

# Aerosol radiation cloud interactions over the tropical Indian Ocean prior to the onset of the summer monsoon

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**Prior to the summer monsoon, from December to April every year, the trade wind blowing from the northeast brings polluted continent air over the Arabian Sea and the tropical Indian Ocean. A variety of fine suspended particles, majority of them man-made, get accumulated over the ocean region till the summer monsoon sets in and cleans the atmosphere. Aerosols have long been recognized for their role in cloud development and rain formation. They act as condensation nuclei for the formation of raindrops and help precipitation. But, if they are present in large quantities they produce a narrow spectrum of small cloud droplets and do not allow the droplets to grow. These clouds do not precipitate and the rain formation is inhibited. The aerosol–cloud interaction could be playing a crucial role on the onset of monsoon over different regions and the spatial distribution of rain over India. Recent observations over the Arabian Sea and the tropical Indian Ocean, conducted as part of the Indian Ocean Experiment, have revealed the presence of large amount of aerosols over these regions. The aerosols brought from the Indian sub-continent and other surrounding regions have relatively high concentration of soot particles, which substantially reduce the amount of solar radiation reaching the ocean surface. Results on this direct effect of aerosols on the radiation budget as well as their impact on cloud properties are presented and discussed.**

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PRIOR to the onset of the Indian summer monsoon, particularly in the winter months, the surface wind over the Indian sub-continent and the surrounding ocean region is predominantly from the northeast (NE) direction. This large-scale circulation feature is responsible for the transport of a variety of anthropogenic gases and suspended particles from the continent to over the ocean surface<sup>1</sup>. The air coming from the continent is dry and does not give any rain over the northern plains, except that it picks up moisture when it blows over the Bay of Bengal and gives torrential rain along the east coast of India, the coastal Orissa and Andhra Pradesh (NE

monsoon). However, during summer, when the Sun moves into the northern hemisphere, the convective activity becomes stronger in the northern hemisphere and the surface winds start blowing from the ocean towards the landmass in the north. The wind predominantly coming from the southwest (SW) direction carries sufficient moisture to give the summer monsoon rain to almost all part of India<sup>2</sup>, though geographical variation does exist in the rainfall distribution<sup>3</sup>. The wind reversal from NE to SW and the summer monsoon rain is a regular annual feature, but the total rainfall received over India during the summer monsoon varies from year to year<sup>4</sup>.

It is known that aerosols scatter the incoming solar radiation back to space and hence can cause a temperature reduction at the surface while particles such as soot absorb the solar radiation and can warm the atmosphere. The aerosols are also found to affect the cloud properties, such as mean droplet size, residence time, etc. and can influence the precipitation regime on local and even global scales. For example, observations show<sup>5</sup> that in polluted areas over Thailand and Indonesia, clouds have very narrow droplet size spectrum which do not precipitate, whereas the similar clouds precipitate in less polluted areas, in about 10 to 15 min after their formation. In recent years, efforts are being made through coordinated field experiments, to investigate in detail the role of aerosols in altering the atmospheric radiation budget as well as the cloud properties. The Central Equatorial Pacific Experiment<sup>6</sup> (CEPEX) conducted over the Pacific Ocean in 1993, the Aerosol Characterization Experiment<sup>7</sup> (ACE) conducted over the southwest Pacific and the Atlantic Oceans, the Tropospheric Aerosol Radiative Forcing Observational Experiment<sup>8</sup> (TARFOX) conducted along the east coast of USA in 1996 and the Indian Ocean Experiment<sup>9</sup> (INDOEX) conducted in 1999 are some examples. Among these experiments, INDOEX is of relevance to India and to the Indian summer monsoon.

INDOEX was a major field experiment<sup>9</sup> conducted from 1996 to 1999 over the tropical Indian Ocean. The initial cruise experiments<sup>10–13</sup> conducted from 1996 to 1998 had culminated in an intensive and final experiment in 1999 involving two ships (*ORV Sagar Kanya*, belong-

