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# Monitoring spatio-temporal aerosol patterns over Pakistan based on MODIS, TOMS and MISR satellite data and a HYSPLIT model

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## ABSTRACT

Three different satellite-borne sensors, namely the Total Ozone Mapping Spectrometer (TOMS), the Moderate Resolution Imaging Spectroradiometer (MODIS), and the Multi-angle Imaging Spectroradiometer (MISR), were used to investigate the spatial and temporal variations of aerosols over several cities in Pakistan. A Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model was used for trajectory analysis in order to reconstruct the origins of air masses and understand the spatio-temporal variability of aerosol concentrations. Recent MODIS aerosol data (2002-2008) and earlier TOMS data (1979-2001) revealed increasing concentrations of aerosols over Pakistan and adjacent areas. Validation of MODIS and MISR derived aerosol optical depths (AODs) with Aerosol Robotic Network (AERONET) data for 2007 demonstrated that the MISR data was more accurate when close to the ocean, while the MODIS was more accurate over vegetated areas. The relationship between MODIS and MISR AOD data from 2002 to 2008 was analyzed, revealing a strong correlation between the two datasets. An assessment of seasonal variability in AOD for industrial, urban, semi-urban, rural, and semi-arid areas revealed maximum AOD values during the summer over all the areas investigated. Back trajectory analyses indicated that while winter air masses reaching Pakistan had travelled long distances, summer air masses had travelled only short distances. The higher aerosol concentrations during the summer are interpreted to be a result of the air masses spending more time over land during the summer than they do during the winter. While monsoonal rainfall tends to reduce aerosol concentrations by washing aerosols out of the atmosphere, this effect is mainly restricted to the eastern and south-eastern parts of Pakistan.

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# 1. Introduction

Atmospheric aerosols play an important part in the energy balance of the earth-atmosphere system. Changes in the atmospheric aerosol load, greenhouse gases, solar radiation, and land surface properties alter the energy balance of earth's atmosphere (Papadimas et al., 2008). Kosmopoulos et al. (2008) stated that "In the last two decades aerosols have been recognized as a major factor in determining the global climate change (IPCC, 2007), since they play a crucial role in the solar and thermal radiative transfer in the atmosphere". Aerosols have a major effect on the solar radiation budget, both at the surface of the earth and in the atmosphere, as well as on the hydrological cycle and precipitation rate (Charlson et al., 1992; Kosmopoulos et al., 2008; Lohmann and Feichter, 2005; Ramanathan et al., 2001).

Various aerosol types have distinct effects on the sign and magnitude of aerosol radiative forcing (Kaskaoustis et al., 2007). Black carbon aerosols have, for example, contributed considerably to global warming (Jacobson, 2001; Penner et al., 2003) and appear to be responsible for increased rainfall in the southern parts of China and, at the same time, for drought conditions in the northern parts (Menon et al., 2002; Sarkar et al., 2006). Dust particles modify the transmission of both short-wave and long-wave radiation through the atmosphere through atmospheric scattering and absorption processes (Otto et al., 2007) and consequently produce heating in the atmospheric column due to dust absorption (Haywood et al., 2001). Aerosols also have significant effects on non-climatic related processes e.g., human health and ecosystem services (Smart et al., 2011). Biomass burning and boreal forest fire aerosol events often result in huge smoke plumes that spread thousands of kilometres from their sources, causing serious problems with regard to air quality and public health (Torres et al., 2002).

Aerosols concentrations vary in both time and space, influencing the microphysics of clouds. There is a growing body of literature has reporting the use of satellite measurements to investigate the





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indirect effects of spatial and temporal patterns of aerosols, at both regional and global scales (Kaufman et al., 2005; Matheson et al., 2005; Nakajima et al., 2001). Satellite remote sensing is an essential tool for monitoring the global aerosol budget and the radiative effects that aerosols have on climate (Penner et al., 1992; Tripathi et al., 2005). In theory, satellite remote sensing allows to the spatial distribution and properties of aerosols to be assessed, and it also has the major benefit of allowing complete and synoptic mapping of a large area in a single snapshot (Kosmopoulos et al., 2008). Satellitebased remote sensing undoubtedly provides a unique opportunity to derive regional, global, and seasonal spatial patterns for aerosol loads and aerosol properties. The spatio-temporal processes involved can then be indirectly described and mapped through modelling techniques, e.g., by using the USA's National Oceanic and Atmospheric Administration (NOAA) HYSPLIT model (Draxler and Rolph, 2003). In order to understand the effects that aerosols have on the earth's climate system and human health they must be routinely monitored, both on a global scale and on regional or local scales, in particularly by analyzing their spatial and temporal patterns.

Nevertheless, ground-based measurements require expensive software or permanent automatic monitoring stations. In several urban areas of Europe, the United States, Australia, and some regions of Asia, the mass concentration of ambient aerosol particles is routinely measured by ground stations. However, individual ground-based observations represent point measurements and do not have coverage required to map regional or global distributions of aerosols. One major advancement in this respect has been the introduction of the AERONET Aerosol Robotic Network (Holben et al., 1998), which means that satellite remote sensing of aerosols no longer needs to be largely independent but can be tied in to this coordinated and harmonised ground data. Satellite remote sensing has made tremendous progress within the last 10-15 years: recent MODIS (Chu et al., 2003; Wang and Christopher, 2003) and MISR (Liu et al., 2007) satellite data has been shown to have tremendous potential for mapping the distribution and properties of aerosols, and for deriving indirect estimates of particulate matter. Since ground measurements are sparse in many regions of the world, satellite data is able to serve as a surrogate measure for the monitoring of particulate matter air quality (Gupta et al., 2008).

