 to 40 $\mu\text{g m}^{-3}$ over the Arabian Sea and less than 20 $\mu\text{g m}^{-3}$ over the pristine tropical Indian Ocean. The aerosol number density distribution derived from the mass concentration obtained at

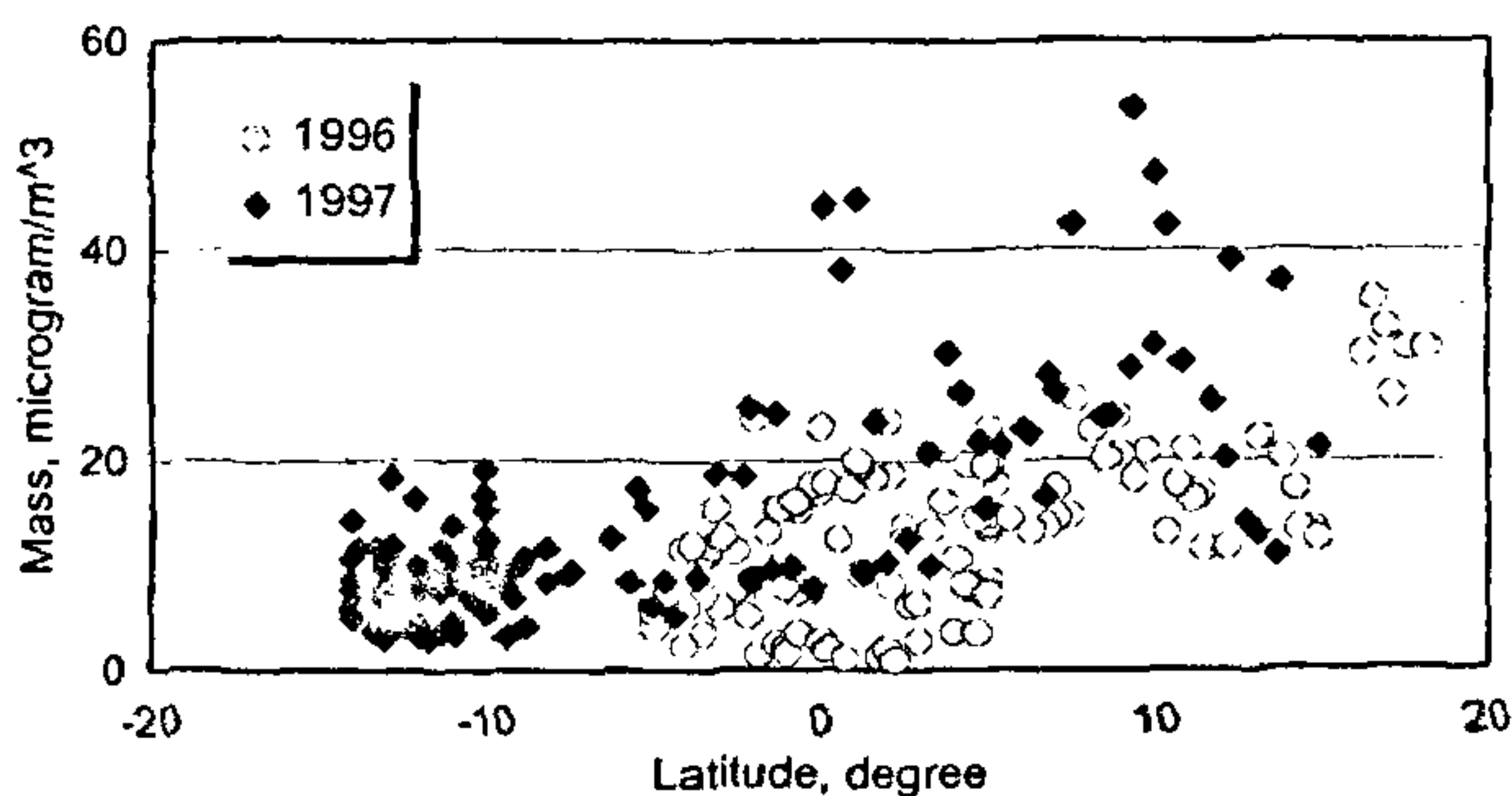


Figure 3. Measured aerosol mass concentrations in the study area. Values corresponding to only the submicron particles (sum of the mass values obtained from the last 5 channels of the QCM, corresponding to particle diameter less than 0.8 μm) present the similarity between the mass data and the AOD data in Figure 2.

