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Seasonal Variability of Atmospheric Aerosols in Karachi, Pakistan

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Abstract: A variety of *in situ* and satellite-derived data of aerosols like atmospheric black carbon concentrations were used to probe the seasonal differences of aerosol concentration in Karachi, Pakistan for one year. Daily [black carbon] varied from about 4000 to 50,000 ng/m^3 with the mean maximum of 14700 ng/m^3 in February, primarily during mornings and evenings. The [black carbon] concentrations were at a maximum during winter months of November to February i.e. around 12000 ng/m^3 and were at minimum value during summer from June to September (3000 ng/m^3). Short term and long-term variabilities were mostly affected by meteorological parameters. Apart from industrial and indiscriminate solid waste burning, most important source of BC emissions in Karachi was vehicular traffic, since over a million vehicles were registered in the city. Aerosol Optical Depth (AOD) from multi-band AOD, AERONET, and MODIS satellites showed a similar trend of its concentrations similar to BC. Aeronet 500 nm AODs were at a maximum for July (0.95 monsoons) and minimum (around 0.4) in November-February. Seasonal variation of AOD (Aeronet) was matching at other wavelengths, while the deviation in the spectral dependency of AOD was uncertain. It implied that a columnar spectral optical depth represented different aerosol type association having advection from various directions and sources. Relevant stakeholders should play their role to reduce BC emissions to mitigate ill health impacts in this metropolitan city.

Keywords: Black carbon aerosol, aethalometer, BC concentrations, urban area and aeronet, MODIS satellite data.

Introduction

The suspended particulate matter in the atmosphere has a number of health and climatic effects. Direct climatic effects are governed by their absorption mechanism and scattering the short and long wave radiations, either coming from the sun or reflected from earth surface. This, in turn, may lead to changed radiative equilibrium between earth and its atmosphere.

Microphysical and radiative properties of the atmosphere are modified by aerosols with varying sizes of combustion products of fossil fuel and biomass related black carbon and mineral dust that originate from human activities besides natural ones. As a result, they exert significant direct or indirect radiative forcing. In fact, energy balance is defined by the change in atmospheric composition.

The magnitude of these effects remains to be determined (IPCC, 2007). These effects originate from the solar radiation absorption and scattering by aerosol particles. Cloud size and density of droplets, alter the albedo, as well as the precipitation (Twomey 1977; Albrecht 1989; Rose field 2000). Radiation scattered by sulfate particles leads to cooling effect through a decrease in radiations reaching earth (Charlson et al., 1992). Contrarily black carbon (BC) absorbs radiations in visible and IR part of spectrum resulting in warming of atmosphere and cooling of surface. Volatile organic or inorganic components of BC are relatively non-absorbing. Much of the absorption of carbonaceous particles is due to the fraction of elemental or graphitic carbon (Rosen et al., 1978). Black carbon aerosol strongly absorbs solar radiation. According to IPCC (2007) reports, radiative forcing (RF) of black carbon aerosols range is 0.27 – 0.54 Watts per square meters and the range for the organic-carbon in negative values is 0.04 − 0.41 Watts per square meter. For dust aerosols, RF ranges from −2.0 to +0.5.

Ramanathan et al., (2001) reported aerosol emissions from South Asia. Black Carbon aerosol strongly absorbs solar radiation and it has been termed as a significant climate forcing agent. Global fossil fuel related BC emission are estimated to be ~ 6-8 Tg C yr^{-1} (Haywood and Boucher, 2000). Bond et al. (2004) estimate these emissions of [black carbon] to be 8 TgC yr^{-1}, with a component of 4.6 TgC yr^{-1} from fossil fuel and bio-fuel combustion and 3.3TgC yr^{-1} for uncovered garbage a flaming.

