Polar aerosol characterization, sources and impacts

S. M. SONBAWNE, P. C. S. DEVARA*, R. C. REDDY, P. D. SAFAI and P. S. SALVEKAR

Indian Institute of Tropical Meteorology, Dr. Homi Bhabha Road, Pune - 411 008, India

*e mail : devara7@gmail.com

सार – ध्रुवीय क्षेत्रों के मौसम, जलवायु एवं उनके ध्रुवीय सतह–वायुमंडलीय प्रणाली के विकिरण संतुलन विशेषरूप से उच्च पृष्ठ–परावर्तनीयता परिस्थितियों द्वारा क्षेत्रों के लक्ष्णीकरण में वायुविलयों के महत्वपूर्ण प्रभावों के कारण जाना जाता है जिसमें वायु विलयों का विष्मजातीय रसायन प्रचलित है। अतः प्रकाशीय एवं कालिक मापक्रम दानों पर वायुविलय के भौतिकीय एवं प्रकाशीय गुणधर्मों के ज्ञान को विकसित करने की जरूरत है। इन भौतिकी–रसायनिक एवं प्रकाशीय गुणधर्मों की विशेषताएँ जानने हेतु ग्रीष्म ऋतु के दौरान 24वें (2004–2005) तथा 26वें (2006–2007) भारतीय एंटार्कटिका अभियान एवं 2010 में आयोजित 14वें भारतीय एंटार्कटिक अभियान के दौरान दोनों ध्रुवीय क्षेत्रों ख्अंटार्कटिका, 'मैत्री' (70.76 डिग्री द., 11.74 डिग्री पू.), आर्कटिक हिमाद्री (79 डिग्री उ., 11 डिग्री पू.) पर उध्ययन किए गए मल्टी–चैनल सोलर–रेडियोमीटर (माईक्रोटोप्स II) का प्रयोग करते हुए कुल स्तंभीय वायुविलय प्रकाशीय गहराई (ए.ओ.डी.), ओजोन (टी.सी.ओ.), अवक्षेपणीय जल मात्रा (पी.डब्ल्यू.सी) एवं प्रत्यक्ष विकिरणीय बलन एवं उनकी लाक्षणिकताओं के लिए वाइड–बैंड पाइरानोमीटर का प्रयोग करते हुए लघु–तरंग ग्लोबल विकिरणीय पलक्स। आर्कटिक में, वायुविलयों के रासायनिक गुणधर्मों को निर्धारित करने हेतु एक एंडेरसन सैम्पलर. ब्लैक कार्बन एथैलोमीटर का भी प्रचालन किया गया।

वायुविलय प्रकाशीय, भौतिकीय एवं विकिरणीय गुणधर्म एवं उनके परस्परूप से मापित गैसों के साथ अंतरापृष्ठ एवं उनके रासायनिक घटकों की जाँच की गई। परिणामों से यह दर्शाया है कि 500 एन. एम. की लाक्षाणिक तरगदैर्ध्य पर दैनिक माप ए.ओ.डी. 0.042 औसत एंगस्टॉर्म गुणांक के साथ 0.24 सहित पाया गया, एंटार्कटिका में कोर्स—मोड कणों की प्रचुरता प्रकट होती है, तथा फाइन—मोड कणों को दर्शाते हुए आर्कटिक औसत ए.ओ.डी. 2.84 के साथ एंगस्टॉर्म गुणांक औसत पर 0.11 प्रेक्षित किया गया। सतह—आधारित ओज़ोन मॉनीटर द्वारा मापित टी.सी.ओ. जो कि कुल ओज़ोन मैपिंग स्पेक्ट्रोमीटर (टी.ओ.एम.एस.) सैटेलाइट सेन्सर के द्वारा मापित 5 प्रतिशत के भीतर समुचित रूप से संगत था। अध्ययन अवधि के दौरान दैनिक मापक्रमों पर ओज़ोन में परिवर्तनीयता, एंटार्कटिका क्षेत्र पर 4 प्रतिशत से कम थी एवं आर्कटिक पर भी कमोवेश उतनी ही थी।

जनवरी 2005 में पाए गए फ्लक्स फरवरी 2005 की तुलना में लगभग 20 प्रतिशत कम थे। वायुवियलों के कारण औसत लघु—तरंग प्रत्यक्ष विकिरणीय बलन ने अध्ययन अवधि के दौरान सतह पर —0.47 डब्ल्यू ⁄ एम² के औसत मान की शीतलता दर्शायी। इस शोध पत्र में, प्रयोग किए गए उपस्कर, डाटा अभिलेख तथा उनकी विशलेषण तकनीकों तथा भारतीय ध्रुवीय स्थानों, 'मैत्री' एवं 'हिमाद्रि' पर प्रमुख परिणामों की हमनें सक्षिप्त रूप से विवेचना की है।

ABSTRACT. Aerosols are known to cause important effects on weather and climate of Polar Regions and their radiation balance of the polar surface-atmosphere system, especially in the regions characterized by high surface-reflectance conditions, which also prevails the heterogeneous chemistry of aerosols. Therefore, the knowledge of the aerosol physical and optical properties needs to be improved on both spatial and temporal scales. To characterize these physico-chemical and optical properties, studies have been carried out over both the polar regions [Antarctica ('Maitri' (70.76°S, 11.74°E) and Arctic "Himadri" (79°N, 11°E) during the summer period of 24th (2004-05), 26th (2006-07) Indian Antarctica Expedition, and during 14th Indian Arctic Expedition in 2010. Total column aerosol optical depth (AOD), ozone (TCO), precipitable water content (PWC), and direct radiative forcing using a multi-channel solar-radiometer (Microtops II); and short-wave global radiative flux using a wide-band pyranometer for their characteristics. In the Arctic, an Andersen Sampler, Black Carbon Aethalometer was also operated to determine the chemical properties of aerosols.