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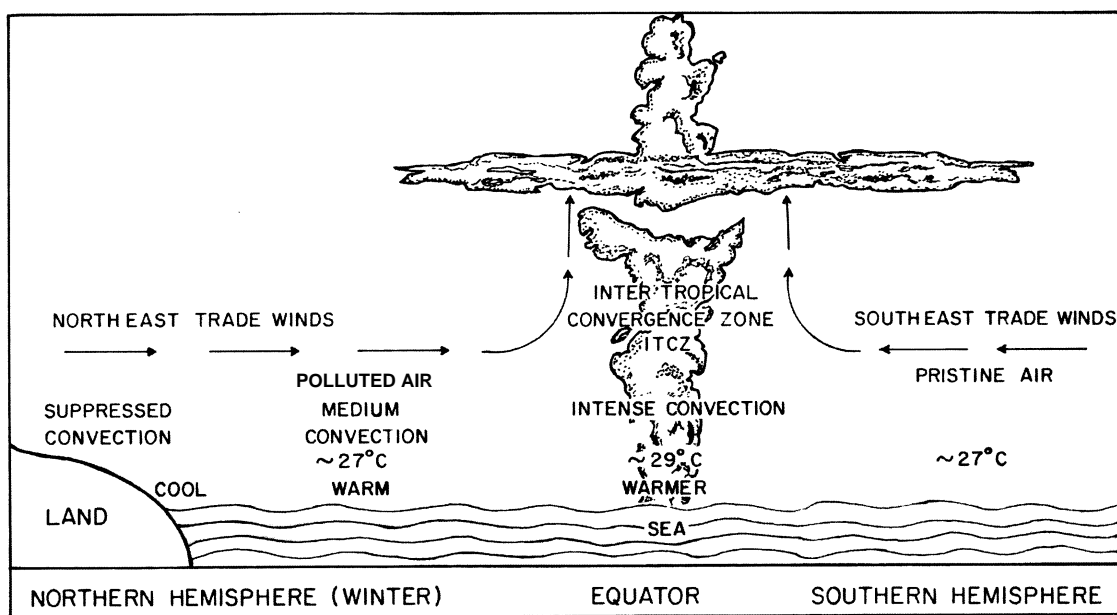
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ing to Department of Ocean Development, India and *R/V Ronald H. Brown*, belonging to NOAA, USA), aircraft, satellites and numerous field measurements in India and other island stations, with an objective to assess the climate forcing caused by both the natural and anthropogenic aerosols over the tropical Indian Ocean. On a global average, the climatic cooling effect of aerosols is shown<sup>14</sup> to compete with the warming caused by the increase in the greenhouse gases concentration. But, over regions of large aerosol concentration, such as over the ocean region surrounding the peninsular India, the aerosol effect could be much larger than the greenhouse effect and the forcing sign could be positive or negative, depending on whether the particles are absorbing or scattering. There are also studies<sup>15,16</sup> suggesting that the concentration of aerosols has increased during the last few decades. INDOEX has shown<sup>17,18</sup> that during winter months anthropogenic haze spreads over much of the Asian region and the northern Indian Ocean. As this happens, prior to the onset of the summer monsoon it is important to examine the impact of this continental size haze on the radiation budget and climate system. The data obtained from INDOEX form the main basis of discussion in this paper.

**Study area**

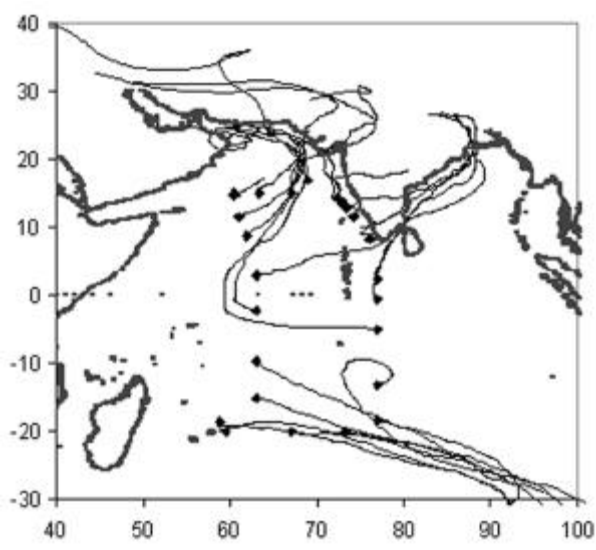
Over the tropical Indian Ocean, during winter months the northeast (NE) trade winds originating from the northern landmasses and the southeast (SE) trade winds originating from the sub-tropical high converge at the inter-tropical

convergence zone (ITCZ) (Figure 1). The polluted air brought from the northern land mass covers almost the entire region north of the ITCZ bound by the land masses of Arabia, Africa in the west and the southeast Asian regions, Myanmar, Thailand, Malaysia in the east. The tropical Indian Ocean region, including the Arabian Sea and the Bay of Bengal, is ideally suited to investigate the aerosol, radiation and cloud interactions. While the north of the ITCZ is polluted by the intense influx of anthropogenic aerosols, trace gases and their reaction products, at the south of the ITCZ the air is very pristine and is influenced very little by the continents. The polluted air from the north and the pristine air from the south are connected by a cross-equatorial monsoon flow into the ITCZ. During the northern winter months the ITCZ is generally located between the equator and 10°S latitude. The NE wind is responsible for a low-level temperature inversion and a suppressed convection over the northern landmass, leading to minimal rain in this region. This enables the continent air laden with pollutants to accumulate and spread north of the ITCZ. China and India being the two topmost populated countries of the world without very stringent pollution control norms, the study area becomes unique in the whole globe to examine the impact of aerosols on radiation and climate system. The wind patterns at the north and south of the ITCZ are distinctly different as shown by the air back trajectories (Figure 2) computed by the HYSPLIT-4 model using NCEP wind data for the January to March 1999 period (courtesy: NCMRWF, New Delhi). The thin lines in Figure 2 show the path an air parcel would have taken before reaching the ocean surface and the end points



**Figure 1.** Schematic diagram showing the convergence of the northeast trade wind from the land masses (India, China, etc.) and the southeast trade wind originating from the southern sub-tropical high at the inter-tropical convergence zone (ITCZ). The polluted air from the north and the pristine air from the south are connected by a cross-equatorial monsoon flow into the ITCZ.

are the locations visited by the ship during its 1999 cruise. The end point is taken 10 m above the ocean surface in the present computation, as most of the air sampling equipments were located at this height level in the ship. Air parcel originating as far as Turkey (40°N, 40°E), Iran and Pakistan could find its way to the mid-Arabian Sea whereas along the west coast of India the wind is predominantly from the southern Indian region. The air trajectories were computed for 10 days back in time and the parcel is found to lie mostly within the planetary boundary layer, below about 1 km altitude, all along its path. *In situ* air sampling over the Bay of Bengal region is not attempted, however the computed air trajectories are found (not shown in the figure) to be mainly from north and northeast India, China and Myanmar. At the south of the ITCZ, below about 10°S, the air is from the pristine southern ocean region (Figure 2) void of any major pollution from continents. The residence time of aerosol particles in the boundary layer is about a week and hence if the air parcel has to remain over the ocean for more than a week most of the aerosol particles would have settled on the ocean surface. Particles settled on the ocean surface are lost from the atmosphere and will not be airlifted again which however can happen over land surfaces. The study area in the present case is limited to over ocean surfaces, north of the ITCZ, from about 10°S to the coastal boundaries of the northern landmass. For comparison, data obtained over the pristine ocean region south of the ITCZ are also presented wherever applicable.