The relationship between particle mass and mortality/morbidity is well established but recent findings have also suggested linkages between carbonaceous particles and cardiovascular, respiratory, and neurological problems. Particle size is the prime parameter which assesses its impact of BC on human health and on the environment. Particulate matter (PM) of diameter of 2.5μm or less is being investigated to study links between the concentration of PM2.5 which can cause respiratory and cardiac disorders (Lopez-Reyes, Antonio et al., 2016). The IPCC (2007) reported RF of BC aerosols from garbage burning to be +0.27 to +0.54Wm^{-2}. Therefore, the quantitative assessment of AOD is important for climate change and other impact assessments.

Populations of mega cities are globally facing exposure to major pollutants including NOx, O3 and COx and the monitoring of these particles is not an easy task (Deary, et al., 2016).

The objectives of this study were to examine the temporal changes of black carbon aerosols and their relationship with AOD values as obtained from ground and satellite-based measurements in Karachi (24° 56' N and 67° E) which is the port and biggest city of Pakistan. A quantitative study of aerosol and spectral deviations of AOD is vital to understand impacts of aerosol in the local area and in neighboring countries. The city houses a large number of low-tech industries with uncontrolled combustion, causing emissions of pollutants into atmosphere (de Miranda et al., 2012). Round the clock measurements of atmospheric BC concentrations using an Aethalometer in Karachi, Pakistan were made from January to December 2007 08 to study trends of black carbon. This helped in assessing the importance of various emission sources and their contributions. In parallel, to correlate these BC values, aerosol profile from Aeronet Station at Karachi and AOD maps from Modis on Terra and Aqua satellites were obtained.

Study Area

This study has been conducted for the mega city of Karachi having a population of approximately 18 million. The urban area of city cover is 700 sq km. Large percentage of population uses combustion vehicles in city causing most of its air pollution. Overall climate is sub-tropical with more moisture in summer and low moisture in winter. Winds in Karachi mostly blow from south and south west except during winter months of November to February when it blows from north east. Daytime temperatures is nearly ~27˚C.

The sources of air pollution including particulate matter in Karachi are both natural and anthropogenic. The natural sources include dust entrainment, dust storm and sea salt sprays. The city has many types of manufacturing, textile, food and chemical industries, and there are power production plants on oil and gas combustion. The main population is spatially distributed along the sea side. The soil is mostly sandy and the areas around the city are arid, but there are some agricultural sites along the Malir river and some are irrigated by Hub river.

The PM 2.5 level is higher and it mostly exceeds safety levels by a factor of two. The maximum level recorded is 279 µg / m^3 from Aug 2008 to Aug. 2009.

Table 1: Meteorological data for Karachi 2007 -8

Year	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Annual
Monthly Amount of Precipitation (Mm)													
2007-8	4	7	17	0	0	55	47.5	143	Trace	0	0	19	294
Mean Monthly Maximum Temperature (°C)													
2007-8	26	**28**	33	35	35	**36**	34	32	35	36	33	27	32
Mean Monthly Minimum Temperature (°C)													
2007-8	**12**	14	20	24	**27**	29	28	27	27	23	18	14	22
Mean Monthly Relative Humidity %													
2007-8	30	37	40	48	62	63	65	70	59	42	36	39	49
Mean Monthly Wind Speed (M/Sec)													
2007-8	2	4	4	5	6	5	5	5	4	3	3	2	4

Aeronet

Sun photometer AERONET (AErosolR Obotic NET work) was used for the in-situ measurements of spectral AOD. The CIMEL radiometer is used to measure direct solar radiance at seven spectral channels viz 340,380,440,500,675,870 and 1020 nano meters (Holben et al., 1998). AOD measurement is taken into a column of atmosphere. The network of sun photometer spread all over the globe provides the observations in every 10 minutes interval for this purpose. A robotic system takes the readings by an auto sun tracking system. The radiometer retrieval accuracy is explained by Dubovik et al. (2000). Level 1.5 AOD data were obtained from GSFC AERONET. Daily averaged AOD at 500 nano meters direct solar data for year 2007 and 2008 have been used. The study also interpolates the daily average AOD at 500 nano meters from AERONET to 550 nm using appended relationship.

$$AOD_{550nm} = AOD_{500nm} (550/500)^{-\alpha}$$

Where α is the Angstrom exponent (Prasad et. al., 2004). Its value is 440 - 870 nm.