individual size bins show in general three modes, one with mode radius around 0.05 μm or less, other with mode radius around 0.5 μm and a coarse particle mode with radii above 5 μm . The size distributions obtained for the coast (within about 200 km from the coastline), and the open ocean compare well with the size distributions proposed by Jaenicke⁸ for urban and remote continental regions respectively.

As the continental air moves off the coast the smaller particle (less than 0.05 μm radius) concentration decreases due to both coagulation and condensational growth; they then evolve into bigger particles (accumulation mode) with mode radius shifting to higher values. The concentration of these particles over the Arabian Sea and near the coast increases greatly compared to the Indian Ocean region. These submicron sized particles are responsible for the observed increase in the aerosol optical depth at lower wavelengths (e.g. at 399 nm) measured near the coastal region. The concentration of 0.05 μm radius particle continuously decreases from the coast until the ITCZ. However south of the ITCZ a sudden drop in the concentration of these particles occurs. Figure 4 shows the mass of aerosol particles measured at the last 6 channels of the QCM (corresponding to the smaller size end of the spectrum) during the 1998 cruise (cr132), at selected locations south and north of the ITCZ. A large difference is seen in the concentration of 0.05 μm radius particles (channel IX of the QCM). The

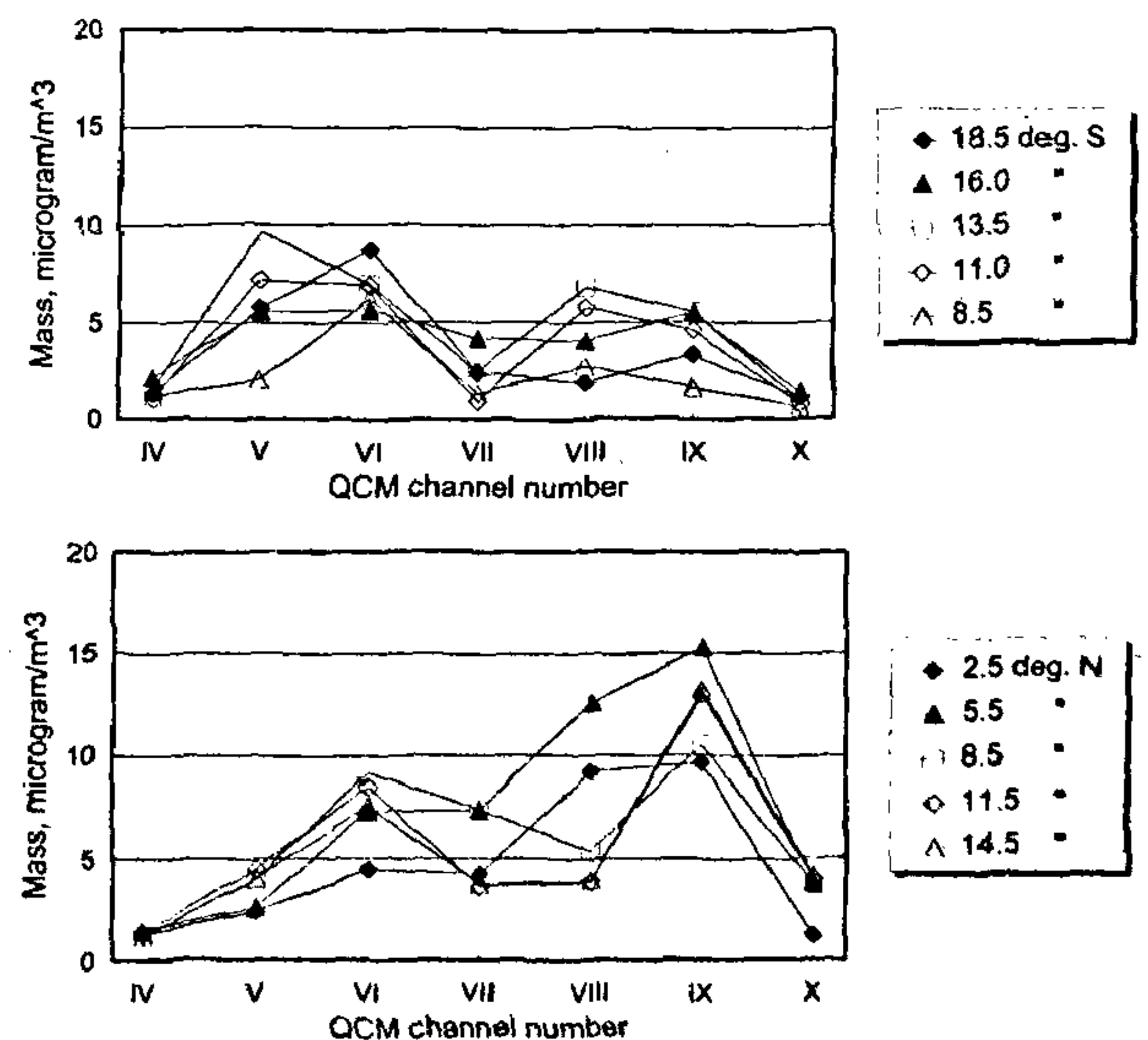


Figure 4. Mass of aerosol particles measured at the last 6 channels of the QCM, which correspond to the smaller size end of the spectrum, during the 1998 cruise (cr132), at few selected locations, south (above) and north (below) of the ITCZ. A large difference is seen in the concentration of 0.05 μm radius particles (channel IX of the QCM) measured in the two regions.

mass concentration of these particles (with a range of 1 to $5 \mu\text{g m}^{-3}$) south of the ITCZ increases almost three-fold at the north side of the ITCZ. The main difference in the two regions is the direction of the mean wind flow, which is southward in the northern region and northward in the southern region of the ITCZ. At the southern side of the ITCZ the pristine air flowing from the Antarctica lacks any man-made aerosols or precursor gases. The existing aerosols are mostly naturally produced, which include mainly sea salt particles and non-sea salt sulfate particles produced from dimethylsulfide (DMS). On northern side of the ITCZ, apart