**Figure 2.** Air back trajectories computed by the HYSPLIT-4 model using NCEP wind data for the January to March 1999 period (courtesy: NCMRWF, New Delhi). The thin lines show the path an air parcel would have taken before reaching the ocean surface. Air parcels originating as far as Turkey (40°N, 40°E), Iran and Pakistan could find its way to the mid-Arabian Sea, while below about 8°S the wind is predominantly from the southeast direction. During the 10 days air back trajectories the air parcels were found to lie within about 1 km altitude, all along its path.

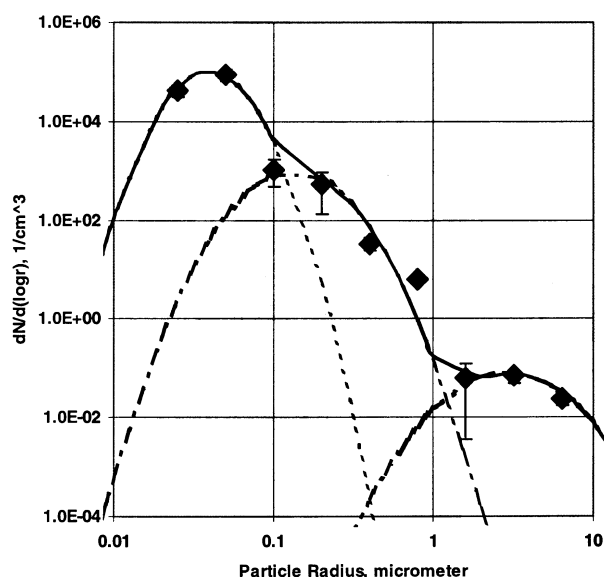
### Aerosol characteristics over the study region

Aerosols are particles of micron or sub-micron size found suspended in the atmosphere. Particles such as mineral dust, sea-salt, soot are directly injected into the atmosphere and are known as primary particles whereas particles such as sulphates are produced from precursor gases released into the atmosphere and are known as secondary particles. Natural (wind, forest fire, volcanic eruption) and anthropogenic activities (industries, automobiles, biomass burning) are responsible for the production of both primary and secondary particles. The Indian subcontinent and the surrounding regions are rich sources of a variety of aerosol particles. Fine crushed mineral (sand) particles from the arid and semi-arid regions, soot (carbon) particles from automobiles, industries, biomass burning, and sulphates from fossil fuel burning are the major components of aerosol found over the continent. Aerosols over the ocean region are mainly the sea salt (NaCl) particles produced by winds and bursting of air bubbles at the ocean surface. Aerosols are produced in varying proportions with large spatial and temporal inhomogeneity, and reside in the atmosphere, mostly in the first few kilometers, from few hours to few days depending on their size. Low-level surface winds and convective activities are responsible for the dispersal and transport of these particles to very long distances, to as far as the deep interior ocean region. Taking the average wind speed as 10 m/s we can show that the particles can be carried to distances, about 6000 km in seven days. The important removal mechanisms for the particles are gravitational settling and rain-wash. The sub-micron size particles can also coagulate to form bigger particles and the bigger particles are removed more readily from the atmosphere compared to smaller particles. We are concerned with the interactions of particles when they are airborne with the incoming solar radiation as well as with clouds.

For convenience the entire aerosol size range is divided into three major categories, viz. the 'nucleation mode' containing particles of size less than 0.1  $\mu\text{m}$ , the 'accumulation mode' particles having size between 0.1 and 1  $\mu\text{m}$  and the 'coarse mode' particles of size greater than 1.0  $\mu\text{m}$ . The nucleation mode particles are the secondary particles formed from gas to particle conversion mechanism and the accumulation mode particles are formed mostly by the coagulation of two or more nucleation particles. The coarse mode particles are the primary particles that are directly injected into the atmosphere, such as sea salt particles over the ocean and fine sand particles and biogenic dust over the land.