Satellite aerosol data from MODIS were used to analyze the variability of aerosols over Karachi (24.87 N, 67.03 E, and elevation 49 m). The MODIS sensors provide 36 spectral channels with an abundance of information on atmosphere, land and ocean. Aerosol retrieval is different over land areas (Kaufman et al., 2005). This study used MOD04_L2 product, which is a diurnal averaged Level 2 dataset.

BC Aerosols Measurements

A multisource data has been used for this research. Ground based measurements included BC concentrations using AE21 Aethalometer (Magee Scientific USA). Aerosol vertical column loading in term Aerosol Optical Thickness (AOD) were obtained from a CIMEL Sun Photometer (in operation at Karachi), as a part of the AERONET (Aerosol Robotic NETwork, Holben et al. 1998) and from MODIS sensors on Aqua and Terra Satellites. The AE21 quantify BC by gathering particulate matter on quartz filter and continuously observing transmission intensity. These beams pass through the loaded filter and are compared to reference beams that pass through an unloaded one. The AE21 was operated with a cyclone inlet (BGI corporation) equipped with an insect and rain guard at a flow rate of 4 LPM (liters per min) making the cut-point 3.2 µm.

NOAA's HYSPLIT model was used to determine back air trajectories, with meteorological variables.

Results and Discussion

Seasonal variability of Aerosols

The meteorological observations for Karachi as obtained from Pakistan Meteorological Department were reported in Table 1, while PM 2.5 data during the year October 2007 to July 2008 for the city appears in Table 2. Mean concentrations of the fine particulate (PM 2.5) as obtained using DustTrak™ 8520 Aerosol Monitors varied from 50 - 75 µg/m3 during fall and winter months of October - December 2007, January - February 2008. The same varied from 26 - 43 µg/m3 during spring/ summer months of February - July. The ground level concentrations of PM 2.5 were normally higher than the prescribed level of 35 µg/m3 of USEPA causing serious health effects. Two folds concentrations were observed during fall and winter which coincide with AOD Aeronet and AOD MODIS data. Same is the case for ground level BC measurements during 2007-2008 (Table 2).

Seasonal Variability of BC

Higher aerosol concentrations were reported over South Asia because of ever increasing population and industrialization (Dey et al., 2004). The same is true for the mega city of Karachi. Black Carbon concentrations varied from about 4000 - 50,000 ng/ m^3 with mean maximum of 14700 ng/ m^3 in February 2007 primarily during the morning and evenings (Fig. 1). Black Carbon values were high during winter months that is around 12000 ng/m^3, and these remain low during summer, i.e. 3000 ng/ m^3. Apart from industrial and indiscriminate solid waste burning, the most important source of BC emissions in Karachi was vehicular traffic, since over a million vehicles are registered. Majority of them are not properly maintained as they spew heavy amounts of black carbon into the atmosphere.

Monthly mean black carbon concentrations were determined and plotted in (Fig 2a, b). Winter months (November to January) show higher [black carbon] values compared to summer months of March to July. Seasonal [black carbon] values were mostly dependent on prevailing meteorological conditions. During summer months (March to September), temperatures vary from 27 to 36ºC and winds in Karachi blow from south west to north east that is from Arabian sea to inland, carrying mostly the black carbon emissions from coastal areas to the monitoring site. Furthermore, higher temperatures result in unstable atmosphere and enhanced vertical mixing leading to lower ground level black carbon concentrations. In winter months the meteorological conditions are just the opposite of those in summer, as winds having lower wind velocity blow from northeast to south west while temperatures vary from 12 to 28ºC (Table 1), leading to stable atmospheric conditions and accumulation of black carbon particulates and hence its higher black carbon concentrations.

