Developments in tropospheric aerosols studies in India

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The paper traces the development of aerosol studies in India in the last 5 decades. Measurements on atmospheric transparency, rain water chemistry and air sampling were the techniques employed up to 1990. From mid 1980s other systems like radiometers and lidar have been introduced. Aerosol research got a boost in India under the IMAP in 1980s, which enabled Indian research community to participate actively in INDOEX (1996-1999) phases. Large number of studies undertaken in India in pre-INDOEX and INDOEX phases reveal that aerosol concentrations are increasing and so is the component due to anthropogenic sources, in all seasons. There could be implications of these trends on regional climate system. The need for enhanced monitoring on a regular network mode, archival of data and modelling is emphasized under a well coordinated multi-agency programme.

1 Introduction

Tropospheric aerosols refer to suspended particles in the atmosphere from surface to the tropopause level. These particles have varying diameters from less than a micrometre (µm) to about 10 µm, known as fine and coarse particles, respectively. These particles owe their origin to natural and man-made (anthropogenic) sources. On the global scale, natural sources contribute much larger to the total aerosol loading than the anthropogenic sources on annual average basis. On regional and local scales, anthropogenic aerosols can dominate significantly. Over India, natural sources contribute 60-70% to the total aerosols in the pre-monsoon, monsoon and post-monsoon seasons. The contribution of anthropogenic sources is substantial in winter. Aerosols of natural origin are like mineral dust particles, particles originating from volcanic eruptions and sea sprays, etc. Volcanic aerosols can be thrown up to the stratosphere and resides there for even a year or more. Residence time of tropospheric aerosols is about a week during which period they are transported over long distances (=1000 km) horizontally and up to 3-5 km in the vertical. During this period they change their chemical and physical characteristics by heterogeneous gas to particle chemistry or nucleation and get scavenged by coagulation, cloud and precipitating processes.

There are many types of aerosols; two specially important ones for climate processes are carbonaceous (organics and black carbon) and watersoluble (sulphate and nitrates) aerosols. Aerosols exert both direct (scattering and absorption) and indirect (cloud formation and changing of microphysics of clouds) forcings on the climate system. Important variables of aerosols are their mass, size, molecular concentration, optical depth, chemical composition and vertical distribution density. Accurate regional measurements are required to yield quantitative information on their characteristics, which when combined with spatial and temporal information (obtained through surface, aircraft and satellite monitoring and model calculations) determine the regional scale climate forcing due to aerosols.

Many of the characteristic properties of aerosols are being investigated for over four decades1-3. The total top of the atmosphere (TOA) forcing due to tropospheric aerosols is estimated to be −0.5 Wm⁻² (soot particles +0.1 Wm⁻²), opposite to the total TOA forcing by green house gases which at present is estimated as +2.45 Wm⁻² with an estimated uncertainty of 15%. The global scale TOA indirect effect of tropospheric forcing has a large uncertainty varying from 0 to −1.5 Wm⁻². Accepting the higher limits of the above forcing, tropospheric aerosols are expected to contribute −2.0 Wm⁻² to global radiative forcing which is almost equal to the green house forcing. This explains the interest which aerosol research has generated in the last two decades, resulting in the launching of several regional aerosol characterizations field experiments in different climatic environments. These experiments have determined regional direct and indirect aerosol
climate forcings and currently it is speculated that aerosols may play crucial role in the modelling of the regional climate.

Aerosol studies have attracted Indian research community for nearly 5 decades and extensive results on rain-chemistry, air sampling techniques, measurements on depletion of solar radiation, atmospheric turbidity, etc. have been reported. In the last two decades vertical distributions of aerosols by lidar measurements have also been undertaken at Trivandrum and Pune. Till 1980 two major groups were involved in these studies, namely, (i) the India Meteorological Department (IMD) and (ii) the Indian Institute of Tropical Meteorology (IITM), Pune. However, under the Indian Middle Atmosphere Programme (IMAP), organized in 1980s, several groups in other research institutes notably the Space Physics Laboratory (SPL), Trivandrum, the National Physical Laboratory (NPL), New Delhi, the Physical Research laboratory (PRL), Ahmedabad and universities (Andhra University, Mysore University and others) have taken up organized programmes on tropospheric aerosol research. Such programmes were particularly promoted by the Indian component of INDOEX under the dynamic leadership of Dr A P Mitra. In this paper, the extensive works done by these groups have been reviewed.

2 Tropospheric aerosols studies in India prior to INDOEX

Studies on tropospheric aerosols were initiated in India in 1950s, under IMD in mid 1950s and under CSIR at their Rain and Cloud Physics (RCPR) unit attached to NPL, New Delhi (established in 1955 and later merged with IITM, Pune in 1967). Mukherjee studied the rain chemistry at Calcutta, an industrialized metropolitan centre in India. In 1960s and 1970s, IMD set up a network of radiation stations whose data have now become a benchmark for determining the changing transparency of the atmosphere under the impact of urbanization and industrialization. In 1970s, IMD established a network of atmospheric turbidity measurements and the background air pollution (BAPMON) network of 10 stations located in different urban, semi-urban and remote island stations. The IITM also launched a major effort under its Weather Modification and Atmospheric Chemistry Programme to measure air, cloud and rain samples for detecting the concentrations of chemical constituents of the tropospheric particulates. It has now data from its regular stations at New Delhi and Pune for nearly 30 years and has also collected data in campaign modes from different stations (urban, rural, hilly, industrial, forest and along the coastal waters) and samples drawn from clouds and fog. Most of its data refer to rainfall samples during the summer monsoon season (June-September), though data for other seasons in campaign mode have been collected for some years. The study of aerosols in India took another leap forward in 1980s when the SPL, Trivandrum, of the Department of Space (DOS) established a regular programme on aerosol research. By the time the international INDOEX was being planned there were several research groups who could collaborate in INDOEX under a coordinated effort contributed by India and carry it further in the post-INDOEX period as a regular programme. Vast scientific reports have come out as a result of nearly 50 years of work from the Indian research community. Some of the salient features of this research efforts are summed up in the following sub-sections.