One of the important features observed over the Arabian Sea is that the concentration of the nucleation mode particles is very high, in the range of 20–50  $\mu\text{g}/\text{m}^3$ . As the precursor gases for the formation of these sub-micron particles come mostly from the continents<sup>19</sup>, the

newly nucleated particles are also found in great abundance near the coastal region. In contrast, the concentration of the nucleation mode particles south of the ITCZ is less than  $10 \mu\text{g}/\text{m}^3$ . Aerosol mass concentration measurements are made by a quartz crystal microbalance (QCM) cascade impactor system employed onboard the ship. Mass concentration values are also used to obtain the number density distribution taking appropriate aerosol density. More details on the measurements are available elsewhere<sup>20</sup>. Figure 3 shows the typical aerosol size distribution for the Arabian Sea region, constructed from a mean of more than 50 individual measurements. The standard deviation of the mean is shown as vertical bars. Three distinctive modes could be seen in the distribution and each mode can be fitted using a log-normal distribution curve as shown in the figure. The major difference between the aerosol size distribution obtained over the Arabian Sea and that for a 'standard ocean atmosphere' available in the literature<sup>21</sup> is the amount of nucleation mode particles. There is more than two order of magnitude in number concentration observed over the Arabian Sea compared to the pristine ocean region, and the concentration is comparable to that commonly found in a polluted urban location. Particle concentration obtained in the accumulation mode (0.1 to  $1 \mu\text{m}$ ) is more or less similar to model values available in the literature for other ocean regions while the coarse mode concentration is again higher which is due to the direct influence of primary particles brought from the continent. This indicates the strong influence of the continental aerosols in the marine boundary layer over the Arabian Sea. The



**Figure 3.** Typical aerosol size distribution for the Arabian Sea region, constructed from a mean of more than 50 individual measurements. The standard deviation of the mean is shown as vertical bars. Three distinctive modes could be seen in the distribution and each mode is fitted using a log-normal distribution curve.

aerosol optical depth (to be discussed later) in the visible region of the solar spectrum that is important to the radiative forcing is determined mainly by the amount of nucleation and accumulation mode particles.

The high concentration of aerosols found within the marine boundary layer north of the ITCZ is further verified by measuring the scattering and absorption efficiency of the particles. These optical properties of the aerosols, which are of direct relevance to the aerosol radiative forcing, are obtained by directly sampling the marine boundary layer air continuously using a nephelometer and a particle/soot absorption photometer (PSAP). The aerosol scattering coefficient is found to be in the range of  $0.075\text{--}0.125 \text{ km}^{-1}$  over the Arabian Sea and around  $0.150 \text{ km}^{-1}$  near the coastal region. A scattering coefficient value of  $0.025 \text{ km}^{-1}$  or less is considered to be 'low' between  $0.025$  and  $0.05 \text{ km}^{-1}$  represent 'medium' aerosol loading and values above  $0.05 \text{ km}^{-1}$  are due to 'high' aerosol concentration. High values over the entire study area, north of the ITCZ show the strong continental influence. Low values around  $0.015 \text{ km}^{-1}$  are obtained south of the ITCZ that can be considered as a representative background value for the pristine ocean region not influenced by continent air mass.

The chemical composition and the size of a particle determine its efficiency to scatter and absorb radiation. Aerosol samples collected over the Arabian Sea region and analysed<sup>18,19</sup> for their chemical constituents reveal that the major components are, sulphate, sea salt, mineral dust, black carbon (soot), nitrate, ammonium and organic matter. The chemical composition determines the refractive index of the particle and the scattering and absorption cross sections depend on the refractive index. The sulphate particles have stronger scattering efficiency and absorb very little visible radiation while the soot particles exhibits strong absorption and contribute significantly to a positive aerosol radiative forcing. The absorption coefficient measured using PSAP reveals that the absorbing particles (soot) are mainly concentrated near the coastal region and over the Arabian Sea and the values are in the range of  $0.01\text{--}0.02 \text{ km}^{-1}$ , which is an order of magnitude higher compared to the pristine ocean value of about  $0.001 \text{ km}^{-1}$ .

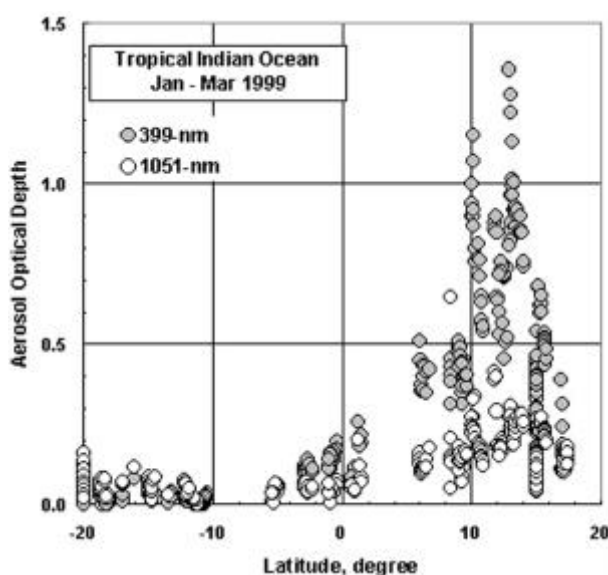
By combining the scattering and absorption coefficients the single scattering albedo,  $w$  for aerosols could be estimated, which is the ratio of the scattering coefficient to the total extinction (scattering + absorption) coefficient. The  $w$  values are found to be in the range of  $0.8\text{--}0.9$  (with an uncertainty of about  $\pm 4\%$ ) for the polluted regions and about  $0.95$  or more for the pristine ocean region, south of the ITCZ. Low  $w$  values are found more close to the Indian coastal region, where the measured columnar aerosol optical depth values also showed high values. For a  $w$  value of  $0.85$ , and aerosol optical depth in the range  $0.4\text{--}0.7$  in the visible region, the columnar aerosol absorption amounts to  $0.06$  to  $0.11$ . This is much higher than

the combined absorption by all known molecular gases, in the visible region of the solar spectrum. Carbonaceous aerosols (soot particles) from fossil fuel and biomass burning are the main contributors to the aerosol absorption, which contributes to positive radiative forcing similar to the greenhouse gases that absorb infrared radiation.