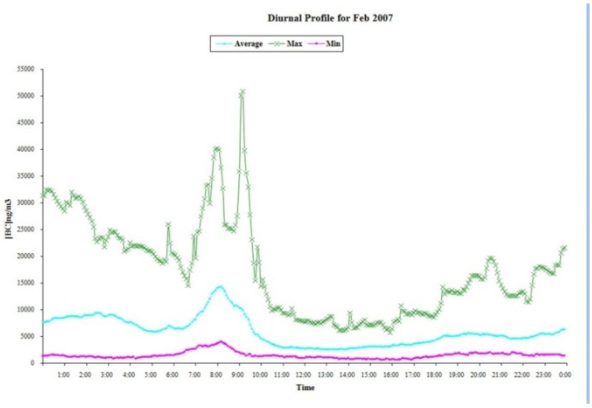

Fig. 1 Diurnal profile of black carbon concentrations Feb 2007

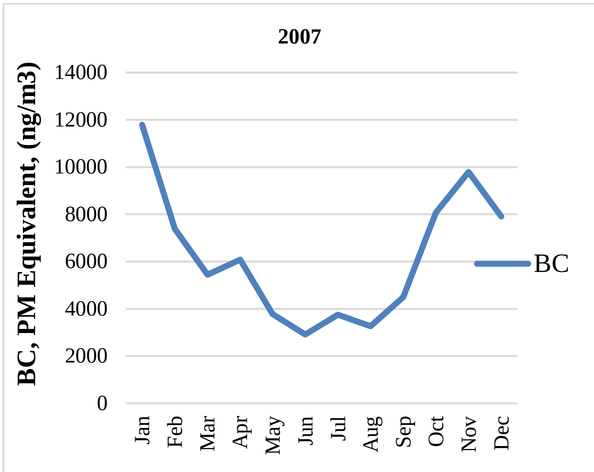

Fig. 2a: Seasonal variation of black carbon concentrations.

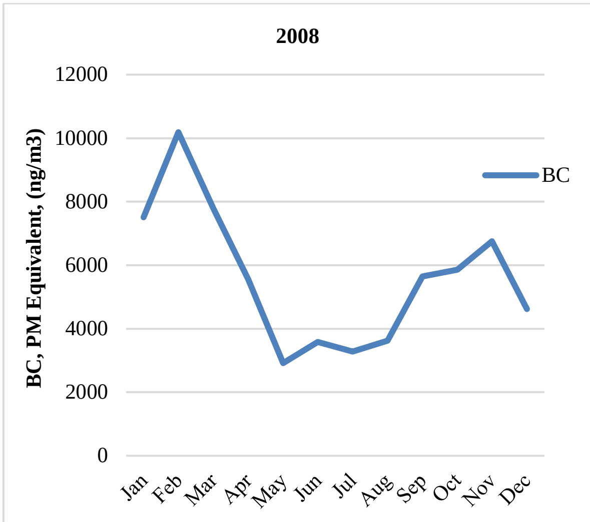

Fig. 2b: Seasonal variation of black carbon concentrations.

Seasonal Variability of AOD

Daily mean and monthly mean spectral AODs were estimated for Karachi during 2007 and 2008. Considering the local meteorology, AOD values extracted from Aeronet sensor were seen to be extremely high (around >1 at 500nm) due to the monsoon season from July to September (Fig. 3a, b). Apart from these months, AOD remained low during summer and high in winter. Aeronet AOD at all wavelengths also followed the same pattern as that of seasonal BC concentrations. Aeronet AODs on the whole were maximum for winter months with a decreasing trend for summer season. Figure 4a shows maximum AOD values for 4 Feb 2007 (0.55 at 500 nm around 0900 GMT) and maximum AOD values of 0.5 at 500 nm on 24 May 2007 at 0500 GMT (Fig. 4b). Figure 5 provides a better insight into the high level of uncertainties of AOD in its seasonal variation. This also implies that a columnar spectral optical depth depicts a different aerosol type associated with having advection from various directions and sources.