The aerosol optical, physical and radiative properties, and their interface with simultaneously measured gases and their chemical composition have been investigated. The results showed that the daily mean AOD at a characteristic wavelength of 500 nm was found to be 0.042 with an average Angstrom coefficient of 0.24, revealing abundance of coarse-mode particles in Antarctica, and Arctic average AOD was observed 0.11 with an average Angstrom coefficient of 2.84, suggesting fine-mode particles. The TCO measured by the surface-based ozone monitor matched reasonably within 5% with that of the Total Ozone Mapping Spectrometer (TOMS) satellite sensor. Variability in ozone on daily scale, during the study period, was less than 4% over the Antarctica region and more or less same for Arctic.

The January 2005 fluxes were found to be less by about 20% as compared to those in February 2005. The average short-wave direct radiative forcing due to aerosols showed cooling at the surface with an average value of -0.47 W/m^2 during the study period. In this paper, we briefly describe the equipment deployed, data archival, their analysis techniques and salient results obtained over the Indian polar stations, 'Maitri' and 'Himadri'.

Key words - Polar region, AOD, Angstrom coefficient, Aerosol radiative forcing.

1. Introduction

The aerosols, and pre-cursor gases over the polar region play great role not only in the Earth's radiation budget but provide reference levels for all the environmental and pollution studies due to their remoteness and restricted human activities. The attenuation of solar irradiance and the processes of scattering and absorption by the aerosol particles may cause appreciable effects on the weather and climate of the Polar Regions. The importance of aerosols on climate forcing has been very well established (Penner et al., 2001; Ramanathan et al., 2001). Aerosols can change Earth's radiation budget both directly by scattering and absorbing radiation and indirectly by affecting cloud properties (Russell et al., 1999). Changing the net flux of radiation above or within the atmosphere alters the energy available for driving the Earth's climate. Hence such a net flux change is termed as radiative forcing of climate (IPCC, 1995). Particles in the atmosphere scatter the incoming solar radiation and thus reduce the radiation flux reaching the Earth (dimming). This is known as 'direct radiative effect' and it accounts for negative forcing. Negative forcing tends to cool the climate & positive forcing tends to warm it. Aerosol particles, particularly those with high black carbon (BC) content, are capable of absorbing incoming solar radiation. While this also decreases the radiation flux at the surface, it heats the atmosphere locally. This heating can, in turn, cause a third climate forcing effect known as the 'semi-direct radiative effect'; the heating of the atmosphere can suppress the local relative humidity (RH) and there by reduce the change of cloud formation.

The polar-regions play an important role in global change studies because enhanced warming is predicted for high-latitudes as greenhouse gas and aerosol concentrations continue to increase in the atmosphere. Aerosols and pre-cursor gas observations over polar region play great role not only in the Earth's radiation budget but provide reference levels for all environmental and pollution studies due to its remoteness and restricted human activities. Thus, in this region, while the total mass of the aerosol particles suspended in the vertical atmospheric column of unit cross section yields smaller values, the lower aerosol optical depth at all the visible and near-infrared wavelengths is of great concern for Earth's radiation budget studies. The attenuation of solar irradiance and the processes of scattering and absorption by the aerosol particles may cause appreciable effects on the radiative exchange mechanisms occurring in the polar atmosphere. Hence the aerosol properties and their effects on weather and climate of this region are certainly different from other continental areas. Moreover, the study of heterogeneous chemistry of aerosols - a unique phenomenon of Polar Regions, provides information on gas-to-particle conversion processes leading to formation of secondary aerosol particles and their influence on ozone content. Studies have shown that the warming can be nearly 4-5 times more over the poles compared to that over tropics. Thus the atmosphere over poles is very sensitive to human-induced changes in any part of the globe and dynamics of the atmosphere over poles itself seems to modify global weather and climate. Two important problems or concerns of the present-day scientific community are the ozone hole and the aforementioned global greenhouse warming; the former already having manifested prominently over polar region during the past decade (WMO, 2003), and the latter having started showing indications of its effects at mid and polar latitudes. Thus there is a need to continuously keep monitoring of the atmospheric constituents over these sensitive Polar Regions.

Ever since the establishment of Indian stations, Dakshin Gangotri in 1983-84 and Maitri in 1988-89 at Antarctica, Indian scientists have been very much involved in Antarctic/Arctic research (Rajaram and Reddy, 1993). Several summer and winter expeditions have been undertaken since the beginning where atmospheric scientists have taken active part in monitoring and studying atmospheric constituents (Gadhavi and Jayaraman, 2003; Jain *et al.*, 2004). Within the frame work of the $24^{\text{th}} \& 26^{\text{th}}$ Indian Antarctica Expedition (IAE) and 14th Indian Arctic Expedition, we have carried out observations of total column aerosol optical depth (AOD), ozone (TCO) and precipitable water content (PWC) using a multi-channel solar-radiometer (Microtops II); and short-wave global radiative flux using a wideband pyranometer over the Indian Antarctica station 'Maitri' (70.76° S, 11.74° E) and In addition, Aethalometer for Black carbon and Andersen sampler were used for aerosol chemical compositions.