2.1 Transparency of the atmosphere

(i) Long-term measurements on incoming solar radiation at Pune and other stations in India show reduction in direct solar radiation by about 15% over the years 1972-1992. Whereas global solar radiation has been found to have a decreasing trend, and the diffuse solar radiation shows an increasing trend. The analysis of the data needs to be done on cloudy and cloud-free days so as to remove the modulations in solar radiation by inter-annual and any long-term variations in the cloudiness and water vapour.

(ii) The trends in turbidity measurements are also found to be negative since observations were started by IMD.

(iii) Horizontal visibility data at several airport stations, which are usually located near semi-urban surroundings, show a significant decreasing trend.

The above information coming from diverse measurements clearly point to increasing aerosol load over India, which could be ascribed to increasing urbanization, changes in land-use pattern and development of small, medium and large industries.

2.2 Chemical nature of rain and air samples

Anthropogenic emissions of acid precursors, like particulates, SO₂, NOₓ and NH₃ from industrial, transportation and agricultural sectors, lead to
acidification of rain water. India had almost negligible emissions from these sources till 1960s, prior to rapid increase of industry and green revolution in agriculture. The situation has considerably altered since then with the increasing use of fossil fuel as a source of energy and massive fertilizer inputs for intensive agriculture. Measurements on rainwater pH and chemical constituents of rainfall and air samples undertaken by IMD, IITM, National Environmental Engineering Research Institute (NEERI), Nagpur, and Central Pollution Control Board (CPCB) in different environments have been showing an increase towards acidity, though on the whole, rain and air samples over India are still alkaline in nature (except in the vicinity of petrochemical industries and thermal power stations). This is mainly ascribed to the presence of mineral dust with high composition of calcium, magnesium and other similar metallic constituents. However, the total suspended particulate (TSP) matter has considerably increased and is above the internationally recommended limits in high urban centres except in rural, hilly and remote environments. Some of the important results of extensive studies undertaken by different groups are the following:

(i) There is no large-scale episodes of acid rain over India, except in the vicinity of highly localized emission sources. This is primarily due to the buffering action of natural mineral dust which neutralizes the gaseous particles like sulphates and nitrates and others. The cations in all type of environments are dominated by calcium, which is of soil origin. The concentrations of anions like SO$_4^{2-}$ and NO$_3^-$ are small even in urban environments, and these acidic components are much smaller in rural, hilly forest and remote environments. Empirical orthogonal function analyses reveal that Cl$^-$, NO$_3^-$, Na$^+$, Ca$^{++}$ and Mg$^{++}$ constitute the factor 1 accounting for nearly 40% of the variance. Factor 2 is dominated by NH$_4^+$, K$^+$ and they, in combination, constitute 17% of the variance. Factor 3 is dominated by SO$_4^{2-}$ and Na$^+$ which together accounts for 11% of the variance. The potential acidity of the rain is given by:

\[
\text{Potential acidity} = [\text{SO}_4^{2-}] + [\text{NO}_3^-] - [\text{NH}_4^+] - [\text{Ca}^{++}] - [\text{Mg}^{++}] - [\text{K}^+] 
\]

(ii) Since the cations dominate in all the three factors in India, the overall balance is towards the cations, thereby keeping the pH of rain water within the alkaline range. However, long-period records of BAPMON stations and IITM-managed stations at New Delhi and Pune clearly show that pH over the years has a decreasing trend. This would mean that anthropogenic emissions of SO$_4^{2-}$ and NO$_3^-$ are on the increase. Average of major ionic components are given in Table 1.

(iii) High chlorine and sodium content is due to some stations being coastal, whereas the SO$_4^{2-}$ and NO$_3^-$ are obviously higher in urban centres. Higher concentrations of sodium, calcium and magnesium are able to neutralize acidic effects. There is no marked difference between urban and rural centres with regard to NH$_4^+$ and K$^+$. Over urban centres, nearly 60% of the chemical constituents are of anthropogenic origin. These cannot be regarded as high anthropogenic emissions over India at present, though the situation may change in favour of anthropogenic sources in the next several years. However, there is significant inter-annual variability near the urban centres due to persistent stable and low ventilation of the atmospheric boundary layer (ABL) near urban centres, particularly in winter season. The effect with regard to gradual decline of alkalinity (or increasing acidity) is rather slow over India as compared to countries of Western Europe and North America, due to slow pace of industrialization in India. Indian coal has a low sulphur content, and increasing use of natural gas as industrial fuel in India is also likely to keep the acidic components of the emissions on the lower side. Naik et al. and Rao et al. have examined the rainwater chemistry of consecutive showers at Pune and have shown marked decrease in the concentrations of soil-origin elements followed by decrease in pH of the subsequent samples. As the soil-origin elements get washed off preferentially, sequential sampling during a single shower gives