For aerosol radiative forcing studies we are concerned with the total column concentration of aerosols, distributed from surface to top of the atmosphere. This can be inferred by measuring the attenuation of the incoming solar radiation at selected wavelength bands in and around the visible region of the electromagnetic spectrum. The aerosol optical depth,  $d_I$  at a wavelength  $I$  is defined such that, the surface reaching direct solar radiation intensity decreases by  $\exp(-d_I/\cos q)$  due to aerosol scattering and absorption, where  $q$  is the solar zenith angle measured from the vertical. Figure 4 shows the aerosol optical depth measured at  $I = 399$  nm and 1051 nm during the *Sagar Kanya* cruise conducted from January to March in 1999. The observed feature is similar to the one seen in the earlier cruises<sup>10,12</sup> though differences exist in the absolute values. The steep latitude gradient seen in  $d_\lambda$  for  $I = 399$  nm, increasing from about 0.1 near 20°S to more than 1.0 around 15°N shows the increasing concentration of the sub-micron size particles as one nears the continent. Typically  $d_I$  value of 0.2 or less for the visible region is considered to represent the background value and any value above 0.4 is considered to represent polluted air.  $d_I$  measured at 1051 nm (Figure 4) show an increase from about 0.1 at the pristine region (20°S) to about 0.3 near the polluted coastal region, which is less compared to a

ten-fold increase observed in the case of 399 nm. Coarse particles, of size greater than about 1  $\mu\text{m}$ , contribute to  $d_I$  at higher wavelengths, which are mainly wind-derived soil dust or sea salt particles. Soil dust is mainly confined over the arid and semi-arid regions of the continents, and has limited range over the ocean surface<sup>22</sup>, while the sea salt particles are the dominant contributor to the coarse mode aerosols in the marine atmosphere<sup>23</sup>.

Though high aerosol optical depth observed over the Arabian Sea is an annual feature, year-to-year variation exists in the total aerosol amount. For example, the 1999 aerosol optical depth values are found to be higher than those obtained during the earlier years. Observations show that the 1996 and 1998 aerosol optical depth values are lower compared to the 1997 and 1999 values. Also, the amplitude of the annual variation is larger for the lower wavelength optical depth than compared to at higher wavelengths. As discussed earlier, anthropogenically produced sub-micron particles brought from the continents contribute significantly to the lower wavelength optical depth and hence the observed variation indicates annual variation in the transport and dispersal of the pollutants from the continent. As it is known, 1998 was an *El Niño* year and 1999 was more a *La Niña* year. A stronger inversion and larger number of anti-cyclones observed during the early 1999 had resulted in trapping the pollutants in the surface layer, which were transported to the ocean region and got accumulated. However, during 1998 the inversion was weaker over India that resulted in pollutants raising higher, where a sizeable portion got caught in the westerlies and moved east with a proportionate reduction in the amount of pollutants reaching the Indian Ocean (T. N. Krishnamurti, Florida State University, private communication). In the present study we consider only the average aerosol loading around the continent for the radiative forcing and cloud impact studies.



**Figure 4.** Aerosol optical depth measured at wavelengths 399 nm and 1051 nm during the *Sagar Kanya* cruise conducted from January to March 1999. The steep latitude gradient seen in the case of  $I = 399$  nm shows the increasing concentration of the sub-micron size particles in the 10° to 15°N region, close to coastal India.

### Aerosol radiative forcing

The Intergovernmental Panel on Climate Change (IPCC) defines<sup>24</sup> radiative forcing as a change in the average net radiation (either solar or terrestrial in origin) in  $\text{W}/\text{m}^2$  at the top of the troposphere (tropopause) due to changes in the concentration of atmospheric constituents with respect to their pre-industrial values. This definition helps to bring out the anthropogenic impact on the Earth-atmosphere system as a whole that is of relevance to climate change studies. Nevertheless, to study the instantaneous radiative forcing, a forcing efficiency term is defined in the present study, as well as in similar other studies<sup>10,25,26</sup> which is the ratio of the change in the surface reaching solar radiation intensity,  $\Delta F_\downarrow$  in  $\text{W}/\text{m}^2$  or the upscattered solar radiation flux  $\Delta F_\uparrow$ , leaving the atmosphere to the change in column aerosol amount

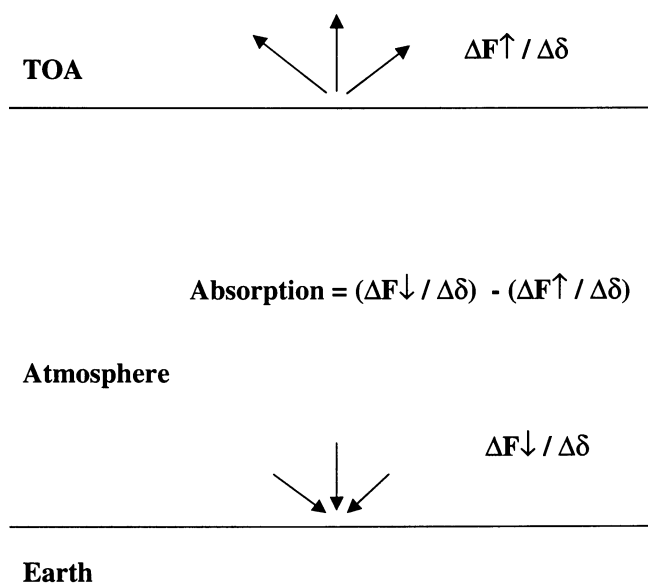
(usually the columnar aerosol optical depth,  $\Delta d$ ). The forcing efficiencies at the surface ( $\Delta F_{\downarrow}/\Delta d$ ) and at the top of the atmosphere ( $\Delta F_{\uparrow}/\Delta d$ ) are estimated separately and the difference between the two terms is defined as the net aerosol absorption efficiency within the atmosphere (Figure 5). The net forcing is obtained by multiplying the forcing efficiency and the average aerosol optical depth over the study area. This estimation helps in quantifying the role of aerosols in altering the radiation budget at the surface and top of the atmosphere (TOA) separately and the radiation loss due to absorption in the atmosphere. Moreover it is found convenient in analysing field data where the effects of aerosols are studied specific to a region and season.