The present paper essentially deals with measurement of optical, physico-chemical and radiative properties of aerosols and pre-cursor gases, and estimation of direct aerosol radiative forcing and heterogeneous

587

chemical effects of aerosols in the polar region. In order to investigate these aspects, extensive field experiments have been undertaken at Indian Antarctica station, Maitri and Indian Arctic station, Himadri to characterize aerosols by means of using above said observations. Such studies are very sparse and almost non-exist over the Polar station, and hence the proposed measurement would help bridging this gap to certain extent. The experimental and data details along with other interesting results obtained will be presented.

2. Data

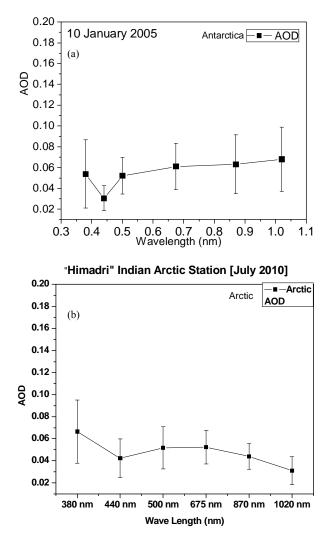
A compact, hand held ozonometer (Microtops-II), which utilizes direct solar light observations at discrete wavelengths from VIS - NIR regions to compute total column aerosol optical depth, ozone, water vapour and short-wave global radiative fluxes using a wide band pyranometer, In addition, to this Anderson sampler for study the chemical compositions and the Aethalometer was used to study the loading of Black carbon over Arctic station. Data over the both polar region has been collected and continuous observations were made at the Indian station 'Maitri' (70.76° S, 11.74° E) during the southern summer months of 24th IAE (2004-05) & 26th IAE (2006-07) and also observation were taken at 14th Indian Arctic station 'Himadri' during the 14th Expedition (2010). These extensive data sets, collected over both polar region during above said expeditions have been utilized to investigate the aerosol optical, physico-chemical, and radiative properties, and their interface with simultaneously measured gases such as total column ozone, water vapour and surface level meteorological parameters.

3. Measurement location & techniques

Observations were carried out on both Polar Regions at the Indian station. Indian Antarctica Station, "Maitri", is located in the Schirmacher Oasis in the Dronning Maud Land, East Antarctica (117 m msl). The nearest steep cliff of the east-west trending glacier on the southern side of the station is more than 700 m away from the station and is 300 m (amsl) in height. Where as Indian Arctic Station "Himadri" situated at Ny-Aalesund, Spits Bergen, (79° N, 11° E) is a small Norwegian settlement focused on scientific research in the Arctic, it is located about 1000 km off the North Pole and mid way on a straight line between Kiruna, Sweden, the base of SOLVE, and the Pole. The snow covered surface during summer season was more than 0.5 km away from the station at both poles. The instruments were installed on barren land near the station. The surface of the station area is mainly covered by sandy and loamy sand types of soil (Panneerselvam, et al., 2004). Antarctica has a desert like climate with clear skies, very low atmospheric aerosol content. The cloud cover over the station occurs mainly under the influence of sub polar low-pressure systems and shows an alternating sequence of clear sky changing over to the overcast and again clearing as the system moves away.

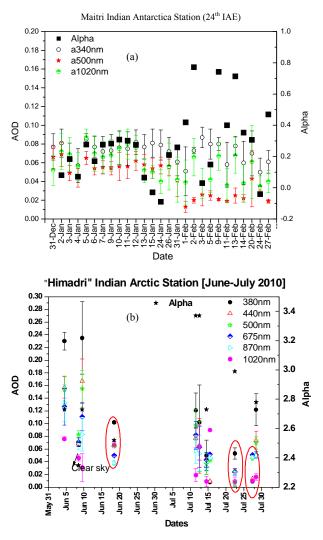
Multi-channel solar-radiometer (Microtops II) & Pyranometer operated at both the poles. In addition, Aethalometer and Andersen sampler were operated at Arctic Indian station. Multi-channel solar-radiometer (Microtops II) measures the columnar AOD and Ozone, water vapor at discrete wavelengths which in turn provide the attenuation in sunlight reaching the ground due to the presence of aerosol pollutants. This radiometer provides integrated AOD (extinction) at six wavelengths covering form UV to NIR and ozonometer determines total column ozone (at UV wavelengths) and perceptible water content (at NIR band) simultaneously. The built-in solid state pressure sensor provides the current atmospheric pressure needed for calculation of scattering due to air molecules (Rayleigh). The global positioning system GPS receiver provides the geographical coordinates of the site, which are used for estimating the local air mass. When the instrument is switched on, the radiation captured by its collimators pass through the band-pass filters and falls onto the photodiode. These signals are amplified and at the same time they store the background value for all the filters (sets of over 25 observations for each filter). The average value, obtained for each filter is used to compute the spectral variation of columnar AOD, ozone and water vapor instantaneously and display for quick look and the pyranometer used in the present study to measure direct and diffuse shortwave radiation (i.e., total radiation) with an un-obstructive view. This has a circular multi-junction thermopile of the plated (copper-constantan) wire wound type, which is temperature, compensated in order to get a response that is independent of ambient temperature. The temperature sensed by the detector is nearly linear with the flux density of incident radiation. The instrument is supplied with a pair of removable precision ground and polished concentric hemispheres of Schott optical glass. The inner hemisphere is transparent to a wavelength of 0.28-3.0 µm. The outer dome can be replaced by Schott glass hemispherical filters which transmit within specified bandwidths. Desiccant is placed in the side of the instrument to absorb humidity inside the glass domes. An Aethalometer was also operated at Indian Artic station for studying the Black Carbon aerosol concentration.