<table>
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<tr>
<th>Stations</th>
<th>Concentration of (mg/l)</th>
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<th>Concentration of (mg/l)</th>
<th>Concentration of (mg/l)</th>
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<tbody>
<tr>
<td>Concentration of</td>
<td>Cl$^-$</td>
<td>SO$_4^{2-}$</td>
<td>NO$_3^-$</td>
<td>NH$_4^+$</td>
</tr>
<tr>
<td>Urban centres</td>
<td>3.2</td>
<td>2.3</td>
<td>1.9</td>
<td>0.4</td>
</tr>
<tr>
<td>Rural, hilly and forest areas</td>
<td>1.3</td>
<td>1.5</td>
<td>1.2</td>
<td>0.4</td>
</tr>
</tbody>
</table>
better information about the ionic balance as compared to bulk sampling for long duration. Therefore, there is a need to collect regular data on sequential sampling to monitor acidic rain.

(iv) The concentrations of SO₂ and NO₂ are higher in industrial and commercial areas, as they are near the source regions of anthropogenic emissions, with inland centres (Mumbai, Calcutta, New Delhi, Kanpur, Ahmedabad) registering higher values than the near-coastal centres (Cochin, Chennai, Trivandrum).

(v) The IITM has also made campaign-mode measurements of TSP over several places. The data suggest large variations of TSP in India ranging from nearly 40 µg m⁻³ to 510 µg m⁻³ between remote rural locations and urban centres. In the Indian environment, concentrations of soil-oriented acid-soluble elements (like Al, Fe and Mn) are much higher compared to other elements (like Pb, Cd, Zn, Cu, Ni, etc.) contributed by anthropogenic sources. However, overall concentrations are lower than those in Europe and America. The average concentrations of water-soluble TSPs are high in India as compared to western countries. Table 2, after Pillai et al.²⁹ and Safai et al.³⁰ provides data on averaged concentration for different environments.

(vi) Concentrations of natural dust-borne metallic elements are considerably higher than those of the anthropogenic origin in India, though near the power plants and urban centres the anthropogenic components are also substantial. Again season-wise separation of data shows that the TSPs in pre-monsoon and winter seasons have higher values than in the monsoon and the post-monsoon seasons. Also greater contributions come from anthropogenic sources in the pre-monsoon and winter seasons. This behaviour could be explained as the cleaning of the tropospheric aerosols by cloud scavenging and washout processes in monsoon season and the air being cleaner in the post-monsoon season. The relative stability of the boundary layer in the winter season and high raising of the soil dust in the pre-monsoon season due to mechanical turbulence account for higher values in these two seasons.

2.3 Influence of atmospheric pollutions on cloud microphysics (indirect effects of aerosols)

The IITM group has also made several investigations on the role of anthropogenic particles in cloud microphysical and rain processes by comparing the concentrations of different ionic components over the industrial and nearby rural complexes as well as upwind and downwind of Mumbai industrial complex. These studies have shown the following aspects:

(i) Rainfall in February (winter), which is quite small around Jamshedpur-Burnpur (steel manufacturing towns) complex, has undergone a significant decrease during the period 1872-1955, suggesting the sensitivity of rain formation in winter season to effluents from steel mills.³¹,³²

(ii) During increasing industrialization epoch of Mumbai between 1941 and 1969, the rainfall of the region, downwind of Mumbai, increased by 15% (statistically significant) compared to rainfall in nearby non-urban regions.³³

(iii) The air temperature at the cloud-base level was found to be higher by about 1 K in the urban (industrial) environment than in the non-urban environment close to Mumbai, suggesting the absorption of direct solar energy by anthropogenic aerosols.³⁴

(iv) Surface and aircraft observations of trace gases (SO₂ and NOₓ), and giant size hygroscopic and non-hygroscopic aerosols showed that chemical and microphysical conditions of clouds are markedly different in the upwind and downwind regions of the industrial complex of Mumbai region. The observed increase in rainfall in the downwind region (Kalyan)

Table 2—Average concentration of TSP and their acid-soluble and water-soluble components in India

<table>
<thead>
<tr>
<th>Location</th>
<th>TSP (µgm⁻³)</th>
<th>Concentration (ngm⁻³) of</th>
<th>Concentration (µgm⁻³) of</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Al</td>
<td>Fe</td>
</tr>
<tr>
<td>Marine</td>
<td>39</td>
<td>699</td>
<td>834</td>
</tr>
<tr>
<td>Coastal</td>
<td>51</td>
<td>480</td>
<td>511</td>
</tr>
<tr>
<td>Power plants</td>
<td>350</td>
<td>3557</td>
<td>4967</td>
</tr>
<tr>
<td>Urban</td>
<td>131</td>
<td>2881</td>
<td>2247</td>
</tr>
<tr>
<td>Rural</td>
<td>86</td>
<td>2038</td>
<td>1775</td>
</tr>
<tr>
<td>Forest</td>
<td>41</td>
<td>997</td>
<td>1030</td>
</tr>
</tbody>
</table>
of the Mumbai industrial complex was ascribed to the increase in number of giant size particles and higher droplet concentration as compared to those in upwind regions (off Mumbai coastal region). Concentrations of hygroscopic aerosols (sea-salt particles) were more in the upwind region than in the downwind region suggesting their fast decrease away from the coast, such that they are only 25% of their coastal values at a distance of about 50 km from the coast – though the wind is fairly strong from the coast. On the contrary, the concentrations of anthropogenic aerosols (SO2 and NOx) were significantly higher in downwind side (Kalyan) compared to the upwind side (Colaba and Alibag) of Mumbai.