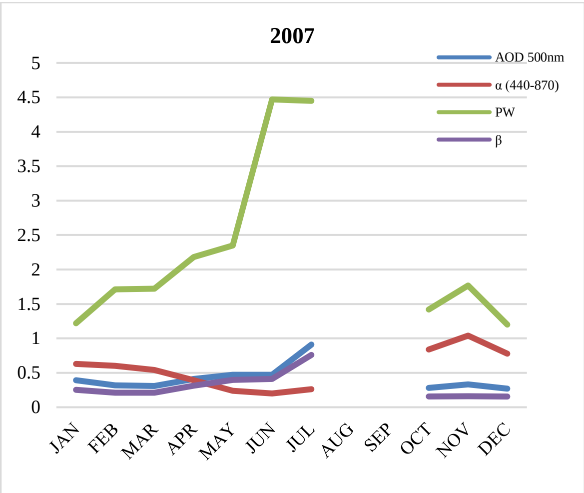

Fig. 3a Seasonal variation of AOD 500nm, α (440-870nm), PW & β.

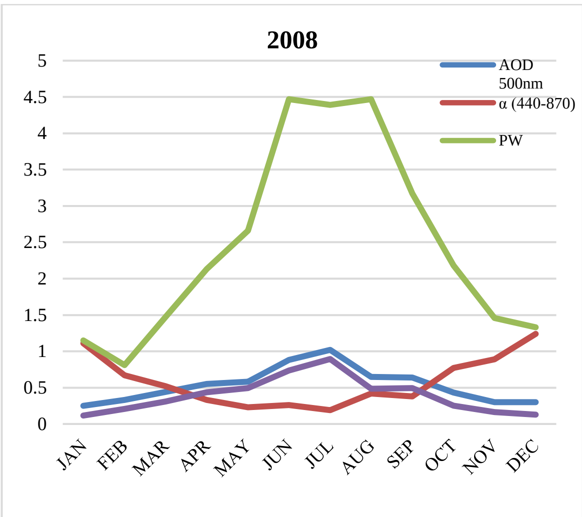

Fig. 3b Seasonal variation of AOD 500nm, α (440-870nm), PW & β.

AOD maps obtained from MODIS for Karachi on these dates (4 February 2007 and 24 May 2007) also confirm the AOD seasonal trend as presented above using Aeronet and in situ Aethalometer measurements. The AOD values from these maps are 0.54 and 0.39 for 04 Feb. and 24 May 2007 respectively.

Angstrom exponent α and angstrom turbidity coefficient β are known as Angstrom parameters. The angstrom turbidity coefficient (β) has been calculated by employing the following equation.

$\beta = \tau \lambda^{\alpha}$

τ = aerosol optical depth for Aeronet observations,

λ = wavelength in microns

α = corresponding angstrom exponent value

The time series of the β were plotted along AOD, Angstrom Exponent and PW taken from Aero-net (Fig. 4a, b).

Seasonal Variations

Alpha-Beta Relation

The Alpha-Beta Relationship has been shown in Figure 4a, b). A positive correlation between AOD (τ) and Turbidity Coefficient (B) has been seen with a very high correlation value around 0.98 irrespective of seasonal variations. This is perhaps due to the closeness of the data for the particular concentration in terms of AOD and atmospheric turbidity recorded during these two years. The positive correlation between two parameters harmonizes well with the field measurements data (Table 2). High PM levels imply high AOD and hence high angstrom turbidity. The value of β represents particles in vertical column, which is typically 0 - 0.5. The value of β which is close to 0.1 depicts a turbid atmosphere. In fact, β is the aerosol optical depth at λ= 1 µm. AOD and β vary together. Higher atmospheric particulate concentrations, which are a regional characteristic give rise to higher AOD values which in turn means higher fine particulate concentrations and strong correlation to high angstrom turbidity β. The value of α represents size distribution. Ranjan *et al.*, (2007) reported that positive values of Angstrom exponent are characteristics of fine-mode-dominated aerosols while near zero and negative values are characteristics of coarse mode (Eck *et al.*, 1999). In this case, greater angstrom exponents were determined during winter months and α<1 occurred during summer months indicating fine particle dominance during winter having regional sources such as agricultural waste burning, industrial and vehicular emissions. While, lower angstrom exponent (α) during summer indicates coarser particle entrained in the atmosphere under strong wind flows.

