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Measurement and Analysis of Fine Particulate Matter ($PM_{2.5}$) in Urban Areas of Pakistan

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ABSTRACT

In order to assess the extent of air quality within the major urban environments in Pakistan, $PM_{2.5}$ pollutant has been analyzed during the period 2007–2011 in Islamabad; and 2007 to 2008 in Lahore, Peshawar and Quetta (high elevation, 1680 m MSL). Seasonal and diurnal variations of $PM_{2.5}$ mass concentration formation and accumulation within these areas have been analyzed. Air quality monitoring data and meteorological data (both QA/QCed) were obtained from Federal and Provincial Pakistan Environmental Protection Agencies. In Islamabad, the annual average $PM_{2.5}$ mass concentrations were 81.1 ± 48.4 µg/m^3, 93.0 ± 49.9 µg/m^3, 47.8 ± 33.2 µg/m^3, 79.0 ± 49.2 µg/m^3, and 66.1 ± 52.1 µg/m^3 during 2007 to 2011 respectively. Comparison of the four cities during summer 2007 to spring 2008 shows that all the four cities had $PM_{2.5}$ concentration exceeding the Pakistan National Environmental Quality Standards (annual average concentration of 25 µg/m^3; and 24 hourly average concentration of 40 µg/m^3) for ambient air. During the same time period, the highest seasonal $PM_{2.5}$ mass concentrations for Islamabad were observed as 98.5 µg/m^3 during spring 2008; 150.4 ± 87.9 µg/m^3; 104.1 ± 51.1 µg/m^3 and 72.7 ± 55.2 µg/m^3 for Lahore, Peshawar, and Quetta during fall 2007, respectively. Wind speed and temperature have a negative correlation with the mass concentration of $PM_{2.5}$. Diurnal profile for all the cities suggests an association of $PM_{2.5}$ with vehicular traffic. Back trajectory analysis conducted using the NOAA HYSPLIT model indicates that air trajectories, during high pollution episodes, influencing the urban regions commonly originate from either western India, especially in summer as part of the prevailing monsoon circulation; or are located in eastern Afghanistan. The source areas in Western India i.e., states of Gujarat, Rajasthan and Punjab have high concentration of industrial activities and crop residue burning, and are likely sources of enhanced $PM_{2.5}$ concentrations, in addition to the local sources.

Keywords: Pakistan; Fine particulate matter; Pollution; Meteorology.

INTRODUCTION

Air pollution has become a major environmental issue in Pakistan. Major causes of air pollution are rapid urbanization, economic growth and unplanned industrialization. Pakistan has an area of 796 095 km^2 with a population of 180.71 million (Government of Pakistan, 2012). The World Bank (2006) estimated an annual environmental degradation cost of about Rs. 365 billion or 6% of GDP for Pakistan. Estimated environmental health cost due to urban particulate matter pollution in the country is around Rs. 65 billion causing about 22,000 premature deaths among adults and 700 deaths among young children. Particulate matter is one of the significant pollutants with adverse health impacts like premature mortality, lungs and cardiovascular diseases (Pope *et al.*, 2002; NARSTO, 2004; Pope *et al.*, 2009; Moldanová *et al.*, 2011; Silva *et al.*, 2013). Indoor air pollution costs approximately Rs. 67 billion causing 28,000 deaths and 40 million cases of respiratory diseases annually (World Bank, 2006).

In urban areas of Pakistan, major sources of fine particulates are automobiles exhaust, combustion of fossil fuels/biomass, power plants and industrial emissions. Vehicles with two-stroke engines are inefficient in burning fuel and contribute to emission of fine particulate matter. Two-stroke vehicles had a growth of 117 percent in 2010–11 as compared to the year 2000–01 (Government of Pakistan, 2012). Continued increase in the number of two-stroke vehicles and diesel driven heavy duty vehicles add up to emission of pollutants into the atmosphere (ADB and CAI-Asia, 2006; World Bank, 2006). Sulfur content is 0.5–1% in diesel and is 1–3.5% in furnace oil leading to high emissions of sulfur dioxide and particulate matter (ADB and CAI-Asia, 2006).

Particulate matter consists of various organic and inorganic species with both natural and anthropogenic origin. Fine particles ($PM_{2.5}$) are composed of constituents like elemental

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carbon, organic carbon compounds, sulfates, nitrates, ammonium, heavy metals, H^+ and condensed metal vapours (NARSTO, 2004). Precursor gases for secondary fine particulates are sulfur dioxide (SO_2), nitrogen oxides (NO_x = NO + NO_2), volatile organic compounds (VOCs) and ammonia (NH_3) emitted from a number of natural and anthropogenic sources (NARSTO, 2004; Hidy and Pennell, 2010). Oxidation of SO_2 and NO_x leads to the formation of secondary inorganic PM i.e., sulfates and, nitrates, however, oxidation of VOCs emitted by transportation, industrial and biogenic sources contribute to the formation of organic carbon fraction of particulates (NARSTO, 2004; Zhang *et al.*, 2008; Jacob and Winner, 2009; Stone *et al.*, 2010). Ammonia enhances this conversion process (Aneja *et al.*, 2009). About 90% of the black carbon aerosols during winter season in Lahore, Pakistan, are present in the form of $PM_{2.5}$ contributing to approximately 5–15% of $PM_{2.5}$ present in the atmosphere (Viidanoja *et al.*, 2002; Husain *et al.*, 2007). The ambient concentrations of pollutants are largely dependent on meteorological conditions as well as chemical processes within the atmosphere involving air mass transport and mixing and solar radiation interactions (Aneja *et al.*, 2001; Hidy and Pennell, 2010). High relative humidity and low temperature in presence of excess ammonia give a significant rise to particulate ammonium nitrate (Pitchford, *et al.*, 2009), however, nitrate particulates decrease with increase in temperature (Aw and Kleeman, 2003; Jacob and Winner, 2009). An increase in temperature causes higher sulfate particulate concentrations due to faster SO_2 oxidation (Kleeman, 2008; Liu *et al.*, 2009). Precipitation is another factor affecting the lifetime of particulate matter in the atmosphere (Jacob and Winner, 2009). Meteorological conditions, specifically winter fog, enhance the level of pollution in some urban areas of Pakistan (Hameed *et al.*, 2000; Biswas *et al.*, 2008). The level of primary $PM_{2.5}$ depends more on the source distribution than on meteorological conditions (Liu *et al.*, 2009), however, meteorological factors like surface-based inversions are quite significant in accumulation of pollutants in certain areas (Incesik, 1996).