(v) Concentrations of Na, K, Ca, Mg, do not show significant differences on upwind and down sites, whereas concentration of NH4+ was found to be low at upwind and high at downwind sites.

The industrial establishments in Mumbai region have increased several folds in the last 5 decades (refining petrochemicals, synthetic material plants, fertilizer plants, textile plants, etc.). As a result, the annual mean concentration of TSP has increased by 30-50% and the total annual mean concentration of NO2 has increased by four folds. The indirect effects of such a large increase in CCN over the last 5 decades on cloud processes have led to the reduction in the efficiency of warm cloud mechanism for producing rainfall over the region. The rainfall data, as examined by Khemani and Ramamurty33 for the period 1941-1969 showed an increase in the rainfall for Mumbai region over the period 1901-1940. However, Naik and Khemani35 showed decrease in rainfall over some stations (Panvel and Kalyan) for the period 1970-1986. Apparently there is natural variability of the rainfall on decadal scale which would not allow a clear signal to emerge with respect to the modification of rain processes by indirect effects of anthropogenic aerosols.

2.4 Characteristics of urban aerosols

Several investigations have been made by the IITM group about aerosol size distribution over Pune and other stations by surface and aircraft measurements over Deccan plateau, which have brought out the following aspects36,37:

(i) Bimodal distribution in total aerosol content with one maximum in the size range 0.04-0.06 μm (small particles) and the other in the size range 5-6 μm (large particles or accumulation mode). There is some seasonal dependence to these two modes.

(ii) Multispectral extinction measurements at Pune38 on cloud-free days showed that aerosols in coarse mode are more prominent. Aerosol optical size spectrum has been approximated by a composite power law with different exponents. Average aerosol optical depth (AOD) showed a seasonal dependence with high value of about 0.4 in the pre-monsoon dust-laden season which is reduced to about 0.2 in the clear post-monsoon season and is again increased to about 0.3 in the stable winter season. The AOD also showed a dependence on the integrated relative humidity in atmospheric column within surface to 5 km altitude; the increase being rather slow up to R.H. of 45% and thereafter it was sharp.

The SPL group at Trivandrum has focussed their studies on tropospheric aerosol characteristics along coastal sites of Trivandrum and Visakhapatnam and inland sites of Mysore. The AODs have been measured in different wavelengths using multispectral radiometers. A large number of research publications have resulted during 1980s and 1990s from this group and some of the salient aspects of these are summarized as follows:

(i) Climatology of AOD at Trivandrum peaks during March-August. The peak in pre-monsoon was accounted for sea-salt aerosols as well as urban-origin aerosols, whereas the monsoon season peak is primarily due to aerosol of marine origin39. The peak occurs over the Arabian Sea island, Minicoy, in monsoon season and is primarily contributed by sea-salt, though desert dust from west Asian region is also brought on the stations occasionally. Strengthening of wind speed also increases AODs (Refs 40, 41).

(ii) At the continental stations, New Delhi and Jodhpur, two maxima occur in pre-monsoon and winter seasons. Summer monsoon and post-monsoon seasons show relatively low AODs.

(iii) Diurnal variability in AODs is also prominent which is explained by atmospheric stability and wind conditions42.

(iv) Data for 15 years from different stations showed a clear positive trend39 in AODs which is ascribed to increasing urbanization and other anthropogenic sources. The annual rate of increase is estimated to be 6-9% in visible wavelengths and about 2% in IR spectrum at Trivandrum.

(v) The AODs in coastal belt showed influence of winds, land and sea breezes and relative humidity33-45. A north-south gradient in AOD with scaling distance
varying between 1000-2000 km, depending on wind speed, is observed at shorter wavelengths.

(vi) Spectral AOD at Trivandrum showed the bimodal characteristics\(^{46}\) during pre-monsoon and monsoon seasons and mono-modal behaviour is observed during the other two seasons. The prominence of different modes depends on the modulation by the land and sea breezes. The accumulation mode (sub-micron size) is due to local urban sources and long-distance transport into the region during winter period\(^{47}\). Sub-micron size aerosols account for the bulk of the surface area of the aerosol, but their contribution to the total volume is less. The sub-micron aerosol mass concentration is high (about 86 $\mu$g m\(^{-3}\)) during dry months and winter and decrease to a small value of about 11 $\mu$g m\(^{-3}\) during wet seasons. The mass concentrations in coarse mode aerosols behave in the opposite manner with low values (15 $\mu$g m\(^{-3}\)) in dry months (winter) and high values in wet monsoon period, highlighting the role of sea-salt particles at the station\(^{48}\).

(vii) Particulate matter (PM), PM\(_{10}\) and PM\(_{2.5}\) at Trivandrum showed that PM\(_{10}\) contribution to PM is highest during pre-monsoon and the contribution of PM\(_{2.5}\) to PM peaks during winter. These shares are lowest during the wet monsoon months\(^{49}\). The average diurnal variations in all PMs show a daytime low and nighttime high, suggesting the role played by daytime ventilation of the boundary layer by thermal and mechanical turbulence and nighttime influence of the land breeze.