from these naturally produced aerosols, a large amount of anthropogenically produced particles are seen which are transported from the continents. From the data obtained it can be said that the naturally produced submicron (less than $1 \mu\text{m}$ radius) sized particles over the tropical Indian Ocean surface is of the order of 10 to $15 \mu\text{g m}^{-3}$. The excess concentration is from particles transported from the continents and their amount decreases with increasing distance from the coast. This excess concentration of submicron particles is of the order of 30 to $40 \mu\text{g m}^{-3}$ near the Indian coast; it then decreases and settles to a value of about 10 to $15 \mu\text{g m}^{-3}$ throughout the northern side of the ITCZ. Thus we state that anthropogenic particles form more than 50% of the total submicron particles seen over the tropical Indian Ocean.

The measured columnar aerosol optical depth is correlated with the independently measured radiative flux values to study the aerosol radiative forcing. In order to do so, the instantaneous optical depth values are multiplied by the air mass factor, $1/\cos \theta$, where θ is solar zenith angle, to get the slant path aerosol optical depth at the time of the measurement. Similarly, the measured flux values are multiplied by the $1/\cos \theta$ term, in this case to correct for the angle of incidence of the flux on the sensing area of the pyranometer or the photodiode radiometer. The 497 nm channel optical depth data are used for correlating with the visible flux, because 500 nm optical depth is considered typical for the visible (400–700 nm) region. In one of our earlier studies⁶, the optical depth and the direct as well as the scattered radiations measured during the first pre-INDOEX cruise of this series (cr109) were correlated to show the difference in radiative forcing exhibited by aerosols present near the coastal region and the interior ocean. Those results are not discussed further in this work. In this work we combine the cr109 and cr120 data and determine the diurnal averaged radiative forcing which is representative of the tropical Indian Ocean (north of ITCZ) for January to March. Global radiation data for cr132 are not available and hence not included for the radiative forcing study. However, it can be said, based on the aerosol optical depth data (Figure 1), that the aerosol characteristics measured during cr132 are typical of the region and season and do not significantly differ from the earlier data. Thus, the absence of the cr132 global radia-