Data were archived during different sky conditions. In view of complexity involved in the data analysis procedures, observations collected during clear-sky conditions only were utilized in the present study. Calibration of both photometers was performed by a transfer of calibration constants from reference instruments which were calibrated by the Langley plot technique at a noise-free high-altitude site. The AOD,



Figs. 1(a&b). Wavelength dependence of AOD for (a) Antarctic and (b) Arctic region

TCO and PWC were evaluated from the data reduction. An important advantage with this radiometer as compared to many complementary instruments lies in its portability and on-line data acquisition and analysis. This enabled us to obtain simultaneous estimates of aerosol and pre-cursor gaseous optical depths instantaneously. More details about observational scheme, calibration and data retrieval procedures for this radiometer have been published in the literature (Devara et al., 2001; Ichoku et al., 2002). In addition, short-wave global radiation fluxes using a wideband pyranometer have been carried out. By combining pyranometer and sun-photometer observations, direct radiative forcing due to aerosols (Maheskumar and Devara, 2004; Devara and Sonbawne, 2008) on clear-sky days has been computed for Antarctica region. These extensive data sets, collected during expedition, have been utilized to investigate the aerosol optical, microphysical and radiative properties, and their interface with



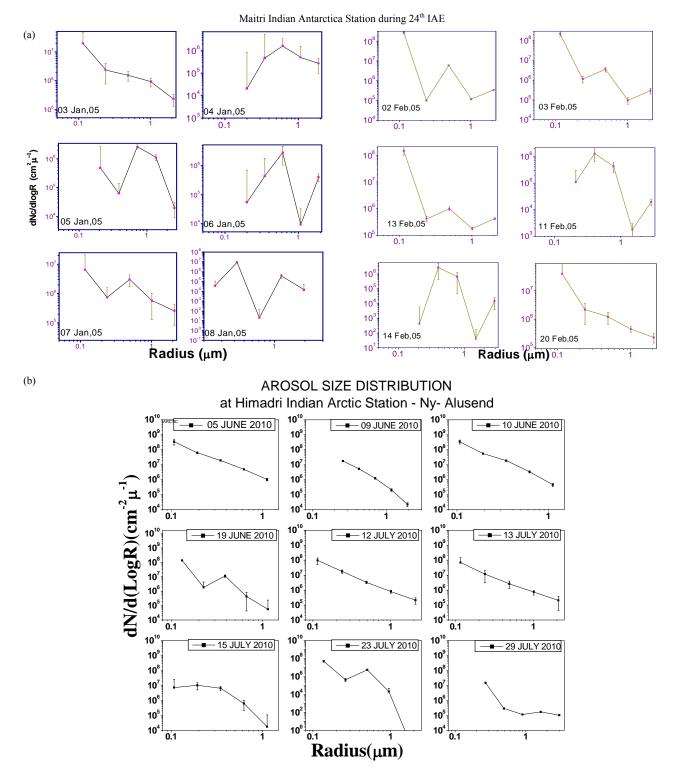
Figs. 2(a&b). Day-to-day mean variation in AOD and Alpha over (a) Maitri and (b) Himadri Indian station

simultaneously measured gases such as total column ozone, water vapor, and surface-level meteorological parameters.

4. Results and discussion

4.1. Spectral characteristics of aerosols

Figs. 1(a&b) depicts the spectral distribution of AOD observed on typical clear-sky days over both poles. These plots reveal a common feature; AOD initially decreases from 340 to 440 nm wavelengths and then slightly increases with increasing wavelength. This implies that loading of lager particles dominates that of the sub-micron particles in the atmosphere. This is also consistent with the observed Angstrom exponent (Alpha) values (indicator of aerosol size distribution), which are discussed further in the next Section. This could be due to

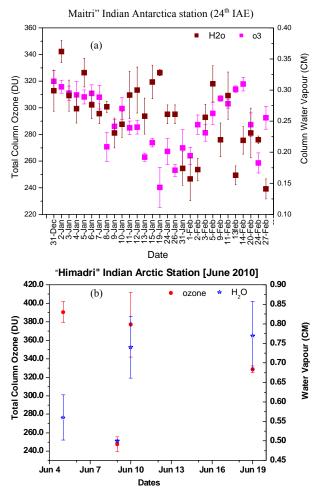


Figs. 3(a&b). Aerosol size distribution observed on some typical days during the study period over (a) Maitri and (b) Himadri Indian station

influx of marine and continental (natural) aerosols that prevails over the experimental site through transport phenomenon. Day-to-day variations in AOD at all wavelengths mainly on 340, 500 and 1020 nm, representing fine, submicron and larger aerosol particles, together with Angstrom exponent (Alpha) observed during clear-sky conditions during Antarctica and Artic Indian Expeditions are shown in Figs. 2(a&b). The daily mean AOD at 500nm was found to be 0.042 in Antarctica and 0.11 in Arctic with an average Angstrom exponent of 0.24 and 2.84 respectively, revealing most of the time the abundance of coarse-mode particles dominate over Antarctica and Arctic region. It is interesting to note from the [Fig. 2(a)] that January AODs are higher as compared to those of February with smaller values of Alpha indicating dominance of larger particles (coarse-mode) in January and larger values of Alpha revealing the dominance of smaller particles (fine-mode) in February and in Arctic the same feature was also observed [Fig. 2(b)] while in June AODs are higher as compared to those of July, with lower values of Alpha indicating dominance of larger particles (coarse-mode) in June and lower values of Alpha revealing the dominance of smaller particles (fine-mode) in July, respectively.