Aerosol concentrations, particularly in Asia, are continually increasing in virtually all urbanized and industrialized regions because of growing populations, rapid urbanization with consequent land use changes, increasing motorized traffic, and increasing industrialization within, and adjacent to, urban areas. There are, to date, relatively few studies on aerosols in Pakistan (e.g., Alam et al., 2010; Alam et al., 2011; Dutkiewicz et al., 2009; Ghauri et al., 2001). Ranjan et al. (2007) analyzed the seasonal AOD variability over Rajkot, in India, reporting lower values in winter and higher values in summer, the latter being due to high wind velocities producing larger quantities of wind-driven dust particles. Dey et al. (2005) investigated the aerosol concentrations over Kanpur, in India, and found that urban and industrial aerosols contributed more than 75% of the observed AOD during the post-monsoon and winter seasons, whereas natural aerosols contributed 60% of the total AOD during the pre-monsoon and monsoon seasons. Prasad et al. (2007) examined aerosol variations over the Indo-Gangetic plain and found that the AOD was higher in the summer season than in the winter season. Similarly, Alam et al. (2010) found higher AOD values in summer and lower values in winter for various cities in Pakistan. The correlation between MODIS and AERONET AOD over Kanpur during the 2004 post-monsoon and winter seasons was almost the same as during the pre-monsoon and monsoon seasons (Tripathi et al., 2005), but Prasad and Singh (2007) found a moderate correlation between MODIS and AERONET AOD over Kanpur during the broader period from 2001 to 2005.

This research has investigated MODIS and MISR data for Pakistan, compared their AOD retrievals, and validated them against groundbased AERONET AODs. The regional spatio-temporal aerosol patterns for various major cities in Pakistan have also been assessed using MODIS and TOMS datasets with a HYSPLIT model. In addition, the seasonal variability of aerosols has been investigated in combination with a back trajectory analysis in an attempt to understand the spatial and temporal variability in aerosol load for the selected cities of Pakistan.

# 2. Methodology

# 2.1. Study area

Pakistan has a wide variety of natural resources: high mountains in the north are home to extensive forests, the western hilly areas are rich in mineral resources, and the eastern region, which is considered the most favourable for monsoonal rainfall, and is used for agriculture. In the extreme south of the country, Karachi is a harbour city, and the business and industrial capital of Pakistan (Fig. 1). The topographic variation makes Pakistan geographically unique for any study of spatio-temporal patterns. Altitudes within Pakistan range from sea level (i.e., the Indian Ocean) to the highest peaks of the greater Himalayan region (e.g., Mt. Godwin Austin/K2, at 8611 m). Since the dawn of independence in 1947, the country's population has continued to grow, placing increasing pressure on its resources. Data for ten selected distinct cities in Pakistan (Fig. 1) have been analyzed and the spatio-temporal variations in aerosol concentrations investigated. These ten cities comprise major urban centres of Pakistan, with generally dense populations and varied distribution patterns for residential, commercial and industrial land use areas.

# 2.2. Datasets and analyses

A variety of datasets and their derived properties have been used for this research. Satellite aerosol properties from the TOMS, MISR, and MODIS sensors were used to understand and analyze the variability of aerosols over different regions of Pakistan. Groundbased AERONET AOD data was used to validate MISR and MODIS AOD products. This study has analyzed aerosol properties, including aerosol concentrations, in terms of the TOMS aerosol index (AI): the AI dataset is archived and available at http://toms. gsfc.nasa.gov. Table 1 provides detailed information on the data employed. Radiances are measured by the TOMS in broad wavelength bands (313, 318, 331, 340, 360 and 380 nm) (Herman et al., 1997). The positive TOMS AI values correspond to absorbing aerosols, while negative values correspond to non-absorbing aerosols (Torres et al., 2002). For the period from 1979 to 1992 we used daily TOMS Nimbus-7 data, and for the period from 1997 to 2001 we used TOMS Earth Probe data. Annual means were calculated for both datasets.

The MODIS sensors onboard NASA's Terra (launched December 1999) and Aqua (launched May 2002) satellites have 36 spectral channels which provide an abundance of information on atmospheric, terrestrial, and oceanic conditions. Aerosol retrieval is different over land (Kaufman et al., 1997) from over oceans (Tanré et al., 1997). Several aerosol parameters are retrieved at a 10 km spatial resolution from MODIS daytime data. For our investigations we have used a MODIS Terra Level-2 daily mean data product (MOD04\_L2. in HDF format) at a spatial resolution of 10 × 10 km. The estimated uncertainty of the MODIS AOD product was reported to be  $\pm 0.05 \pm 0.15$  (AOD) over land (Chu et al., 2003) and  $0.03 \pm 0.05$  (AOD) over the ocean (Remer et al., 2005). The NASA MODIS aerosol retrieval algorithm has recently been improved and the products denoted as C005 were implemented to correct



Fig. 1. Map of study area.

systematic biases in the MODIS algorithm used previously (Remer et al., 2005; Levy et al., 2007).

The MISR Terra Level-2 global data product is available on a daily basis with  $17.6 \times 17.6$  km resolution (http://www-misr.jpl.nasa.gov/). The MISR instrument continuously acquires daytime data over most parts of the world, but with a frequency that is dependent on latitude. Due to the overlap of the swathes (paths) near the poles and their broad separation at the equator, coverage intervals vary from 2 to 9 days, respectively. The MISR acquires systematic multi-angle imagery for global monitoring of top-of-atmosphere and surface albedos, and measures the short-wave radiative properties of aerosols, clouds, and surface scenes, which are used to characterize their impact on the Earth's climate.