2.5 Lidar-based studies

The sources of tropospheric aerosols are located near the earth’s surface and the aerosols are carried upward into the troposphere by vertical mixing processes which depend on atmospheric stability (diffusion) in the boundary layer. Changes in particulate concentrations also depend on long-range transports within the lower-middle troposphere and also on gas-to-particulate heterogeneous processes. Thus, lidar-based measurements of vertical distribution of aerosols become important for elucidating these processes. Lidar studies of tropospheric aerosols began in India in mid 1980s both at IITM, Pune and SPL, Trivandrum, and recently a lidar has been established at PRL, Ahmedabad.

2.5.1 Tropospheric aerosol studies by IITM group with lidar at Pune—Pune is an urban station, situated in the lee side of the Syahadri hills on western ghats.

The IITM installed a lidar facility towards the mid 1980s which has been extensively used for a large number of investigations on vertical distribution and other characteristics of tropospheric aerosols. Main features from these investigations\(^{40-56}\) are summed up as follows:

(i) Based on 1986-1998 statistics, the aerosol columnar content showed a maximum ($280 \times 10^6$ cm\(^{-2}\)) in May in the pre-monsoon season and a minimum ($120 \times 10^6$ cm\(^{-2}\)) in July in the summer monsoon season. The decrease occurs in episodic manner with the onset of monsoon season. The seasonal and annual variability of aerosol load depends upon the inter-annual variability in monsoon rainfall over the station. The columnar aerosol increases, rather rapidly, in the intervening dry period after a rainfall spell in the monsoon season, though it would not reach the high value of the pre-monsoon season, as the soil is moist and clouds are present during the monsoon season.

(ii) There is considerable inter-annual variability in each month in columnar aerosol content with coefficient of variation ranging from 25% to 40%. Larger variability is observed in pre-monsoon months and smaller in winter months.

(iii) Large aerosol concentrations are observed in the layer between surface and 150 m followed by a sharp decline between 150 and 200 m altitude. Thus, most of the aerosols are trapped close to surface. The average annual ratio of the columnar content between 50-200 m layer to the total columnar content in the 50-1100 m layer is 40%, the percentage ratio is highest in the pre-monsoon and winter seasons and lowest in the summer monsoon season.

(iv) Horizontal advection by winds play an important role in determining the particulate concentration at nighttime. With the formation of a stable layer at night, the vertical dispersal can occur only with stronger winds. The concentrations also increase with columnar relative humidity. Higher water vapour in the column changes the scattering coefficient of aerosol. Thus, day-to-day profiles of aerosol in the lower troposphere depend upon previous rainfall, winds in the surface layer and integrated column relative humidity.

(v) Based on 13 years of data at Pune (1986-1998), a rising trend of about 3% in columnar aerosol loading is noted and the superposed intra-seasonal variability due to episodic fall and rise is much higher than the slow long-term rising trend which could be
due to urbanization, land-use change in the vicinity of the station and industrialization.

2.5.2 Tropospheric aerosol studies by the SPL group at Trivandrum with lidar—The SPL group at Trivandrum have also made extensive use of their lidar facility to understand various facets of variation in distribution of vertical density of aerosols in the troposphere. The following important observations have been noted57-64:

(i) Aerosol number density profiles on individual days showed stratified layered structure. The aerosols are mostly trapped in the layer between 200 and 1200 m, the layer of high vertical stability. Significant month-to-month variations are noted in the average aerosol extinction coefficient. Like at Pune, the aerosol number density showed dependence on wind speed and relative humidity.

(ii) Mixing heights of the aerosols are maximum in pre-monsoon, monsoon and post-monsoon seasons. Maximum contents of aerosols exist in the layer surface to 400 m and about 30% of the columnar depth is contributed by the lowest 1 km. When this feature is compared with the data at Pune (an inland station) it is found that vertical mixing is higher at Trivandrum than at Pune. This could be due to the land and sea breeze systems prevailing at Trivandrum which would promote ventilation of the boundary layer and hence deeper vertical mixing.

(iii) The AODs within the boundary layer also showed an increasing trend of about 3% based on 6 years data (1989-1997), which is similar to those observed at Pune.

(iv) An analytical model was validated for the altitude profile of aerosol number density in the boundary layer. Three distinct aerosol layers were observed, namely, the well mixed (150-400 m), the middle layer (0.4-1.5 km) and the upper well mixed layer (above 1.5 km).

(v) Tropospheric vertical diffusion coefficients were determined and are found to decrease in the altitude range 5-14 km. The seasonal altitude variations of aerosols in the free troposphere are mainly governed by wet removal processes under the presence of deeper clouds.

3 INDOEX and south Asian aerosol studies

3.1 INDOEX objectives and major results

The Indian Ocean Experiment (INDOEX) was a major field experiment conceived in 1996 and conducted jointly in 1999 by several countries of Europe, India and USA with 3 inter-related objectives65:

(i) To assess the significance of continental aerosols for global radiative forcing.

(ii) To assess the magnitude of solar absorption at the surface and in the troposphere including ITCZ cloud systems, and

(iii) To assess the role of ITCZ in the transport of pollutants and their radiative forcing.