4.2. Aerosol size distribution

The aerosol size spectra, retrieved from the wavelength dependence of aerosol optical depth, have been obtained by following the constrained linear inversion method. The aerosol size distributions, computed on a few typical clear-sky days for both the poles are shown in Figs. 3(a&b). These distributions, on an average, exhibit bi-modal and tri-model distributions during the study period at both poles. As evidenced from the figure, the size distributions exhibit almost equal dominance in accumulation and coarse modes, and reveal distinct difference but similar feature in both the poles during observation periods. In February, besides dominance of coarse-mode particles. tri-modal distribution (each mode representing different sources & size of particles) with a primary mode at 0.1 - 0.6 micrometer and secondary mode at 0.6 to 1.0 micrometer, and third mode of particle size at 1.0 to 1.2 micrometer (giant mode) may be noted at both the pole. It is also noted that the smaller size particles are more at Arctic region compared to Antarctica, whereas most observational days shows mono or bi-model distribution over Arctic region. This may be due to long-range transport of pollutant over the Arctic region from the neighboring countries, thereby the industrial smoke and getting transported over this region compare to Antarctica as it is very far away from such countries. So the transportation of smaller particle is very less and this region is mainly dominated by naturally origin aerosols. The formation of primary mode particles is considered to be due to gas-to-particle conversion and photochemical reaction processes while that of coarse-mode particles is ascribed to the water bubble bursting (white caps) process

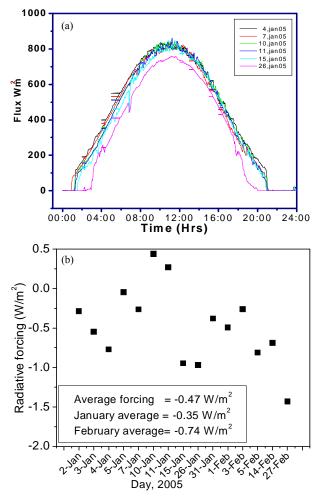


Figs. 4(a&b). Daily mean variations in TCO and PWC over (a) Maitri and (b) Himadri Indian station

over oceanic and wind-blown dust from continental regions as both the station situated at the coast.

4.3. Temporal variations in Total Column Ozone (TCO) & Precipitable Water Content (PWC)

In order to illustrate the nature of variation in ozone & precipitable water content at Antarctica & Arctic, high resolution data collected on a typical clear-sky days and their daily variations are examined. In this context, our earlier results obtained over Antarctica region (Sonbawne *et al.*, 2009; Ernest Raj *et al.*, 2009) are compared with those obtained over the Arctic region in Figs. 4(a&b). The daily mean variations in total column ozone (TCO) and precipitable water content (PWC) are found to exhibit an inverse relationship on particular days associated with low temperature [(Figs. 4(a&b)] at both poles. At Arctic region in the month of June (early-summer period), the ozone and water vapor concentration showed higher (from ~247 to 390.5 DU with an average value of 355 DU) and (0.50 to 0.77 cm with an average value of



Figs. 5(a&b). (a) Diurnal variation in radiant flux on different experimental days over Maitri and (b) Variations in daily average radiative forcing over Maitri

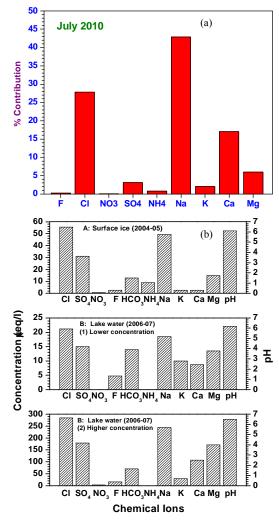
0.64 cm), which is considered to be due to strong convective activity and associated photochemical activity. Where as at Antarctica in the month of January (midsummer period), the ozone concentration showed high (~255 to 280 DU) and low values of PWC (0.14 to 0.36 cm), which could be due to strong convective activity and associated photochemical activity at both poles as the atmospheric condition is more or less similar. As we approach towards winter period *i.e.*, in February, the ozone concentration decreases (between 220 and 310 DU) with increasing PWC from 0.14 to 0.33 cm. The same feature was observed at Arctic station. This could be attributed to the relationship resembles that of heterogeneous chemical processes wherein aerosols act as catalysts in the ozone destruction mechanisms, resultant reduction in ozone amount. During the total observation period at Antarctica Indian station, TCO and PWC are found to vary from 240 DU to 320 DU with an average value of 289 DU, and the variability of ozone is found to be 4% and the observed ozone was showed good agreement with the TOMs data while the surface measurements slightly overestimating, but within 6% deviation. At the same time PWC variation is 0.14 to 0.36 cm with average value of 0.26 cm, respectively. It is found that the Arctic ozone concentration is higher than Antarctica ozone concentration. As ozone-hole phenomenon is more active in Antarctic regions than Arctic regions.