The AERONET CIMEL sky ground-based data are available at three levels; Level 1.0 (unscreened), Level 1.5 (cloud screened) and Level 2.0 (quality assured) (Holben et al., 1998), which can be downloaded from the AERONET website (http://aeronet.gsfc.nasa.gov/). The CIMEL sun/sky radiometer takes measurements of direct sun and diffuses sky radiances within the 340–1020 nm and 440–1020 nm spectral ranges, respectively (Holben et al., 1998). The sun/sky radiometer retrieval accuracy is comprehensively explained and discussed by Dubovik et al. (2000). Version 2 cloud screened daily mean AOD (500 nm) direct sun data over Karachi and Lahore for the year 2007 have been used in this study. For validation, daily mean AODs

#### Table 1

Datasets used in the study.

Satellite sensor Data used Product Special resolution Spectral band MODIS Terra 01/01/2002-31/12/2008 Daily level-2  $10 \times 10 \text{ km}$ 550 nm 01/01/2002-31/12/2008 Daily level-3  $1 \times 1^{\circ}$ MISR Terra 01/01/2007-31/12/2007 Daily level-2  $17.6 \times 17.6$  km 558 nm 01/01/2002-31/12/2008 Daily level-3  $0.5\,\times\,0.5^\circ$ **TOMS Nimbus-7** 01/01/1979-31/12/1992 Daily level-3  $1 \times 1.25^{\circ}$ 340-380 nm Earth Probe 01/01/1997-31/12/2001 AERONET 01/01/2007-31/12/2007 Daily level 1.5 500 nm

(500 nm) from AERONET were interpolated to a common wavelength of 550 nm using the power law.

$$AOD_{550nm} = AOD_{500nm} \left(\frac{550}{500}\right)^{-\alpha}$$

where  $\alpha$  is the (440–870 nm) Angstrom exponent (Prasad et al., 2007).

A HYSPLIT model has also been used to compute simple air parcel trajectories, in particular and backward trajectories, with meteorological variables (ambient temperature, rainfall, relative humidity, and solar radiation flux). The model can be run interactively on the website (http://ready.arl.noaa.gov/HYSPLIT.php).

#### 3. Results and discussion

#### 3.1. Aerosol concentrations

It is generally accepted that increases in aerosol concentrations over Asia are a result of increasing populations, growing economies, urbanization, and industrialization, as has been shown to be the case in India (Dey et al., 2005; Sarkar et al., 2006). An increasing trend in aerosol concentrations can also be observed in Pakistan, following the same general trend. Between 1979 and 2001 there has been a substantial increase in the AI over all ten selected urban/ industrial locations (Fig. 2). This is particularly the case in the southern parts of Pakistan, largely due to the megacity of Karachi which, with its population of 18 million, challenges the environment as a large industrial cluster (Qureshi, 2010). These AI values over Karachi also reflect the occurrence of local dusts that originate within the Thar Desert, in addition to the major regional or subcontinental events originating from the Sahara Desert (EI-Askary et al., 2006). The annual mean AOD in Karachi exceeds 0.3 and the absorption optical depth makes up about 10 per cent of the total AOD, indicating the presence of strongly absorbing soot aerosols (Ramanathan et al., 2008). Habib et al. (2006) found increasing trends in anthropogenic emissions between 1981 and 2000, and concluded that this was contributing to increases in the aerosol load detected by the TOMS over India.

Our study confirms the conclusions from Alam et al. (2010) that the aerosol concentration is high as a result of both natural and anthropogenic influences. Similar increasing trends in aerosol concentrations over these Pakistan cities have been detected from more recent MODIS monthly data, for the seven-year period covering 2002–2008 (Fig. 3). It is evident from Fig. 3 that the AOD increases over all investigated Pakistani cities. For a subset of the cities studied



Fig. 2. Trends in the aerosol index over selected cities in Pakistan.



Fig. 3. Trends in AOD over selected cities in Pakistan.

herein, Alam et al. (2010) found that the AOD increased rapidly from the month of April, reaching its maximum in July. On the other hand AOD started to decrease rapidly in September and reached its minimum in December. The summer increase in AOD is believed to be due to three main factors: (1) mineral dust carried from the Thar Desert in India and the Dasht Desert in Iran (see Fig. 7), (2) sea salt particles caught up by the strong south-westerly winds, and (3) an increase in humidity within the region (Alam et al., 2010). Ranjan et al. (2007) concluded that water vapour (precipitate) and AOD are directly related to each other, and hence a higher concentration of water vapour in summer leads to a higher AOD. Conversely, dry weather and lower humidity in winter result in lower AOD values. Our research found that, as a result of anthropogenic activities (like urbanization, land use change etc.), AOD increased and higher AOD levels persisted longer over industrial and densely populated cities in Pakistan (Karachi, Rohri, Lahore, and Faisalabad). Backward trajectory analyses of both previous and current data (see Section 3.4) indicated the origins of air masses and the sources of aerosol loads arriving in Pakistan, revealing which particular aerosol loads originated from adjacent regions rather than from the investigated cities themselves.

#### 3.2. Intercomparison of MODIS, MISR and AERONET AOD data

An intercomparison of AOD values from different satellite sensors is necessary if a long-term database for climatological studies is to be established, and also to improve the accuracy and the coverage achievable with a single sensor (Prasad and Singh, 2007). Xiao et al. (2009) recently examined MISR and MODIS AOD retrievals for South-East Asia from a spatial perspective and found good correlation between the retrievals from each of these sensors. Prasad and Singh (2007) carried out a validation of monthly average Level-3 MODIS and MISR AODs over Khanpur for summer and winter seasons between 2001 and 2004, and found that MODIS and MISR AODs were correlated with  $R^2 > 0.7$  in 20.5% of all pixels in the Indo-Gangetic region, but in only 13.9% of the pixels at worldwide average. The





spatial correlation for MODIS–MISR AODs has been analyzed using daily average Level-3 MODIS (MOD08\_D3.005) and MISR (MIL3DAE.004) data obtained from the NASA-operated GIOVANNI portal (http://disc.sci.gsfc.nasa.gov/giovanni). MISR products are available with a spatial resolution of 0.5° by 0.5°, whereas MODIS products are only available at a spatial resolution of 1° by 1°. MISR data were converted to the same spatial resolution as MODIS data on the basis of the Giovanni re-girding algorithm, using the box averaging method.