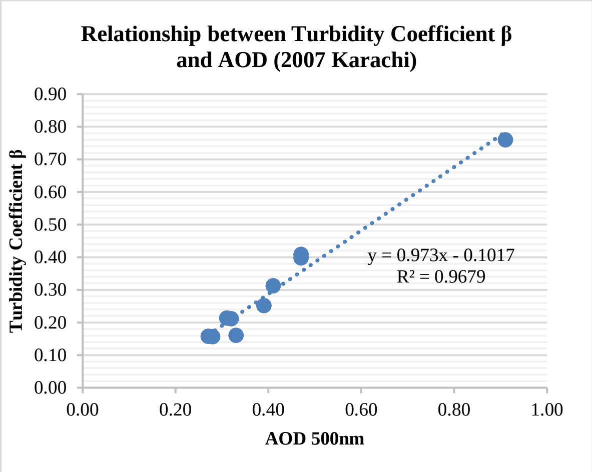

Fig. 4a Correlation of Aerosol parameters β and AOD at Karachi.

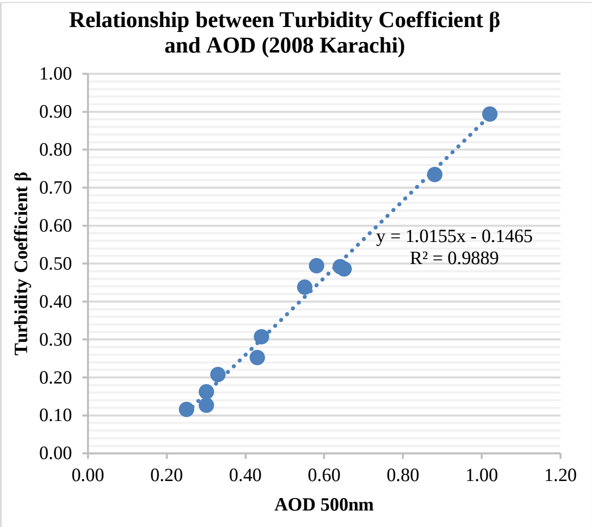

Fig. 4b: Correlation of Aerosol parameters β and AOD at Karachi

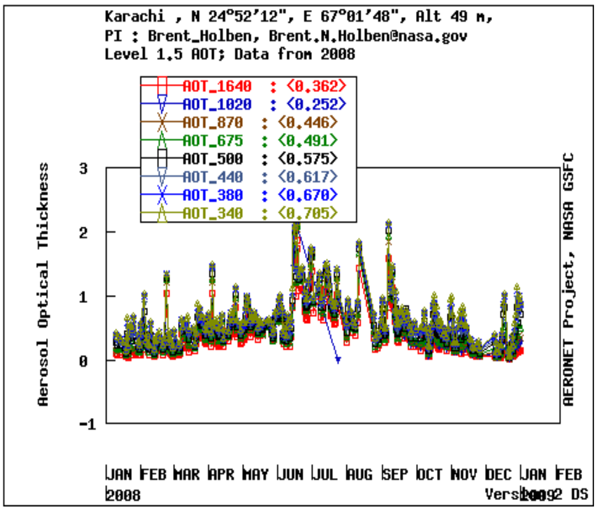

Fig. 5 Seasonal variation of AOD as obtained from Aeronet.

Seasonal variations of PM 2.5, BC (PM 2.5), Aeronet AOD and Angstrom Parameters (Alpha, Beta) indicate type of aerosol and their probable sources. As has been mentioned above, that higher Alpha values prevail during winter months which means finer particles were in abundance during these months. The prevalence of relatively hazy atmosphere is seen in MODIS-AOD maps (Fig 7a,b) during the winter period in 2007-2008 (Table 1) which is due to biomass and solid waste burning, industrial emissions, and natural dust.