tion data does not in any way affect the conclusion drawn on the radiative forcing.

Figure 5 shows the correlation between the global flux in the visible region of the solar spectrum from 400 to 700 nm measured using the photodiode radiometer and the columnar aerosol optical depth at 497 nm measured using the sun-photometer during the January 1996 cruise (cr109). A statistical fit through the data points show a negative gradient, and the estimated slope is -156 ± 8 . This means for a unit increase in aerosol optical depth there is a reduction of about 156 W m^{-2} in the surface reaching solar flux in the 400–700 nm. Similarly for the cr120 data from January 1997 the estimated reduction in the flux value is $-129 \pm 7 \text{ W m}^{-2}$ for every unit increase in aerosol optical depth at 497 nm. A slightly higher value obtained in the case of cr109 data is because the cruise did not venture much into the interior open ocean, and hence, the measured aerosol characteristics are more representative of the Arabian Sea. The higher radiative forcing obtained for this case indicates the presence of more absorbing particles over this region. On the contrary, the cr120 went much deeper into the open ocean, down to 15°S ; therefore, the data do not have bias for the more absorbing aerosols found over the Arabian Sea. The average radiative forcing obtained from these two cruises, which is $-143 \pm 8 \text{ W m}^{-2}$ can be considered representative for the tropical Indian Ocean. Thus, every 0.1 aerosol optical depth over the tropical Indian Ocean causes a reduction of about 14 W m^{-2} in the surface reaching solar flux, which is the direct radiative forcing by aerosols for the visible solar radiation during daytime.