The spatial correlation was analyzed for the period covering 2002–2008 (Fig. 4c). The correlation coefficient between MODIS and MISR AODs was found to be high (>0.96) for most of the regions under investigation. In addition, the spatial correlation between MODIS and MISR AODs (Fig. 4, a and b) was analyzed separately for winter (December 2006–February 2007) and summer (June–August 2007), and the correlation coefficient found to be higher in winter than in summer. The number of data points in the winter period was considered by Prasad and Singh (2007) to be sufficient to produce



Fig. 4. Spatial correlations between MODIS-MISR AOD for (a) winter (b) summer (c) 1st January 2002-31st December 2008 over regions of Pakistan.

1.0

0.6

0.2

-0.2

0.6

1.0

reliable results. The correlation was high (0.9–1.0) for the winter data from seven of the cities (Peshawar, Rawalpindi, Zhob, Faisalabad, Lahore, Multan and Rohri), while for the three other cities or regions (Karachi, in particular, being more of a geographical region than a city - see Qureshi, 2010), i.e., Swabi, D.G. Khan and Karachi, the correlation was only moderate (between 0.6 and 0.8). The correlation in summer was found to be high (0.9-1.0) for five of the regions (Peshawar, Rawalpindi, Faisalabad, Rohri and Karachi), moderate (0.6-0.8) for four regions (Swabi, Zhob, Lahore and Multan), and considerably lower (0.2–0.4) for one of the regions (D.G. Khan). The negative correlation between MODIS and MISR (Fig. 4b) could be seen in the extreme North-East of Pakistan. Snow cover in the region leads to insufficient MODIS and MISR data points in the area, consequently weaker data analyses. Similarly the negative correlation for MODIS-MISR could also be observed over the Kharan desert in the North-Western Balochistan. The MISR AODs were not used for the trend analysis due to insufficient data points.

The correlation between AERONET and MODIS/MISR AODs during 2007 was also analyzed for Karachi and Lahore (Fig. 5). The correlation coefficient for Karachi was found to be relatively high between AERONET and MISR (0.67) and lower between AERONET and MODIS (0.59). In contrast, the correlation coefficient for Lahore was higher between AERONET and MODIS (0.72) than between AERONET and MISR (0.11). The results suggest that the MISR sensor provides better AOD estimates close to the ocean while AOD estimates from the MODIS sensor are better over terrestrial regions (especially over vegetated surfaces). The low correlation between AERONET and MISR AODs over Lahore is again due to the lack of sufficient MISR data points. In general, MODIS AOD values over Karachi are higher than those from AERONET, while for Lahore the AERONET and MODIS values are similar. In summary, good agreement was observed between AERONET and MODIS AODs over Lahore and between AERONET and MISR AODs over Karachi.

Many additional issues arise when using multiple sensors and sources (Xiao et al., 2009), including consistency, revisit times, instrument calibration, sampling differences, and data availability (Liu et al., 2007; Kahn et al., 2007). MODIS data are used to derive AODs using the extended dark target approach (Remer et al., 2005), whereas MISR data are used in clear sky to characterize surface properties in the red and near-infrared bands (Vermote et al., 2007). Some recent studies have suggested useful ideas for improving analyses including (a) dealing with algorithm consistency issues, and their potential upgrades (Kahn et al., 2007), (b) changing the MISR model to provide a better fit for high pollution areas (Jiang et al., 2007), and (c) obtaining data with increased accuracy through satellite retrievals at high spatial resolutions (Liu et al., 2007).

#### 3.3. Annual means and seasonal variations in AOD

Seasonal variations in AOD were analyzed using MODIS data over a period of seven years (see Table 2). Annual means, seasonal means, and standard deviations of AOD at 550 nm were calculated for the years 2002–2008. Our analysis showed that the highest AOD values were observed in the southern parts of Pakistan, particularly over Karachi and Rohri. The annual mean AOD for Karachi was 0.52 (standard deviation: 0.37) over this period, ranging from 0.51 in 2002 to 0.69 in 2008 (not shown in Table 2). The results indicate that aerosol concentrations increased with increased industrial activities. Similar increases in aerosol concentrations were found for other Pakistani cities. However, Rohri, as an arid city adjacent to the Thar Desert, had an annual mean AOD of 0.81 (standard deviation: 0.38). Natural dust aerosols are therefore more dominant at Rohri than those from human sources. Faisalabad, which is the second largest city of the country in terms of industrialization and the third largest city in terms of population, has an annual mean AOD of 0.70 (standard deviation: 0.31). The two urban areas investigated in the central part of Pakistan, Multan and D.G. Khan, comprise a mixture of urban and agricultural sites and are adjacent to the Nara and Cholistan deserts. These two cities have annual mean AOD values of 0.79 and 0.72, respectively. As both Multan and D.G. Khan are close to desert regions, dust aerosols were more persistent, along with local vehicular emissions. It is important to emphasize that increases in the total AOD values for Pakistan cities are due to a complex mixture of causes, and that it is, in particular, the amalgamation of material from different sources that produces the largest increases. The main influences identified were a) increasing populations and associated changes in land use, b) increased industrial development c) increased vehicular emissions, and d) locally derived dust. In northern Pakistan lower AOD values were observed over most of the cities: Swabi and



Fig. 5. Validations of MODIS and MISR AOD with AERONET AOD at 550 nm, over Karachi and Lahore, for the year 2007.