4.4. Radiative forcing

In the present study, the radiative forcing only over Antarctica is discussed here. In Arctic forcing was not calculated due to cloudy condition the radiation data was not useful to compute forcing during the observation period. Radiative forcing was calculated at 500nm wavelength for Antarctica region, combining the AOD data from the Microtops II sun photometer and concurrent radiative flux measured by the pyranometer. Fig. 5(a) shows the diurnal variation in radiation fluxes observed on some typical days, which reveal bell-shape with maximum flux around local noon and minimum around early morning and late evening hours. The decrease in flux in the presence of clouds may be seen very clearly from the figure. Such observations will be useful to discriminate the clear-sky AOD from cloudy-sky AOD. Fig. 5(b) depicts day-to-day variation in surface radiative forcing. These variations in radiative forcing reveal that the forcing swings between positive (warming) and negative (cooling) during January, while it is negative during February, which is considered to be due to influence of local meteorology and also due to long-range transport of aerosol loading. The forcing values are found to vary between -1.44 W/m^2 and 0.44 W/m^2 with an average value of -0.47 W/m^2 during the period of study (Devara et al., 2011. These values are found to be consistent with reported values in the literature, and have strong bearing on changes in surface albedo and Sun-Earth geometry during the experimental period.

4.5. Chemical properties of aerosols and snow

The aerosol samples collected using the multi-stage Andersen aerosol mass-size spectrometer were analyzed for chemical composition of aerosols over the "Himadri" site. Fig. 6(a) shows percentage distribution of different radicals to the total aerosol volume. The results reveal (*i*) alkaline nature (pH = 5.85) of water soluble extract of aerosols indicating more influence of natural sources on the chemistry of aerosols, (*ii*) clear dominance of marine influence with 77 % contribution from Na, Cl and Mg, which was followed by about 17 % contribution from Ca whereas; SO₄ and NO₃ contributed only about 3 and 0.1 % respectively indicating very weak influence of anthropogenic activity.



Figs. 6(a&b). (a) Chemical composition of aerosols over Himadri, an Indian Arctic Station and (b) Average chemical composition of surface snow and lake water collected at different locations in Antarctica during 24th and 26th IAE

In Antarctica, for the chemical composition of surface snow and lake water samples collected at different locations during southern summers of the years 2004-05 (24th IAE) and 2006-07 (26th IAE). The Fig. 6(b) shown the concentration of chemical species, Chemical composition of surface ice reveals that concentration of Na⁺ is maximum (~49µeq/l) among the cations followed by Mg²⁺ (~15µeq/l) with variations of ~22-72µeq/l and ~ 4-45µeq/l respectively. Concentrations of Ca²⁺ and K⁺ are very small or nearly equal. Among the measured anions, Cl⁻ is dominant with concentration level nearly equal to 56µeq/l and it varies between ~25-82µeq/l. Considerable amount of SO₄²⁻ (~31µeq/l) is also found in the samples with variation between ~10-65µeq/l and the concentration of the other measured chemical ions is found to be very

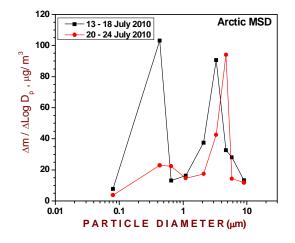


Fig. 7. Mass-size distribution over "Himadri" Indian Arctic Station

small. Sodium and chloride ions may show high concentration because of their being major sea components. The source of appreciable concentration of sulphate may be due to DMS, which is mainly produced by marine phytoplankton, which further oxidized and finally becomes sulphate particles. Dominant chemical ions in the lake water samples-1 [Fig. 5(b)] are Cl⁻ and Na⁺ with average concentration of 21 and 18µeq/l respectively. Concentration of SO42-, Mg2+, K+ and Ca2+ are ~15, 14, 10 and ~9µeq/l respectively. All though average concentration level of individual chemical ions in lake water samples-2 [Fig. 5(c)] is very high as compared to samples-1, also the dominant ions in these samples are Cl⁻ (284 μ eq/l) and Na⁺ (244 μ eq/l) followed by SO₄²⁻ (180 μ eq/l), Mg²⁺ (171 μ eq/l) and Ca²⁺ (107 μ eq/l). Comparing of the two groups of lake water samples on the basis of ratio values between the average values of individual ions, it is found that the ratio value for major ions like Cl⁻, SO_4^2 , Na⁺, Ca²⁺ and Mg²⁺ comes between 12 and 13. Thus it can be inferred that the chemical composition of the samples from both the groups is nearly same. Low pH values and low chemical contents in the lake water samples-1 it may be due to the absence of sufficient mineral neutralizers because these lakes are fed by glaciers, whereas comparatively high pH and high chemical contents in the other lake water samples it may be due to collection of these samples from lakes being fed from inlet run-off water and dissolving ample mineral dust in them. Three times increase in the concentration of K⁺ $(29\mu eq/l)$ is also attributed to the burning of waste material (like wood, waste paper and wastage food).

4.6. Mass-size distribution

Fig. 7 displays mass-size spectra of aerosols observed over Arctic on two typical experimental days in July 2010. The spectra clearly indicate bimodal

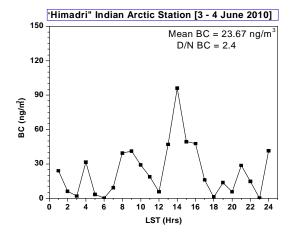


Fig. 8. Daily mean variations in BC loading at "Himadri" during June 2010

distribution with a peak in fine as well as in coarse size. During 13-18 July 2010, fine particles were dominant (58%) whereas, during 20-24 July 2010, coarse particles dominated (54%).