Sources

In Karachi, both natural and anthropogenic sources of air pollution are active due to geographic location of the city. It is an industrial city having many types of manufacturing, textile, food and chemical industries, and there are power production plants on oil and gas combustion. There are over a million registered and unregistered vehicles and is one of the main causes of the air pollution. Karachi is a coastal city on the Arabian sea in Kirthar mountain range in the Northern and north western region of Karachi.

Apart from local sources, long-range transport of aerosols from neighboring areas could change the aerosol properties in a given region (e.g., Moorthy *et al* 2003, Moorthy *et al* 2005). In order to study such changes in AOD values at the monitoring site, we computed the weekly air mass back trajectories using HYSPLIT model for Karachi. A trajectory is the path of infinitesimally small particles of air as they move through time and space. Since adverted mass of aerosol particles in the entire atmospheric column are known to modify AOD values and 3 levels were chosen for drawing these backward trajectories using the criterion given by Moorthy et al 2005 i.e. one at 500m (within the Atmospheric Boundary Layer (ABL); 2000m (above the ABL), and at 3500m (friction free troposphere). The 7-day backward trajectories were considered, since fine aerosol normally can stay for a week or so in the atmosphere before settling through wet removal or absorbed by the earth surface (Jaenicke 1984; Babu and Moorthy, 2002). Due to low rainfall and sandy nature of the soil, it is the most important natural aerosol source of the city. Other natural source is the frequent dust storms blowing from west Asian and Gulf States during pre-monsoon period (April through June).

To further explain the contribution of advection to Karachi station in winter and summer, we plotted seasonal back trajectories for the years 2007 and 2008 i.e. January - Feb 2007 and May - June (Fig.6a and 6b). On the average, the long-distance contribution has been observed to be originated from west Asia, Middle East Iran, Afghanistan, and North Africa. The June 2007 and 2008 showed a contribution from Arabian at lower altitude at 500 AGL. During the summer months, as the contribution is seen from the sea in the form of large sized particles, there is a sudden decrease in AOD values compared to winter ones. This has also been explained earlier in the section on alpha-beta relationship.

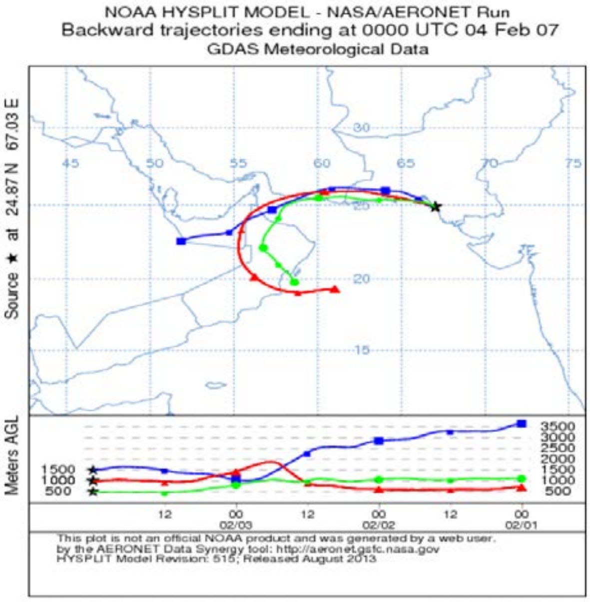

Fig. 6a HYSPLIT backward trajectory 04 February, 2007.