Table	2
Iupic	_

MODIC	Appual and coaconal	moone and standar	d domintions of AOD	at EEO pm in differen	nt citize of Dakistan for th	a partiad 2002 2008
wods -	AIIIIUdi dilu sedsolidi	I IIIEdilis dilu stalludi	u uevialions of AOD	<i>i</i> at 550 mm m umerer	III CILIES OF PARISLAIT IOF LE	e period 2002-2008.

City name	Population	Annual and seasonal means and standard deviations of AOD at 550 nm						
		Annual mean	Winter	Spring	Summer	Autumn		
Karachi	10,272,500	$0.7250 \pm 0.3365$	$0.4465 \pm 0.0671$	$0.6656 \pm 0.1532$	$1.0036 \pm 0.3138$	$0.4543 \pm 0.1699$		
Rohri	44,143	$0.8370 \pm 0.3866$	$0.5361 \pm 0.0756$	$0.7865 \pm 0.2538$	$1.3389 \pm 0.3332$	$0.6067 \pm 0.1525$		
Multan	1,310,400	$0.7930 \pm 0.3250$	$0.4267 \pm 0.1009$	$0.6759 \pm 0.2303$	$1.2328 \pm 0.2422$	$0.7396 \pm 0.1523$		
D.G. Khan	188,149	$0.7210 \pm 0.3276$	$0.4362 \pm 0.0768$	$0.6157 \pm 0.2025$	$1.1764 \pm 0.2503$	$0.6555 \pm 0.1306$		
Lahore	5,611,500	$0.6712\pm0.2771$	$0.5236 \pm 0.0857$	$0.5655 \pm 0.1872$	$1.0237 \pm 0.2380$	$0.6691 \pm 0.1106$		
Faisalabad	2,191,200	$0.7056 \pm 0.3131$	$0.4253 \pm 0.0861$	$0.5749 \pm 0,1958$	$1.1198 \pm 0.2577$	$0.7026 \pm 0.1200$		
Rawalpindi	1,558,400	$0.4179 \pm 0.1349$	$0.2247 \pm 0.0332$	$0.3467 \pm 0.0800$	$0.5267 \pm 0.1064$	$0.3337 \pm 0.0865$		
Zhob	44,248	$0.3311 \pm 0.1406$	$0.1682 \pm 0.0437$	$0.3772 \pm 0.0627$	$0.5075 \pm 0.0480$	$0.2718 \pm 0.0860$		
Peshawar	1,094,900	$0.4771\pm0.1959$	$0.2553 \pm 0.0332$	$0.4476 \pm 0.0851$	$0.7344 \pm 0.1359$	$0.4712\pm0.1018$		
Swabi	78,960	$0.3250 \pm 0.1063$	$0.2373 \pm 0.0397$	$0.2774 \pm 0.0509$	$0.4561\pm0.0906$	$0.2812\pm0.0669$		

Population source: GOP, 2000.

Zhob, for example, which are remote and agricultural sites, had annual mean AODs of 0.32 and 0.33, respectively.

These results are in general agreement with published results for India and the surrounding region, but highlighting some of the details will serve to emphasize their importance. For all of the cities investigated in Pakistan the highest mean AODs were recorded during the summer and the lowest AODs in the winter (Table 2). It is also evident from Fig. 6 that summer AODs were higher than those for all other seasons in the year, followed in order by spring, autumn and winter. Sarkar et al. (2006) found that for mainland India the AOD started to increase from the month of March and reached a maximum in June, but that high AODs persisted over neighbouring Pakistan and the Arabian Sea until August. Ranjan et al. (2007) analyzed the spectral variations in AOD and also found the highest values in summer and low values in winter. They found that the lowest mean AOD values occurred from October to December. This decrease just before the winter season could be due to cloud scavenging and rain washout processes (Ranjan et al., 2007), while the summer increase could be due to high wind velocities producing larger quantities of wind-driven dust particles. Alam et al. (2010)



Fig. 6. Seasonal variations in AOD for (a) winter (b) spring (c) summer (d) autumn, for the period covering 2002–2008, over various cities in Pakistan.





NOAA HYSPLIT MODEL Backward trajectories ending at 1200 UTC 17 Jul 08 GDAS Meteorological Data

NOAA HYSPLIT MODEL Backward trajectories ending at 1200 UTC 28 Jan 08 GDAS Meteorological Data



Fig. 7. Seven-day back trajectories for various cities in Pakistan, for 28th January and 17th July, 2008.

found that the highest mean AODs occurred during the summer season, particularly in the southern part of Pakistan where mean AODs were greater than 0.9. They also found that, although AOD values varied between low values in winter and high values in summer, the pattern of values in spring and autumn were quite similar to each other.

During summer (pre-monsoon) high temperature plays a vital role in heating and lifting loose material from soil due to higher wind speeds consequently higher AOD values observed. The lower AOD value in winter (winter monsoon season) is due to rain washout processes. During pre-monsoon and monsoon season water soluble aerosols grow hygroscopically in the presence of water vapour and contribute to higher AOD values. Water vapour and AOD are directly related to each other, and hence a higher concentration of water vapour in summer leads to a higher AOD (Alam et al., 2010; Ranjan et al., 2007).