4.7. Black Carbon (BC) aerosol loading

Fig. 8 portrays the temporal variation of BC. It shows, higher loading of BC of about 90 ng/m³ in the atmosphere during the afternoon hours from 12:00 to 16:00 hrs (local time) with a mean value of BC of 23 ng/m³ which is about 2 orders of magnitude less than that over a tropical station such as Pune (mean Pune BC = 3.4 microgram/m³) and the day time BC is 2.4 times more than that in the nighttime at Ny-Alusend region. The day-to-day mean variation in BC at Indian Arctic station showed concentration of 30.54 ± 11.03 ng/m³during June and 55.65 ± 62.60 ng/m³ during July 2010. The range of BC concentration observed to vary from 5.63 ng/m³ to 260.21 ng/m³.

5. Summary and conclusions

Aerosols and pre-cursor gases are of considerable interest in atmospheric sciences, mainly due to their potential of changing the radiative transfer of solar radiation. As the remote polar region are characterized by the highest surface albedo and one of the less illuminated by solar radiation for a major time period during the year, detailed studies relating to characterization and impact of these constituents are highly essential. Moreover, the Antarctic continent can be expected to provide reference levels for all environmental and pollution studies due to its remoteness and restricted human activities. Aerosols are currently the largest source of uncertainty for characterizing the radiative forcing of climate (Schwartz and Andreae, 1996). To evaluate the influence of aerosols, we need to quantitatively attribute aerosol forcing to aerosol type (natural and anthropogenic) for highly diverse conditions. (Rathke 2002), the present experimental study, carried out at Maitri and Himadri, as part of the 24th Indian Antarctica Expedition and 14th Indian Arctic Expedition, reveled the following:

Antarctica mean AOD at 500 nm is found average value of 0.042 and in Arctic it average value is 0.11, the Angstrom exponent (Alpha) which is an indicator of aerosol size distribution, is found to vary an average value of 0.24 & 2.84 respectively. The aerosol size distribution shows mainly coarse-mode particles dominate tri-model distribution (each mode representing different sources and sizes of particles) with a primary mode at 0.1 - 0.6micrometer and secondary mode at 0.6 to 1.0 micrometer (accumulation-mode) and the third mode of particle size at 1.0 to 1.2 micrometer (coarse-mode) in Antarctica. As in the Arctic accumulation mode is dominating with the same size range of particle over the Antarctica. The TCO and PWC are found to vary from 240 DU to 320 DU with an average value of 289 DU, and from 0.14 to 0.36 cm with an average value of 0.26 cm over Antarctica. And in Arctic the ozone showed higher concentration varies from \sim 247 to 390.5 DU with an average value of 355 DU and water vapor concentration from 0.50 to 0.77 cm with an average value of 0.64 cm. which exhibited inverse relationship on certain days associated with low temperature, which reveals heterogeneous chemical processes leading to ozone destruction. In the present study on the Antarctica radiation flux where present which shows the fluxes to be dropped by about 20% as we approach to winter seasons. Where the estimated aerosol radiative forcing showed cooling at the surface, with a value varying between -1.44 W/m^2 and 0.44 W/m^2 with an average value of -0.47 W/m^2 during the period of study.

Surface snow and lake water samples collected during 24th Indian Antarctica Expedition (IAE) (December 2004-March 2005), 26th IAE (December 2006-March 2007) and in addition to snow and lake water sample, Anderson sampler were operated at Arctic Samples were analyzed for major chemical ions for both poles. In Artic expedition sample shows fine particles were dominant 58%, coarse particles 54% contributed respectively. In Antarctica samples shows snow and lake waters are slightly acidic in nature, with average pH values of snow 6.09 and pH value of lake water samples is 6.21 with low chemical contents and 6.45 with high chemical contents were found for arctic the pH value is found 5.85 *i.e.*, in alkaline nature. In all the samples, Na⁺ and Cl⁻ shows maximum concentrations based on mass and also among cations and anions and considerable amount of SO_4^{2-} are also found in the surface snow and lake water which is attributed to the oxidation of DMS produced by marine phytoplankton. It is found that in the snow sample NH_4^+

 Mg^{2+} is dominate for neutralization of acidic and component, which indicate the dominance of anthropogenic activity over the natural aerosols and in lake water the Mg^{2+} , Ca^{2+} and K^+ are nearly equally effective in neutralizing the acidic components. Cl^2 , SO_4^2 and Na⁺ respectively more contributed in the total ionic mass of the samples. The NH_4^+ and SO_4^{2-} occur over the Antarctica region mostly in the form of (NH₄)₂SO₄. In the Arctic region we found the clear dominance of marine influence with 77 % contribution from Na, Cl and Mg, which was followed by about 17 % contribution from Ca whereas; SO₄ and NO₃ contributed only about 3 and 0.1 % respectively indicating very weak influence of anthropogenic activity.

Loading of BC shows in Arctic region about 90 ng/m³ in the atmosphere during the afternoon hours from 12:00 to 16:00 hrs (local time) with a mean value of BC 23 ng/m³. The day time BC is 2.4 times more than that in the nighttime at Arctic region. The day-to-day mean variation in BC at Indian Arctic station showed concentration of 30.54 ± 11.03 ng/m³during June and 55.65 ± 62.60 ng/m³ during July 2010.