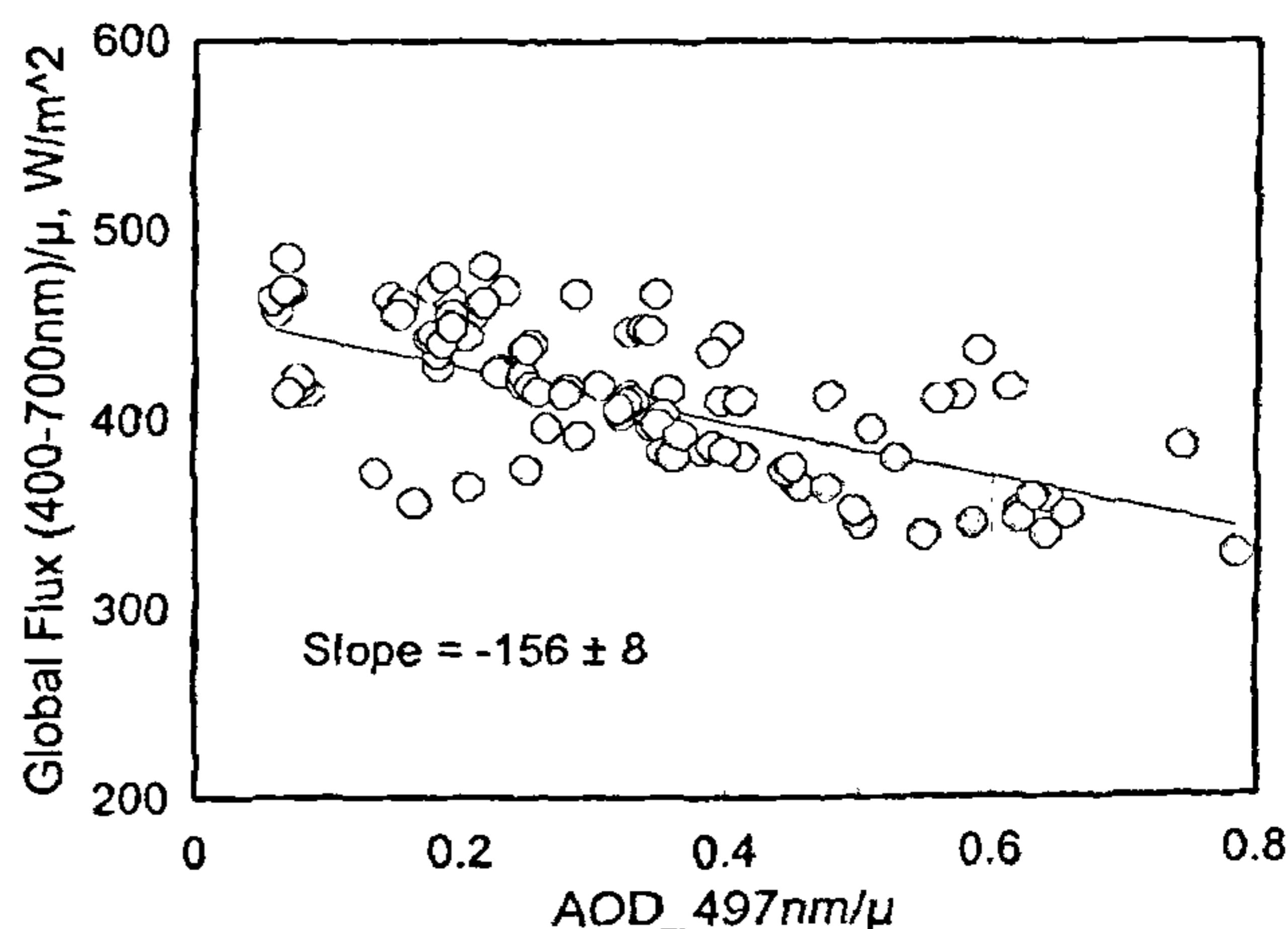


Figure 5. The global flux in the visible region of the solar spectrum from 400 to 700 nm measured using the photodiode radiometer is correlated with the columnar aerosol optical depth at 497 nm measured using the sun-photometer during the January 1996 cruise (cr109). A statistical fit through the data points shows a negative gradient.

Table 3. Results of the statistical fit done for the available data (about 400 data points), grouped into 10° degree solar zenith angle bins from 20° to 70°. The radiative forcing in terms of reduction in the visible solar flux for every 0.1 aerosol optical depth and the associated errors are given

Solar zenith angle (in degrees)	Radiative forcing (W m^{-2})	Uncertainty (W m^{-2})
0 to 20	*	—
20 to 30	11.4	1.6
30 to 40	10.5	1.3
40 to 50	10.8	1.5
50 to 60	8.9	1.5
60 to 70	8.7	1.6
70 to 90	*	—