The first objective was to contribute to a better understanding of the long-term time scale climate forcing, the second to assess the direct effects of aerosol optical and radiative properties on regional scale radiative forcing and the third to assess the indirect effects of aerosols in modulating ITCZ and large scale cloud system processes. The field experiment was carried out in the northern hemisphere winter season of 1996-1999, when the winter N-E trade system of Asia is at its peak and continental aerosols are transported across the equator into the south-Indian Ocean N-W monsoon system, where they ascend into the large-scale convective cloud build-up of the southern ITCZ (mean position 8-12°S). South of ITCZ is the regime of the pristine south-east trade winds of marine origin and the region north of equator is dominated by the continental air from south Asia, east Asia and south-east Asia.

The INDOEX deployed composite observing system consisting of ship transits in the north and south Indian Ocean, the surface measurements network over India, Maldives, Mauritius and Reunion islands, constant level balloon trajectories released from Goa, and research aircraft equipped with sophisticated instruments to measure different properties of aerosols. A specially equipped climate observatory was functional at Kashidhoo island in Maldives from 1998 to 2000. Intensive field phase (IFP) was carried out during mid-February to the end of March 1999 with Male International Airport as the operational base, where over 200 international scientists participated in the interdisciplinary experiments. The integrated IFP data provided ground truth for satellite measurements of aerosols. A general circulation model with an aerosol transport model was used to assimilate the experimental data with the satellite data to produce the big picture of the trans-continenta aerosol transport. The model assimilation of the aerosols helped in deriving the regional radiative forcing of the aerosols which could then be used for estimating the regional climate forcing.
The INDOEX helped the Indian research community, derived from over 15 organizations, to work jointly with the international research teams in the study of long-range aerosol transport and its implication on the regional climate forcing and climate modulation during the winter season over India. The Indian research community has published their findings in two special volumes of the journal Current Science in 1999 and 2001. Besides several papers from India were also published in a special issue of the Journal of Geophysical Research (2001) and other international journals. The widespread pollution plume of anthropogenic origin originating from Asian region was traced in several studies \(^{66,67}\). The four main channels of the plume, which operated with different strengths and in an episodic manner, and the plume’s back and forward trajectories were analyzed by Verver et al.\(^{68}\). At the end of the experiment, UNEP formed a panel of experts to produce an integrated picture of the INDOEX results with possible impacts on regional climate, agriculture and health in the winter season over south Asia, consequent to the prevalence of the widespread polluted haze layer. Indian scientists were involved as co-authors of the UNEP reports. Major results of INDOEX from the Indian efforts were the following:

(i) Estimates of the radiative forcing over the tropical Indian Ocean with appreciable forcing at the surface (cooling) and at the top of the boundary layer (warming) \(^{64,69}\). The ratio of surface to top of the atmosphere forcing, a measure of the aerosol absorbing efficiency, was about 3 for the Indian Ocean aerosols.\(^{70}\), as top of the atmospheric radiative forcing was estimated to be about 25 Wm\(^{-2}\) and the surface forcing 75 Wm\(^{-2}\). The heating due to absorbing aerosols within the boundary layer was found to be 0.3-1.0 K which is an increase of 50-100% of solar heating in an aerosol-free atmosphere. It may be mentioned that in an earlier study by IITM group\(^{59}\), the polluted industrial complex temperature in summer monsoon situation was found to be about 1 K higher than that of the pollution-free environment in Mumbai region.

(ii) The AOD over tropical Indian Ocean due to sulphate and ammonium contributed about 29%, sea-salt and nitrate about 17%, mineral dust about 15%, and soot, organics and flyash contributions were 11%, 20% and 8%, respectively\(^{71}\). It may be mentioned that the higher contribution by sulphate and ammonium over the oceanic region may be due to emissions of sulphate (dimethyl sulphide) and ammonium from marine natural sources. Earlier studies by IITM group (discussed in Sec. 2.1-2.4) allowed for greater contributions from soil dust over India. Of the chemical constituents measured during INDOEX, nearly 30% (sea-salt, mineral dust and marine origin sulphur and ammonium) is from natural sources and remaining 70% originates from the anthropogenic sources. Black carbon (soot) is an absorbing aerosol and its role in radiative forcing is considered to be very important\(^{66,72}\). Role of absorbing aerosol like black carbon, is to deplete the short wave radiation at surface by absorbing radiation within the aerosol layer\(^{72-74}\). Black carbon is found to contribute to the anthropogenic aerosols over the INDOEX region. The following observations were also made:

(i) A model with emissions from Indian sources showed that carbonaceous and flyash aerosols are significant contributors to the radiative forcing\(^{73,76}\).

(ii) Chemical characteristics and sources of aerosols were apportioned over the Indian Ocean\(^{77}\).

(iii) Aerosol optical depth distributions from satellite retrievals over the oceanic region adjacent to south Asia for different seasons were obtained for 5 years (1996-1999). These maps showed enhanced AODs over the west Arabian Sea during the summer monsoon season, ascribed to desert aerosols from neighbouring region, and were maximum over the east Arabian Sea and the Bay of Bengal regions during winter with possible origin from anthropogenic sources of the neighbouring continental Asian region.

(iv) Higher aerosol concentrations were observed in north of the ITCZ (polluted zone) as compared to those in the south (pristine zone), as expected.