#### 3.4. Influence of air masses on AOD, using trajectory analysis

In order to understand the origins of the air masses arriving in the studied region we performed seven-day back trajectory analyses based on the NOAA HYSPLIT model (Draxler and Rolph, 2003). The meteorological input for the trajectory model was the GDAS (Global Data Assimilation) dataset (reprocessed from National Centres for Environmental Prediction (NCEP) by Air Resources Laboratory). These trajectories were computed at several altitudes (2500 m, 1500 m, and 500 m) for January 2008 and July 2008. Fig. 7 reveals that air masses reached Pakistan from the Dasht (Iran), Thar (India), Cholistan (Pakistan), and Sahara deserts, and also from the Arabian Sea, and subsequently passed over the various cities in Pakistan. The trajectories for 28th January and 27th July 2008 are shown in Fig. 7. We also found similar trajectories for other time periods, but only these have been included as they are considered to be representative of the entire time period analyzed.

Local pollution due to locally derived dust, industries, biomass burning, and vehicular emissions (Alam et al., 2011; Lodhi et al., 2009; Dutkiewicz et al., 2009) can be seen to have contributed to an increase in the aerosols concentrations over Karachi and Lahore. Kuniyal et al. (2009) found that, for Mohal-Kullu in northern India, most of the back trajectories identified air mass flows originating either from the Sahara, from sub-Saharan regions, or from the Great Thar Desert of Rajasthan, and that consequently, a significant increase in AOD values could be observed over this area. In summer, the highest AOD values in India occurred over the Indian Ocean due partly to south-westerly winds bringing dust from the Horn of Africa via the Arabian Sea, and also to the mid tropospheric transport of dust from the Arabian Peninsula (Li and Ramanathan, 2002).

Air mass back trajectories shown in Fig. 7 indicate different sources in summer and winter. For example, winter air masses reaching Pakistan and surrounding areas have travelled long distances from the south-west, while the summer air masses have travelled shorter distances. It therefore seems likely that the air masses spent more time over land during the summer than during the winter, which would explain the higher levels of AOD observed during the summer months. This is also due to the uneven distribution of the solar flux reaching the Earth's surface: air at lower latitudes is warmer than at higher latitudes and tropical air therefore rises vertically from equatorial regions and moves towards the poles at high altitudes while the cooler polar air moves towards the equator at lower levels. The air pressure consequently increases at equatorial latitudes due to the outflow of air mass and moist equatorial air moves towards the poles under this pressure gradient. Since the air moves away from the equator in northerly direction at high altitudes over the Pakistan region and the Coriolis force has a significant effect, south and south-westerly winds dominate. It is clear however from Fig. 7 that the air masses have been influenced by the combined effects of land, industries and the Indian Ocean, and that this has resulted in higher AOD values in the southern parts of Pakistan.

### 4. Conclusion

TOMS, MODIS, and MISR aerosol measurements have been used to assess the variability of aerosol concentrations over various cities in Pakistan. Both previous (TOMS) data and current (MODIS) data showed an increase in aerosol concentrations with time. MODIS and MISR AOD retrievals have been compared in order to examine the accuracy of these datasets. An excellent agreement between MODIS and MISR AOD retrievals was noted over most of the regions. Validation of MODIS or MISR using AERONET has confirmed that MISR performs better than MODIS for areas close to the ocean, while MODIS performs better over vegetated regions.

It is also clear that aerosols vary regionally with different geographical circumstances, and also seasonally. Our annual and seasonal analyses showed that the highest annual and seasonal AOD means were found over the southern parts of Pakistan. This is interpreted as being a result of the larger population, rapid urbanization, increased industrialization, and the proximity of desert land masses in the south. Aerosol concentrations were lower in the north because most of the northern cities are semi-urban and agricultural land use dominates in these regions. Examination of these variations in AOD using back trajectory analysis revealed that air masses from Iranian deserts (mainly from the Dasht Desert) and Saharan regions, along with local pollutants (locally derived dust, vehicular emissions, and emissions from biomass burning), arrive via the Arabian Sea (see Fig. 7). These air masses carry dust and sea salt particles, resulting in higher AOD values and influencing the ten selected Pakistani cities in different ways. In order to obtain more comprehensive satellite AOD retrievals utilization of long time period AERONET is planned for future research projects aimed at achieving a better understanding of spatial and temporal variations in aerosols over this region.

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#### References

- Alam, K., Iqbal, M.J., Blaschke, T., Qureshi, S., Khan, G., 2010. Monitoring the spatiotemporal variations in aerosols and aerosol-cloud interactions over Pakistan using MODIS data. Advances in Space Research 46, 1162–1176.
- Alam, K., Trautmann, T., Blaschke, T., 2011. Aerosol Optical Properties and radiative forcing over mega city Karachi. Journal of Atmospheric Research, doi:10.1016/ j.atmosres.2011.05.007.
- Charlson, R.J., Schwartz, S.E., Hales, J.M., Cess, R.D., Coakley, J.A., Hansen, J., Hofmann, D.J., 1992. Climate forcing by anthropogenic aerosols. Science 255, 423–430.
- Chu, D.A., Kaufman, Y.J., Zibordi, G., Chern, J.D., Mao, J., Li, C., Holben, B.N., 2003. Global monitoring air pollution over land from the earth observing systemterra moderate resolution imaging spectroradiometer (MODIS). Journal of Geophysical Research 108, D21. doi:10.1029/2002JD003179.