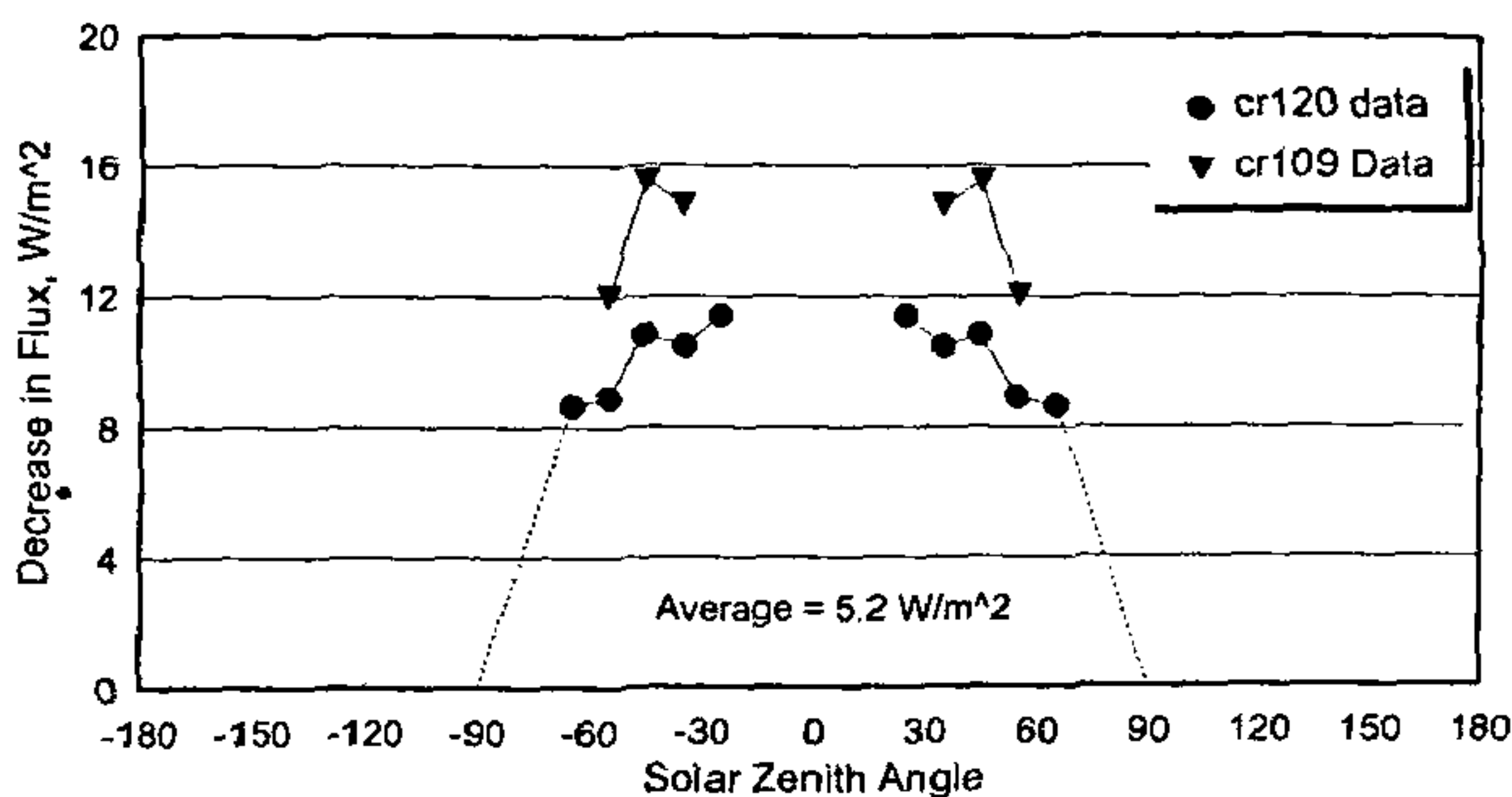


Figure 6. Result of the statistical fit done for about 400 data points, grouped into 10° solar zenith angle bins. The values and the associated errors are shown in Table 3. Taking the solar radiative forcing by aerosols as zero at night, the diurnal averaged radiative forcing is obtained as -5.2 W m^{-2} .

The above referred direct radiative forcing by aerosols, of the order of -14 W m^{-2} is the average value for the daytime measurements. However, theoretical estimates show that the forcing varies as a function of the solar zenith angle. In order to examine this effect, the optical depth and flux (which is about 400 data points) are grouped according to the solar zenith angle at the time of the measurement. To get statistically good sample points, the width of the zenith angle bin is taken as 10 degrees. Statistical fit, similar to the one shown in Figure 5, is made for the individual data bins, and the results are shown in Table 3 as well as in Figure 6. At night, in the absence of the sunlight, the direct radiative forcing by aerosols in the short wavelength is taken as zero. By combining all these results, the diurnal averaged radiative forcing by aerosols is estimated as -5.2 W m^{-2} for the aerosols found over the tropical Indian Ocean during January to March. In the literature, values can be found for the radiative forcing estimated theoretically for different aerosol types. For example, the combined shortwave radiative forcing due to sulfate aerosols and smoke is