(v) Large intra-seasonal oscillations in the AOD and CO are observed at Kashidhoo which are controlled by cloud scavenging and washout processes during convective events occurring in the northern hemisphere equatorial trough (convergence zone) near Maldives\(^{68}\). The Bay of Bengal was thought to be more polluted in 1999 because of the persistent nature of an anticyclonic circulation centre over the central India and the merging of the air streams from the north Indian Ganges plain, China and S-E Asia over the central southern Bay. Sikka\(^{78}\) explained the abnormally high pollution load in IFP-1999 compared to IFP-1998 at Kashidhoo due to the persistent meteorology of the central Indian anticyclonic circulation. He also examined the interannual and intra-seasonal variabilities of AOD and
CO over Kashidhoo and found them to be quite prominent. Thus, meteorology of the area on inter-annual and intra-seasonal scale is very important and the radiative forcing would significantly modulate due to such variability.

(vi) Kamara et al. studied the north-south aerosol gradients from atmospheric electrical conductivity measurements in IFP-1999 and emphasized on large intra-seasonal variability.

4 Important issues emerging out of INDOEX results

South Asian region is becoming an important source of anthropogenic emissions. The INDOEX has shown that, in winter, about 70% of aerosol load over this region is from anthropogenic sources. Earlier studies by the IITM, IMD and other groups for the last 20 years, using primarily rain chemistry, dry deposition, solar radiation, turbidity measurements, etc., have again and again emphasized the rising trends in anthropogenic emissions over India. Similar results coming from ground-based AOD measurements and lidar-based measurements have emphasized the rising trends in anthropogenic aerosols during last 15 years or so. Thus, it should not come as a surprise that the regional atmosphere over India is taking the burden of increased urbanization, transportation and industrial activities. Reduction of solar radiation by about 15%, as estimated in earlier Indian studies and as obtained from INDOEX is quite large as the global average from 1958 to 1985 is only 5%. India is a developing economy with a large population struggling to enhance the living standards. Anthropogenic activities have increased in the last 2 decades and would increase further. Therefore, the results of the previous aerosol measurements over India and their extension across the neighbouring ocean and land regions due to INDOEX should not be surprising at all, as the meteorology of the winter season supports the accumulation of aerosols from different source regions (as the light winds and stable conditions prevail over central and northern India). Their long-range transports are also facilitated by the persistent N-E trends prevailing over the Arabian Sea and the Bay of Bengal. The widespread Asian haze is a result of continental emissions as well as natural dust and sea-salt. The meteorology of the area helps in its being quasi-persistent over the region in winter season. The burden of winter haze (of anthropogenic as well as natural origins) is a fact of life and well validated by the INDOEX observations and several years of satellite data. Its abnormal thickness over Kashidhoo (Maldives) in the winter of 1999 was due to abnormal meteorology of the area which inhibited ventilation of the atmospheric boundary layer. The INDOEX results have, for the first time, emphasized the potential impact which this widespread haze could play in modulating the winter seasons climate over the south Asian region, primarily through changing the north-south and inter-hemispheric gradients in AOD resulting in changes in heating gradients. The potential climate modulation could arise due to a loop of physical factors, starting from distributing the regional radiative balance (cooling of the surface and slight warming at top of the boundary layer) and decrease in regional evaporation, which may have impact on the regional hydrological cycle. The major culprit in this scenario has been found to be the black carbon aerosols which are chiefly due to emissions from transport, burning of industrial fuels, use of biofuels and biomass burning. Biofuels in the house-hold have been the traditional survival fuel in south Asia and cannot be taken to be luxury fuel. Biomass burning is not likely to be widespread in India as the bulk of agricultural biomass is used as fodder for cattles, though there could be some upward trend, after the green revolution, to direct part of the agricultural biomass (rice husk) to small-scale industrial sector for use as fuel. Use of diesel in the transport sector has increased to carry the load of increasing industrial economy, though measures have been taken to reduce subsidy on diesel and also use of CNG in city transport system and natural gas for industrial and power production sectors. Such technological adoptions are likely to reduce black carbon emissions in the atmosphere in the next decade, which could get somewhat upset by increasing urbanization and industrialization. This requires proper monitoring.

The evaluation of impact of haze cloud on regional winter climate has been based on numerical experiments using different GCMs without the haze layer and with the observed haze layer (including the absorbing aerosol). These experiments show one robust result in terms of substantial increase in rainfall in the near-equatorial region of the northern and southern ITCZ (15°S to 15°N) and decrease in rainfall over the north-western India, Pakistan and adjoining Afghanistan. The experiments also indicate changes in the gradient of temperature at surface and up to
1km with decrease of subsidence over central India and decrease of surface pressure and northward shift of ITCZ over about 5° latitude. These experiments are very preliminary in nature and more experiments are needed to establish the results. There is also a necessity to establish statistical significance of the results by making model runs for several decades and using ensemble technique to isolate the model variability (unperturbed and perturbed runs) in winter rainfall over south Asia. The natural variability of winter rains is quite high (40%) on inter-annual scale. Research in the next few years could be devoted to provide scientific evidence about the modulation in south Asian winter season rainfall due to extensive haze layer. Winter and summer monsoon droughts are part of the natural variability of south Asian climate and their occurrence cannot a priori be ascribed to the widespread haze. In fact, south Asian summer monsoon data of nearly 150 years show that there is a multi-decadal and inter-annual variability which has produced higher (lower) incidence of drought (excess rains) years and vice versa in the non-industrial and pre-industrial era of India between 1860 and 1980. In fact, in the industrial era of India commencing from 1980 to the present, only 3 drought years (1982, 1987, 2002) and two excess summer monsoon years (1983 and 1988) have occurred and their incidence is more closely linked to ENSO-Monsoon connections rather than growing haze in winter or pre-monsoon season. Therefore, it is rather premature to overemphasize the potential impact of haze layer in modulating the south Asian climate in winter or summer monsoon season, though the subject needs further examination. Observational data would not be able to support the findings, based on models, due to large inter-annual variability in rainfall regimes arising out of natural factors. Modelling, no doubt, provides a tool to suggest probability of haze layer modulating the climate and such a result, if confirmed in a rigorous scientific manner, could be used to enlighten the policy maker about the potential implications of haze layer.