Simultaneously measured surface reaching solar radiation intensity and the columnar aerosol optical depth are used to estimate the change in flux values (downward radiative forcing) for the different aerosol loading observed near the coast as well as over the interior ocean. The magnitude of the aerosol radiative forcing at any location and time depends on the amount of aerosols present, their optical properties, underlying surface albedo and the solar zenith angle. The direct solar visible (< 780 nm) flux values, measured using calibrated pyrheliometer, are compared with the instantaneous aerosol optical depth normalized for the solar zenith angle variation. The data are treated separately for the coastal region, open Arabian Sea region and the pristine Indian Ocean region south of the ITCZ. We estimate the diurnally averaged net surface forcing

for the three regions as,  $-27.0 \text{ W/m}^2$ ,  $-12.5 \text{ W/m}^2$  and  $-2.0 \text{ W/m}^2$  respectively, with an absolute uncertainty of about  $0.5 \text{ W/m}^2$ . The minus sign indicates a reduction in the flux value with increase in the aerosol optical depth.

From the estimated surface radiative forcing, TOA radiative forcing efficiency ( $DF_{\uparrow}/Dd$ ) could be derived by knowing the scattering efficiency of the atmosphere and the upscatter fraction (fraction of the scattered solar radiation that escapes to space). The downward scattering efficiency,  $e$  is defined<sup>10</sup> as the ratio of the increase in the diffuse sky radiation to the decrease in the direct solar radiation. The obtained  $e$  values are 0.38, 0.47 and 0.63 respectively for the coastal region, the open Arabian Sea and the pristine ocean region south of the ITCZ. Radiative transfer models<sup>27</sup> can be used to calculate the upscatter fraction  $b_c$  for different aerosol size and sun position. In radiative forcing calculations it is necessary to average the upscatter fraction for the entire diurnal variation of the solar zenith angle. The average value for  $b_c$  is about 0.5 for very small particles and about 0.2 for coarse particles. In general, a value of 0.3 is found appropriate for  $b_c$  for realistic aerosol atmosphere. Using the scattering efficiency values and the upscatter fraction, the radiative forcing at the top of the atmosphere due to aerosols is estimated as  $7.0 \text{ W/m}^2$ ,  $4.5 \text{ W/m}^2$  and  $1.5 \text{ W/m}^2$  respectively for the coastal region, the open Arabian Sea and the pristine ocean region south of the ITCZ. The difference between the forcing at the surface and the forcing at the TOA yields the net solar radiation absorption within the atmosphere and the values for the three regions are  $20 \text{ W/m}^2$ ,  $8 \text{ W/m}^2$  and  $0.5 \text{ W/m}^2$ . The atmospheric solar heating by about  $20 \text{ W/m}^2$  caused by aerosol absorption over the west coast of India is about a factor of 8 higher compared to the enhanced greenhouse warming caused by all the known greenhouse gases in the visible region of the solar spectrum. In Figure 6 the diurnally averaged aerosol radiative forcing values are presented individually for the surface, at the top of the atmosphere for the Arabian Sea, coastal India (west coast) and the Bay of Bengal regions. In the absence of *in situ* ship cruise measurements over the Bay of Bengal region, the NOAA satellite data are used to obtain the aerosol optical depth and the radiative forcing. More details can be obtained from Coakley *et al.*<sup>28</sup>. Though differences exist between the methodologies used for analysing the *in situ* ship data as well as the satellite data, the final results can be compared within few  $\text{W/m}^2$ . Radiative forcing detected over the ocean regions surrounding the Indian sub-continent is about an order of magnitude higher compared to the pristine ocean region south of the ITCZ. This is of great concern and its effect on the heat budget of the atmosphere and the impact on dynamics need to be studied in greater detail and depth.

In summary, it can be said that the surface forcing is about three times more than the TOA forcing in the polluted regions (in the BOB region it is more than four times) while the difference between the surface and TOA