Acknowledgements

This work was supported jointly by the IITM, Pune and NCAOR, Goa. The authors are grateful to the Director of the Indian Institute of Tropical Meteorology, Pune for support and encouragement. Support received from the scientists of National Center for Antarctic and Ocean Research (NCAOR), Goa and from the Ministry of Earth Sciences (MoES), Government of India. Exceptional logistic expertise provided by the NPI, (Ny-Alesund) staff is appreciated and acknowledged. Thanks are also due to all Members of the "Aerosol and Cloud Physics Laboratory for Weather and Climate Studies" particularly to P. Ernest Raj, K. K. Dani and P. S. P. Rao.

References

- Devara, P. C. S., Maheskumar, R. S., Ernest Raj, P., Dani, K. K. and Sonbawne, S. M., 2001, "Some features of aerosol optical depth, ozone and precipitable water content observed over land during the INDOEX-IFP99", *Meteorologische Zeirschrift*, 10, 901-908.
- Devara, P. C. S. and Sonbawne, S. M., 2008, "Some features of aerosols and pre-cursor gases observed over Antarctic region during 24th IAE", NCAOR Report (In Press).
- Devara, P.C.S., Sonbawne, S. M., Dani, K. K., Saha, S. K. and Ernest Raj, P., 2011, "Characterization of aerosols and pre-cursor gases over Maitri during 24th Indian Antarctic Expedition", *International Journal of Remote Sensing*, 32,1-12.
- Ernest Raj, P., Sonbawne, S. M., Dani, K. K., Saha, S. K., Pandithurai, G. and Devara, P. C. S., 2009, "Changes observed in sun photometer derived total column ozone and possible implications on surface-reaching UV radiation over a tropical Indian station", *International Journal of Remote Sensing*, 30, 4153-4165.

- Gadhavi, H. and Jayaraman, A., 2003, "Aerosol characteristics and aerosol radiative forcing over Maitri-Antarctica", *Current Science*, 80, 296-304.
- Ichoku, C., Levy, L., Kaufman, Y. J., Remer, L. A., Rong-Rong, Li, Martins, V. J., Holben, B. N., Abuhassan, N., Slutsker, I., Eck, T. F. and Pietras, C., 2002, "Analysis of the performance characteristics of the five-channel Microtops-II sun-photometer for measuring aerosol optical thickness and precipitable water vapor", J. Geophys. Res., 107, D13, 10.1029/200IJD001302.
- IPCC, 1995, Climate Change 1995 : The Science of Climate Change, [Houghton, J. T. et al., [eds.)]. Cambridge Univ. Press, 572, 15.
- Jain, S. L., Arya, B. C., Ghude, S., Kumar, A. and Pandey, P. C., 2004, "Measurement of green-house gases and ozone at Maitri, Antarctica", Proc. XXVIII SCAR Open Science Conference, Brochure 2004.
- Maheskumar, R. S. and Devara, P. C. S., 2004, "Observational estimation of direct radiative forcing by atmospheric aerosols, Proc. International Conference on 'Aerosols, Clouds and Indian Monsoon", IIT, Kanpur, India, November 15-17, 2004, *IASTA Bulletin*, 16, 30-33.
- Panneerselvam, C., Jeeva, K., Nair, K. U., Gurubaran, S., Rajaram, R., Dhar, Ajay and Rajaram, G., 2004, "Observation of Atmospheric Maxwell Current at Indian Antarctica Station, Maitri", 19th Indian Antarctica Expedition, Scientific Report, 17, 99-105.
- Penner, J. E., Andreae, M. O., Annegarn, L., Barrie, J., Feichter, G., Hegg, D., Jayaraman, A., Leaitch, R., Murphy, D., Nagana, J. and Pitari, G., 2001, "Aerosols, their direct and indirect effects in climate change: IPCC Report", Cambridge University Press, 289-348.
- Rajaram, G. and Reddy, B. M., 1993, "Atmospheric Research from Antarctica – The Indian Contribution", DST/ CSIR Publ., p104.
- Ramanathan, V., Crutzen, P. J., Kiehl, J. T. and Rosenfeld, D., 2001, "Atmosphere aerosol Climate and the hydrological cycle", *Science*, 294, 2119-2124.
- Rathke, C., 2002, "Properties of Coastal Antarctica aerosols from combined FTIR Spectrometer and Sun-photometer measurements", *Geophys. Res. Lett.*, 29, 2123-2131.
- Russell, P. B., Livingston, J. M., Hignett, P., Kinney, S., Wong, J. and Hobbs, P. V., 1999, "Aerosol - induced radiative flux changes off the United State mid-Atlantic coast; comparison of values calculated from sun photometer and in situ data with those measured by airborne pyranometer", J. Geophys. Res., 104, 2289-2307.
- Schwartz, S. E. and Andreae, M.O., 1996, "Uncertainty in climate change caused by aerosols", *Science*, **272**, 1121-1122.
- Sonbawne, S. M., Raj, P. E., Devara, P. C. S. and Dani, K. K., 2009, "Variability in summertime total column ozone over the Indian station, Maitri in the Antarctic region", *International Journal of Remote Sensing*, **30**, 4331-4341.
- WMO (World Meteorological Organization), 2003, Scientific Assessment of Ozone Depletion : Global ozone research and monitoring project, 47, p498.