5 Future strategy for tropospheric aerosol monitoring and research

In view of the potential hazards of haze layer, it is essential for countries in south Asian region, particularly India which is the biggest and also more industrialized in the region, to establish a good monitoring network for tropospheric aerosols over its different climatological, urban and rural environments. The programme may be jointly supported by different Governments with the following major objectives:

(i) To monitor the physical characteristics of aerosols at a network of surface and lidar stations in different climatological environments to assess the large spatial variability and inter-annual and intra-seasonal variability. Variations in north-south gradients in winter and east-west gradient in pre-monsoon and monsoon seasons are large.

(ii) To monitor the chemical conditions of aerosols, including black carbon load, at surface stations and, if possible, by taking grab samples by aircraft probes.

(iii) To use satellite aerosol measurements for producing large-scale space-time distribution of aerosols over the south Asian regions following the method developed by Rajeev et al.

(iv) To use the data on monthly and annual basis for assessing the inter-annual variability of aerosol radiative forcing.

(v) To continue monitoring the chemical constituents of rainfall and dry deposition of a network at stations and visit experimental (campaign mode) stations once in 5 years for assessing the changing scenario.

(vi) To develop and adopt numerical models for long-range transport of aerosols and also prepare back trajectories of episodic high concentration plumes in all seasons on year-to-year basis, so that the source regions of trans-boundary plumes and the role played by meteorology of the region could be examined.

(vii) To run global circulation models (atmosphere and ocean-atmosphere coupled models) on multi-decadal basis for summer monsoon with and without haze layer of April-May, which is the season of highest AODs in view of widespread natural dust load (possibly mixed with anthropogenic emission), for assessing the impact of dust load variability in the pre-monsoon season on the ensuing summer monsoon rainfall.

(viii) To launch national field campaigns during different seasons over the Indian seas for aerosol physics and chemistry measurements and for providing ground truth of satellite monitoring. Satheesh undertook a campaign over the Bay of Bengal in March 2001 and found the AODs to be as large as those estimated in INDOEX phase.

(ix) To establish data archival and distribution procedures.
The Department of Space had constituted a committee to propose a national programme on aerosol and radiation budget studies (ARBS) and the committee’s report has been adopted for a phased implementation under their Geosphere-Biosphere Programme. The IMD is also having a plan to enhance their network for aerosol monitoring. The National Physical Laboratory, New Delhi and the Physical Research Laboratory, Ahemdabad, under the guidance of Dr A P Mitra have also included some more stations (Port Blair, Darjeeling, Leh and other places) for aerosol physics and chemistry measurements. A co-ordinated programme on tropospheric aerosols with components on observations (monitoring and data archival) and modelling (theoretical and numerical) must be formulated.

Climate related issues are becoming increasingly relevant to several socio-economic purposes. In future, inter-state as well as international politico-economic systems would be linked to intervene in the natural climate system by human activities. South Asia is one of the most rapidly developing regions in the world with high population growth and low per capita income. Its consumption of energy is bound to rise in the next 2-5 decades. Its economies still hinge on the bounty of the monsoon rains. There is an overriding priority for economic development and the model chosen for development is (after the western Europe and north America) through industrialization. Such a developmental pathway may have profound impacts through the ways sources and sinks are used. Thus, human-induced changes in every aspect of the environment are expected to occur. Their consequences could appear on natural resources like climate and, hence, on human welfare too. Since it is imperative that development must continue, it is urgent that a comprehensive and integral programme is undertaken which includes the systematic monitoring of physical and chemical nature of the regional monsoon system.

Developing societies like India have so far contributed, in a very small measure, to global change and hence, at this early stage, nations in the south Asian region must strengthen their hands by sound quantified scientific information about their environment to counteract arguments against their rapid development as well as to make mid-way corrections to policies for energy and technology options. Even though monsoon system is driven by powerful natural mechanisms and the south Asian summer monsoon is one of the most stable systems of the global climate system in terms of its arrival and total seasonal performance, it has substantial inter-decadal and inter-annual variability. The possible impact of human-induced changes may appear small and uncertain at this stage, but one cannot guarantee that the natural climate system may not abruptly get affected and give a surprise to the fragile national economic systems of the regions. Environment-human interaction with monsoon climate has multiscale interacting loops. Many of the environment-human interaction loops are linked processes. Therefore, ecosystem adjustment with the regional climate system under development efforts must be assigned high priority in India, as monsoons have been continuing and shall continue to be the main stay of the ancient civilization of this country.

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