estimated as -2 W m^{-2} . In an independent calculation, Harshavardhan⁹ shows that the optical depth for the global mean sulfate burden is 0.04 which corresponds to a radiative forcing of about -1.2 W m^{-2} . This becomes -3.0 W m^{-2} for 0.1 optical depth which is still less than -5.2 W m^{-2} obtained in the present study. This excess radiative forcing could be explained by invoking additional absorption by aerosols or any unknown absorbing gases in the visible region. Further studies could include a closed experiment where, apart from the above measurements, the backscattered radiation to space should be measured with preferably similar set of equipments from high flying aircrafts or satellites. Such an experiment will further explain the measured excess radiative forcing.

Summary

A series of three pre-INDOEX cruises were conducted during 1996, 1997 and 1998 in January–March to assess the climate forcing caused by both natural and anthropogenic aerosols. The detailed measurements of aerosol characteristics show a large hemispherical difference, with very high values of aerosol optical depth in the north of the equator, supporting the argument that large amount of aerosols are transported from the landmass surrounding the Arabian Sea. Though it is difficult to say the individual contribution from different aerosol types to the measured optical depth, the larger variation seen in the optical depth for smaller wavelength indicate that it is the submicron size particles, in the size range 0.1 to 0.4 micron radius, which are responsible for the observed large increase in the Arabian Sea. It is found that, the low level wind flow can transport aerosols within about to 6 to 7 days from the landmass to the inter-tropical convergence zone where the air from the northern hemisphere meets the relatively pristine air from the southern hemisphere. A systematic latitude gradient is seen in the mass concentration of the submicron particles from 15°S to about 22°N, similar to the behaviour observed in the case of the aerosol optical depth. South of the ITCZ, a sudden

drop in the concentration of these particles is observed showing the pristine nature of the atmosphere. It is found that anthropogenic particles form more than 50% of the total submicron particles seen over the tropical Indian Ocean. The measured columnar aerosol optical depth is correlated with the independently measured radiative flux values to study the aerosol radiative forcing. The diurnal averaged radiative forcing by aerosols is found to be about -5.2 W m^{-2} for the aerosols found over the tropical Indian Ocean during January–March. To the best of my knowledge, this is the first ever estimate on the direct radiative forcing by aerosols based entirely on the actually measured experimental data. Theoretical values for the combined shortwave radiative forcing due to sulphate aerosols and smoke is estimated as -2 W m^{-2} . The measured excess radiative forcing could be explained only by invoking additional absorbing aerosols or any absorbing gases in the visible region which is hitherto unaccounted for. Future closed experiments with satellite and air-borne measurements of the backscattered radiation may help to explain the measured excess radiative forcing.

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MEETINGS/SYMPOSIUMS/SEMINARS

International Symposium on Adhesion Aspects of Thin Films

Date: 28–29 October 1999
Place: New Jersey, USA

Topics include: Factors influencing adhesion – Residual stress, mechanical properties, contamination, etc.; Bond durability, corrosion prevention; Adhesion promoters.

Polymeric films: Plasma polymerized films; Photoresists; Organic insulators; Barrier layers; Effects of aging and environment on adhesion.

General systems: Polymer to metal and metal to polymer adhesion; Multilevel laminates involving glass, ceramic, metal and polymer thin films.

Fundamental issues: Role of surface chemistry, wettability and morphology; Fundamental adhesion mechanisms including film/substrate interactions.

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International Conference on Industry and Environment (ICIE 99)

Date: 23–25 December 1999
Place: Karad, India

Topics include: Impact of industrial pollution on air quality, forests, aquatic plants and animals, estuarine and marine ecosystems, agricultural crops and soils including case studies; Water pollution management in industries; Air pollution control in industries; Environmental regulations and laws; ISO 14000 and environmental management systems; Low cost or no waste industrial technologies; Solid waste management in industries; Hazardous waste management in industries; Impact of industrialization on social and economic environment.

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