- Dey, S., Tripathi, S.N., Singh, R.P., Holben, B.N., 2005. Seasonal variability of aerosol parameters over Kanpur, an urban site in Indo-Gangetic basin. Advances in Space Research 36, 778–782.
- Draxler, R.R., Rolph, G.D., 2003. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model Access via the NOAA ARL READY Website. NOAA Air Resour. Lab., Silver Spring, Md. Available at: http://www.arl.noaa.gov/ready/hysplit4.html.
- Dubovik, O., Smirnov, A., Holben, B.N., King, M.D., Kaufman, Y.J., Eck, T.F., Slutsker, I., 2000. Accuracy assessments of aerosol optical properties retrieved from aerosol robotic network (AERONET) sun and sky radiance measurements. Journal of Geophysical Research 105, 9791–9806.
- Dutkiewicz, V.A., Alvi, S., Ghauri, B., Choudhary, M.I., Husain, L., 2009. Black carbon aerosols in urban air in South Asia. Atmospheric Environment 43, 1737–1744.
- El-Askary, H., Gautam, R., Singh, R.P., Kafatos, M., 2006. Dust storms detection over the Indo-Gangetic basin using multi sensor data. Advances in Space Research 37, 728–733.
- Ghauri, B.M., Mirza, M.I., Richter, R., Dutkiewicz, V.A., Rusheed, A., Khan, A.R., Husain, L., 2001. Composition of aerosols and cloud water at a remote mountain site (2.8 kms) in Pakistan. Chemosphere: Global Science Change 3, 51–63.
- Government of Pakistan (GOP), 2000. Provincial Census Reports, Pakistan: Demographic Survey 1998. Federal Bureau of Statistics, Statistics Division, Government of Pakistan, Islamabad.
- Gupta, P., Christopher, P.A., Wang, J., Gehring, R., Lee, Y., Kumar, N., 2008. Satellite remote sensing of particulate matter and air quality assessment over global cities. Atmospheric Environment 40, 5880–5892.
- Habib, G., Venkataraman, C., Chiapello, I., Ramachandran, S., Boucher, O., Reddy, M.S., 2006. Seasonal and interannual variability in absorbing aerosols over India derived from TOMS: relationship to regional meteorology and emissions. Atmospheric Environment 40, 1909–1921.
- Haywood, J.M., Francis, P.N., Glew, M.D., Taylor, J.P., 2001. Optical properties and direct radiative effect of Saharan dust: a case study of two Saharan dust outbreaks using aircraft data. Journal of Geophysical Research 106, 18417–18430.
- Herman, J.R., Bhartia, P.B., Torres, O., Hsu, C., Seftor, C.J., Celarier, E., 1997. Global distribution of UV-absorbing aerosols from Nimbus 7/TOMS data. Journal of Geophysical Research 102, 16911–16921.
- Holben, B.N., Eck, T.F., Slutsker, I., Tanré, D., Buis, J.P., Setzer, A., Vermote, E., Reagan, J.A., Kaufman, Y.J., Nakajima, T., Lavenu, F., Jankowiak, I., Smirnov, A., 1998. AERONET-A federated instrument network and data archive for aerosol characterization. Remote Sensing of Environment 66, 1–16.
- Intergovernmental Panel on Climate Change (IPCC), 2007. Climate Change: The Physical Science Basis. Cambridge University Press, New York, pp. 131–216.
- Jacobson, M.Z., 2001. Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols. Nature 409, 695–698.
- Jiang, X., Liu, Y., Yu, B., Jiang, M., 2007. Comparison of MISR aerosol optical thickness with AERONET measurements in Beijing metropolitan area. Remote Sensing of Environment 107, 45–53.
- Kahn, R.A., Garay, M.J., Nelson, D.L., Yau, K.K., Bull, M.A., Gaitley, B.J., Martonchik, J.V., Levy, R.C., 2007. Satellite-derived aerosol optical depth over dark water from MISR and MODIS: comparisons with AERONET and implications for climatological studies. Journal of Geophysical Research 112. doi:10.1029/2006JD008175.
- Kaskaoustis, D.G., Kambezidis, H.D., Hatzianastassiou, N., Kosmopoulos, P.G., Badarinath, V.S., 2007. Aerosol climatology: on the discrimination of aerosol types over four AERONET sites. Atmospheric Chemistry and Physics 7, 6357–6411.
- Kaufman, Y.J., Tanré, D., Remer, LA., Vermote, E.F., Chu, A., Holben, B.N., 1997. Operational remote sensing of tropospheric aerosol over land from EOS Moderate-resolution Imaging Spectroradiometer. Journal of Geophysical Research 102, 17051–17067.
- Kaufman, Y.M., Koren, I., Remer, L.A., Rosenfield, D., Rudich, Y., 2005. The effect of smoke, dust, and pollution aerosol on shallow cloud development over the Atlantic Ocean. Proceedings of the National Academy of Sciences 102, 11207–11212.
- Kosmopoulos, P.G., Kaskaoutis, D.G., Nastos, P.T., Kambezidis, H.D., 2008. Seasonal variation of columnar aerosol optical properties over Athens, Greece, based on MODIS data. Remote Sensing of Environment 112, 2354–2366.
- Kuniyal, J.C., Thakur, A., Thakur, H.K., Sharma, S., Pant, P., Rawat, P.S., Moorthy, K.K., 2009. Aerosol optical depths at Mohal-Kullu in the northwestern Indian Himalayan high altitude station during ICARB. Journal of Earth System Science 118, 41–48.
- Levy, R.C., Remer, L.A., Mattoo, S., Vermote, E.F., Kaufman, Y.J., 2007. Second-generation operational algorithm: retrieval of aerosol properties over land from inversion of moderate resolution imaging spectroradiometer spectral reflectance. Journal of Geophysical Research 112, D13211. doi:10.1029/2006JD007811.
- Li, F., Ramanathan, V., 2002. Winter to summer monsoon variation of aerosol optical depth over the tropical Indian Ocean. Journal of Geophysical Research 107 (D16) 1029/2001 JD000949.