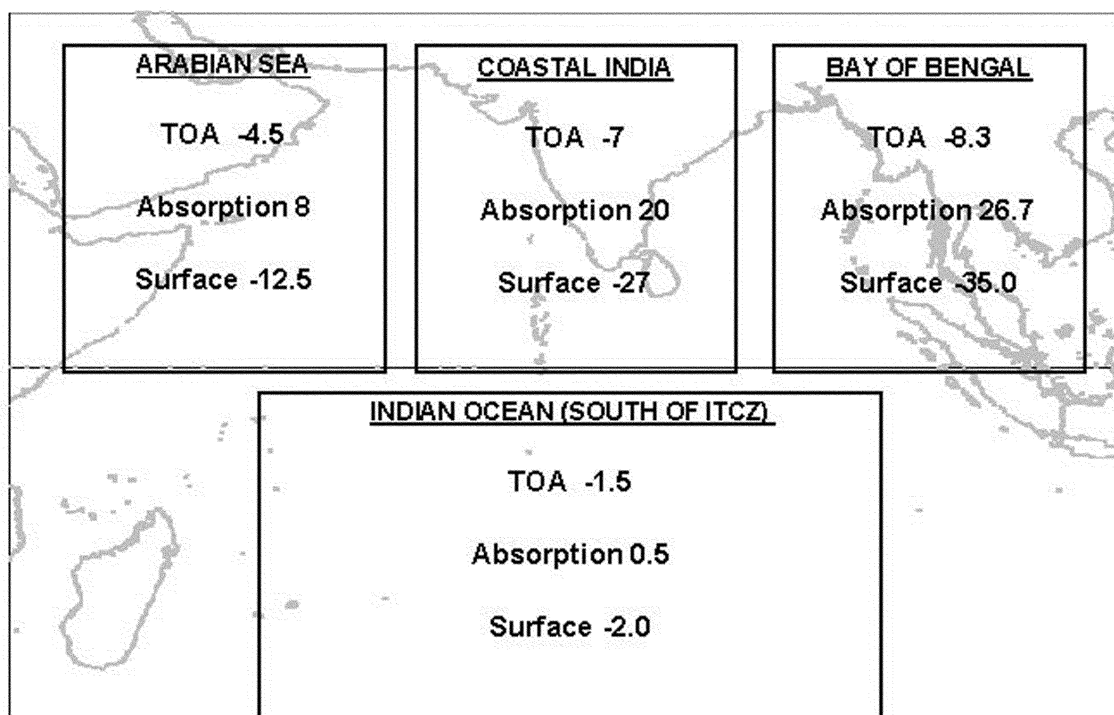


**Figure 5.** The instantaneous aerosol radiative forcing efficiency is defined as the change in the surface reaching solar radiation intensity,  $DF_{\downarrow}$  in  $\text{W/m}^2$  or the upscattered solar radiation flux  $DF_{\uparrow}$ , leaving the atmosphere to a change in columnar aerosol optical depth,  $Dd$ . The forcing efficiencies at the surface ( $DF_{\downarrow}/Dd$ ) and at the top of the atmosphere ( $DF_{\uparrow}/Dd$ ) are estimated separately and the difference between the two is defined as the net aerosol absorption efficiency within the atmosphere.

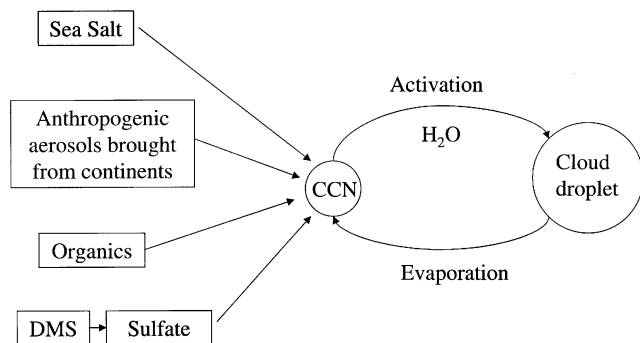
forcing is much less (surface forcing is about 30% higher than the TOA forcing) for the pristine region. Large difference between the surface and TOA forcing indicates aerosol absorption within the atmosphere. As presented earlier, the values of the single scattering albedo,  $w$  of the aerosols found near the coastal region (as well as over the BOB region) are very low, between 0.8 and 0.9 compared to the pristine ocean value of more than 0.95. Other studies<sup>18</sup> show that the column average of  $w$  lie generally in the range between 0.86 and 0.89 for the polluted regions. These  $w$  values represent a highly absorbing aerosol mixture. More or less similar aerosol characteristics over a very wide area indicate that the NE monsoon flow is responsible for the dispersion and mixing of continental aerosols to a very large region over the ocean surface. It should be noted that the above estimations are for clear sky conditions only as the radiation and aerosol optical depth data used in the above computations are screened for cloud contamination. In the presence of clouds, it is not sufficient to consider only the direct radiative effects of aerosols but also the indirect effect caused due to the cloud reflection. For example, computations<sup>18</sup> show that during overcast sky condition, the TOA forcing nearly doubles if the aerosol layer is above the clouds than below, because the elevated aerosol layer absorbs more solar radiation reflected by the clouds.

### Effects of aerosols on clouds

While the aerosol transport in the horizontal direction is controlled by winds and is a mesoscale process, the vertical transport and scavenging is determined by the convective motions and takes place in convective scales. The aerosol particles caught in the convective updrafts serve as cloud condensation nuclei ( $C_{CN}$ ) and help in the cloud droplet formation. The efficiency of an aerosol particle to serve as a  $C_{CN}$  depends on its size and the amount of water adsorbing material in it. Different aerosol types act as  $C_{CN}$  and help in cloud droplet formation (Figure 7). It is estimated<sup>29</sup> that about 50% of the total aerosol particles serve as  $C_{CN}$ . The fraction of aerosol particles which do not experience the  $C_{CN}$  activation is transported to high levels in which a certain fraction serve as ice nuclei and are responsible for ice crystal formation in the upper troposphere. It has been observed<sup>5</sup> that the cloud development is different for different air masses such as over ocean or continent. For example, the data for Thailand and Indonesia indicate that in over-polluted areas the clouds are found not precipitating because of the smaller droplet size while the similar clouds precipitate in clear air within 10 to 15 min after their formation. The precipitation rate, vertical distribution of latent heat and other cloud-related phenomenon in clear and polluted environment could significantly be different.