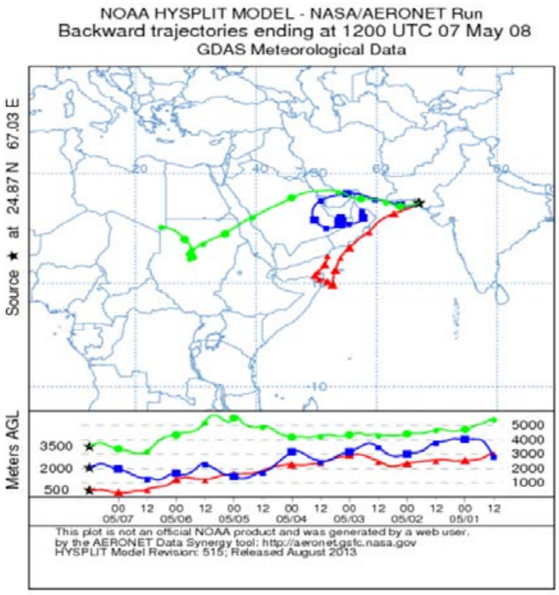

Fig. 6b HYSPLIT backward trajectory 7 May, 2008.

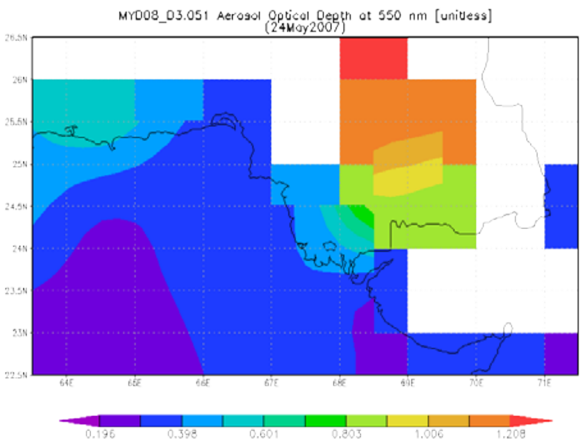

Fig. 7a AOD MODIS Map of May 24th 2007.

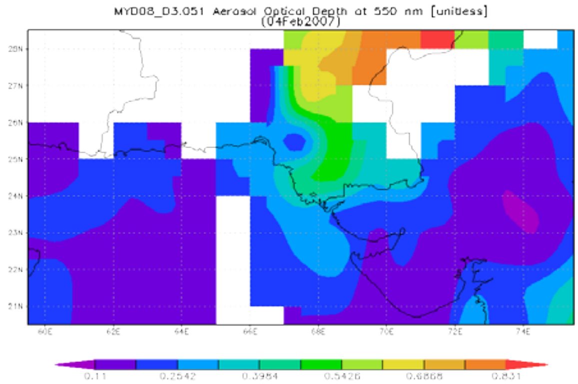

Fig. 7a: AOD MODIS map of May 24th 2007.

Conclusion

The BC was high in winter months that is around 12000 ng/m^3, but it remained low in summer, i.e. 3000 ng/ m^3. These trends were mostly affected by meteorological parameters and emissions from industrial and indiscriminate solid waste burning and the most important source of black carbon emissions in Karachi was vehicular traffic. Aerosols in terms of Aerosol Optical Depth (AOD) from AERONET and MODIS satellites showed similar trends as that of black carbon. Aeronet 500nm. AODs were maximum for July (0.95 monsoon) and minimum (around 0.4) in November-February. However, changes in spectral dependency were uncertain. This also implied that a columnar spectral optical depth represents a different aerosol type associated with having advection from various directions and sources. Seasonal AOD maps obtained from MODIS for Karachi also confirm the AOD seasonal trend as stated in case of Aeronet and in situ Aethalometer measurements.

Relationship between Aeronet AOD and aerosol parameters (α and β) were also determined. In our case, greater angstrom exponent α during winter months and lower $\alpha<1$ occurred during summer months indicating fine particle dominance during winter having regional sources such as, agricultural waste burning, whereas lower angstrom exponent (α) during summer indicates coarser particle entrained in the atmosphere under strong wind flows. The prevalence of relatively hazy atmosphere as seen in MODIS-AOD maps during the winter period in 2007-2008 were matched well with high PM at monitoring places. On the average, the long-distance contribution was observed to be originated from west Asia, Middle East, Iran, Afghanistan, and North Africa. Back trajectories (HYSPLIT) for February 2007 and May 2008 showed a contribution from Arabian sea at lower altitude at 500 AGL.

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