- Liu, Y., Franklin, M., Kahn, R., Koutrakis, P., 2007. Using aerosol optical thickness to predict ground-level PM concentrations in the St. Louis area: a comparison between MISR and MODIS. Remote Sensing of Environment 107, 33–44.
- Lodhi, A., Ghauri, B., Khan, M.R., Rehman, S., Shafique, S., 2009. Particulate matter (PM<sub>2.5</sub>) concentration and source apportionment in Lahore. Journal of the Brazilian Chemical Society 20, 1811–1820.
- Lohmann, U., Feichter, J., 2005. Global indirect aerosol effects: a review. Atmospheric Chemistry and Physics 5, 715–737.
- Matheson, M.A., Coakley, J.A., Tahnk, W.R., 2005. Aerosol and cloud property relationships for summertime stratiform clouds in the northern Atlantic from advanced very high resolution radiometer observations. Journal of Geophysical Research 110, D24204. doi:10.1029/2005JD006165.
- Menon, S., Hansen, J., Nazarenko, L., Luo, Y., 2002. Climate effects of black carbon aerosols in China and India. Science 297, 2250–2253.
- Nakajima, T., Higurashi, A., Kawamoto, K., Penner, J.E., 2001. A possible correlation between satellite-derived cloud and aerosol microphysical parameters. Journal of Geophysical Research Letters 28, 1171–1174.
  Otto, S., de Reus, M., Trautmann, T., Thomas, A., Wendisch, M., Borrmann, S., 2007.
- Otto, S., de Reus, M., Trautmann, T., Thomas, A., Wendisch, M., Borrmann, S., 2007. Atmospheric radiative effects of an in situ measured Saharan dust plume and the role of large particles. Atmospheric Chemistry and Physics 7, 4887–4903.
- Papadimas, C.D., Hatzianastassiou, N., Mihaloppoulos, N., Kanakidou, M., Katsoulis, B.D., Vardavas, I., 2008. Assessment of the MODIS collections C005 and C004 aerosol optical depth products over the Mediterranean basin. Atmospheric Chemistry and Physics 9, 2987–2999.
- Penner, J.E., Dickinson, R.E., O'Neill, C.A., 1992. Effects of aerosols from biomass burning on the global radiation budget. Science 256, 1432–1434.
- Penner, J.E., Zhang, S.Y., Chuang, C.C., 2003. Soot and smoke may not warm climate. Journal of Geophysical Research 108, D21. doi:10.1029/2003JD003409.
- Prasad, A.K., Singh, R.P., 2007. Comparison of MISR-MODIS aerosol optical depth over the Indo-Gangetic basin during the winter and summer seasons (2000–2005). Remote Sensing of Environment 107, 109–119.
- Prasad, A.K., Singh, S., Chauhan, S.S., Srivastava, M.K., Singh, R.P., Singh, R., 2007. Aerosol radiative forcing over the Indo-Gangetic plains during major dust storms. Atmospheric Environment 41, 6289–6301.
- Qureshi, S., 2010. The fast growing megacity Karachi as a frontier of environmental challenges: urbanization and contemporary urbanism issues. Journal of Geography and Regional Planning 3, 306–321.
- Ramanathan, V., Crutzen, P.J., Kiehl, J.T., Rosenfeld, D., 2001. Aerosol, climate, and hydrological cycle. Science 294, 2119–2124.
- Ranjan, R.R., Joshi, H.P., Iyer, K.N., 2007. Spectral variation of total column aerosol optical depth over Rajkot: a tropical semi-arid Indian station. Aerosol and Air Quality Research 7, 33–45.
- Remer, L.A., Kaufman, Y.J., Tanré, D., Mattoo, S., Chu, D.A., Martins, J.V., Li, R.R., Ichoku, C., Levy, R.C., Kleidman, R.G., Eck, T.F., Vermote, E., Holben, B.N., 2005. The MODIS aerosol algorithm, products and validation. Journal of Atmospheric Science 62, 947–973.
- Sarkar, S., Chokngamwong, R., Cervone, G., Singh, R.P., Kafatos, M., 2006. Variability of aerosol optical depth and aerosol forcing over India. Advances in Space Research 37, 2153–2159.
- Smart, J.C.R., Hicks, K., Morrissey, T., Heinemeyer, A., Sutton, M.A., Ashmore, M., 2011. Applying the ecosystem service concept to air quality management in the UK: a case study for ammonia. Environmetrics 22. doi:10.1002/env.1094 n/a.
- Tanré, D., Kaufman, Y.J., Herman, M., Mattoo, S., 1997. Remote sensing of aerosol properties over oceans using the MODIS/EOS spectral radiances. Journal of Geophysical Research-Atmospheres 102, 16971–16986.
- Torres, O., Bhartia, P.K., Herman, J.R., Sinyuk, A., Ginoux, P., Holben, B.N., 2002. A long-term record of aerosol optical depth from TOMS observations and comparison to AERONET measurements. Journal of Atmospheric Science 59, 398–413.
- Tripathi, S.N., Dey, S., Chandel, A., Srivastava, S., Singh, R.P., Holben, B.N., 2005. Comparison of MODIS and AERONET derived aerosol optical depth over the Ganga Basin, India. Annales Geophysicae 23, 1093–1101.
- Vermote, E.F., Roger, J.C., Sinyuk, A., Saleous, N., Dubovik, O., 2007. Fusion of MODIS–MISR aerosol inversion for estimation of aerosol absorption. Remote Sensing of Environment 107, 81–89.
- Wang, J., Christopher, S.A., 2003. Intercomparison between satellite-derived aerosol optical thickness and PM2.5 mass: implications for air quality studies. Geophysical Research Letters 30. doi:10.1029/2003GL018174.
- Xiao, N., Shi, T., Calder, C.A., Munroe, D.K., Berrett, C., Wolfinbarger, S., Li, D., 2009. Spatial characteristics of the difference between MISR and MODIS aerosol optical depth retrievals over mainland Southeast Asia. Remote Sensing of Environment 113, 1–9.