**Figure 6.** The aerosol radiative forcing (diurnally averaged forcing efficiency multiplied by the average aerosol optical depth over the region) values are presented individually at the surface, at the top of the atmosphere (TOA) and within the atmosphere. The aerosol radiative forcing for the Bay of Bengal region is estimated using NOAA satellite data<sup>28</sup>. The values for the Arabian Sea, coastal India (west coast) and the Bay of Bengal region are about an order of magnitude high compared to the pristine ocean region, south of the ITCZ.

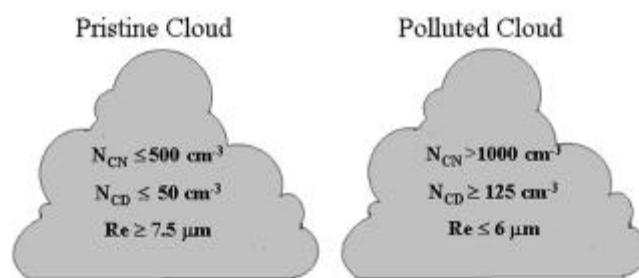


**Figure 7.** Different aerosol types contribute to cloud condensation nuclei (CCN) and are responsible for the formation and development of cloud droplets.

Over the present study area, the aircraft measurements<sup>18</sup> clearly demonstrate that the polluted clouds have the concentration of condensation nuclei exceeding 1000 per cc compared to less than 500 per cc in pristine air. Similarly, for the same average liquid water content (about  $0.1 \text{ g/m}^3$ ) in the polluted clouds, more cloud drops, in the order of 125 per cc and a smaller effective drop radii of less than  $6 \mu\text{m}$  are found compared to pristine air where the cloud drop concentration is less than 50 per cc and the effective drop radii is more than  $7.5 \mu\text{m}$  (Figure 8). An increase in the number of cloud droplets enhances multiple scattering of light within the clouds, which increases the optical depth and cloud albedo and hence influences the radiative forcing. An increase in the aerosol amount thus can suppress precipitation at least from low clouds. The rain formation within the clouds depends on the cloud drop size distribution. For a given liquid water content, precipitation will be accelerated in the case of less number of larger drops than with a larger number of smaller drops. The presence of soot, due to its absorption of solar radiation can also burn off the low clouds. A reduced rainfall leads to a reduced rain scavenging of aerosols that induces a positive feedback, with a possibility of an increase in the haze amount and extent.

## Conclusion

It has been shown that aerosols play a major role in modifying the earth's radiation budget over the polluted region. During clear sky (cloud free) conditions the aerosol direct forcing at the surface is shown to be about three times higher than the top of the atmosphere forcing and the difference is the forcing caused within the atmosphere by absorbing aerosols such as soot particles. More work needs to be done<sup>18</sup> to establish aerosol radiative forcing in the presence of clouds (indirect radiative forcing). Large inter annual variability can exist in the aerosol loading over the ocean surfaces surrounding the peninsular India, caused mainly due to varying meteorological conditions.



**Figure 8.** Aircraft measurements over the INDOEX study area (Ramanathan *et al.*<sup>18</sup>) demonstrate that for the same average liquid water content of about  $0.1 \text{ g/m}^3$ , polluted clouds have more number of condensation nuclei ( $N_{CN}$ ) and cloud droplets ( $N_{CD}$ ) compared to pristine clouds. The effective cloud drop radius ( $R_e$ ) is less in the case of polluted clouds. For a given liquid water content, precipitation will be accelerated in the case of fewer number of larger cloud drops (pristine cloud) than with a larger number of smaller drops (polluted clouds).

The consequences of the increased aerosol amount and the large solar radiation heating are many. For example, the increased aerosol amount decreases the surface reaching solar flux by as much as 15%. This would result in a decreased evaporation<sup>30</sup>, which will result in a decreased rainfall. Since most of the reduction in the surface reaching solar radiation is concentrated in the photosynthetically active visible and UV part of the solar spectrum, the aerosols could have an effect on the marine and terrestrial biological productivity<sup>31</sup>. While the increased aerosol concentration can suppress the precipitation from low clouds, the very large atmospheric heating by aerosol absorption obtained near the Indian coast can also burn off the low clouds.

What needs to be done in the Indian context is to initiate urgently a national effort to characterize both the naturally occurring and anthropogenically produced aerosols in different geographical conditions in India. Reverse transport models and air forward/back trajectory analysis indicate the importance of the northeast Indian region as a source region for the pollutants observed over the Bay of Bengal region which is hitherto not studied in detail. There is also an urgent need to do size resolved chemical analysis of aerosols to identify the sources (diesel vehicles/two-stroke automobiles, biomass burning). Role of large-scale atmospheric heating by aerosol absorption needs to be examined through coordinated modelling efforts. Observations on land surface processes (soil moisture, surface winds, etc.) need to be looked into for studying regional impacts of the atmospheric heating induced by aerosol absorption. The estimated population increase in India and China will make the region more susceptible to the associated effects of pollution. A national/international level coordinated studies and establishment of observatories for baseline monitoring of radiation and atmospheric chemistry should get the highest priority.



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