Pakistan National Environmental Quality Standards (NEQS) for Ambient Air set the 24-hour limit for $PM_{2.5}$ at 40 µg/m^3, and the annual and hourly average has been set as 25 µg/m^3. A revised 24-h limit of 35 µg/m^3 and annual and hourly average of 15 µg/m^3 have become effective from 1st January, 2013 (Pak-EPA, 2010). In this study, the mass concentration of $PM_{2.5}$ has been compared with the NEQS applicable before January, 2013.

The objective of this study is to characterize the mass concentration of $PM_{2.5}$ in four major cities of Pakistan i.e., Islamabad, Lahore, Peshawar and Quetta (high elevation city) in order to investigate the seasonal and diurnal variations, and the effect of meteorological parameters on the level of $PM_{2.5}$ air pollution; and examine the origin of air masses i.e., back-trajectory analysis. The study would be significant for regulatory agencies to develop control strategies in order to improve the air quality of the urban environments of Pakistan. Moreover, this data set would be of immense value to the urban, regional, and global air quality modeling community.

MATERIALS AND METHODS

Description of Sampling Sites

The air quality monitoring was carried out in four major cities of Pakistan i.e., Islamabad, Lahore, Peshawar and Quetta. The sampling cities are indicated in Fig. 1:

Islamabad

Islamabad is the capital city of Pakistan and is located at 33°26′N 73°02′E, with Margalla Hills surrounding the city from two sides (Siddique *et al.*, 2012). It has a population of approximately 1.15 million. The average elevation of Islamabad is 507 metres above sea level with lot of variation having the highest elevation of 1 604 meters. Islamabad has a semi-arid climate with warm to hot humid summers with monsoon season and a cold winter. The total area of Islamabad is 906 km^2. An extended area of 2 717 km^2 including Margalla Hills in the north and northeast has also been specified within the jurisdiction of Islamabad. Air monitoring station is located at the Central Laboratory for Environmental Analysis and Networking (CLEAN), Sector H-8/2. The height of the monitoring station is 2.5 m from the ground. The area consists of official buildings and an industrial estate at a distance of about 2–3 km.

Lahore

Lahore is located at 31°32′N 74°22′E with a municipal area of 332 km^2, which has been extended to 1 000 km^2 due to urbanization. Lahore is the second largest city of Pakistan with a population of about 9.01 million (Bureau of Statistics, 2012). There are approximately 2.7 million motor vehicles and 1986 industries in the city (Bureau of Statistics, 2012). Vehicular and industrial emissions are the main sources of air pollution in the city (Stone *et al.*, 2010). Lahore City is located at an elevation of 217 meters above sea level. The city is characterized by hot semi-arid climate with monsoon season and dry and warm winters. The monitoring site is located in Township, a typical residential area of Lahore city. The height of the monitoring station from the ground is 2.5 m. The major pollution sources in the area are traffic movement and textile industries at a distance of about 3–5 km. Some farmlands are also located on the eastern side of the monitoring station.

Peshawar

Peshawar is the capital city of Khyber-Pakhtunkhwa (KP) province and lies on the Iranian plateau at 34°01′N 71°35′E. It has a population of about 3.6 million. The city has an area of 1 257 km^2 and is located at an elevation of 359 meter above sea level. Semi-arid climate is a characteristic of Peshawar. The air monitoring station is installed at the rooftop of KP-EPA building located at Khyber Road, Peshawar. The height of the monitoring station from the ground is 10 m. The monitoring station is located about 2 km from a traffic congestion intersection. Major sources of air pollution are vehicular emissions.

Quetta

Quetta (the high elevation city) is the provincial capital

Fig. 1. Physical Map of Pakistan showing the sampling sites in Islamabad, Lahore, Peshawar and Quetta.

of the Balochistan Province of Pakistan located at 30°13′N 67°01′E. Quetta is a bowl shaped valley with an elevation of 1680 meters surrounded by mountain ranges with peak height above ~3000 meters above sea level (Muhammad *et al.*, 2006). Quetta has an area of 2 653 km^2 with the population of about 0.9 million inhabitants. Quetta has a semi-arid climate and it does not experience monsoon rainfalls. Air monitoring station is installed at the rooftop of municipality building in Zarghoon Town, Quetta. Major pollution sources in the vicinity of the monitoring station are vehicular emissions. Moreover, brick kilns and an industrial estate lie at a distance of about 8–10 km.

Experimental Methods

Data Collection

The air quality data (QA/QCed) for this research work were obtained from Pakistan Environmental Protection Agency (Pak-EPA). $PM_{2.5}$ is measured by Dust Analyzer (Horiba Ltd; Model APDA-370) with 0–5 mg/m^3 range through β-ray absorption method (ISO6349). Moreover, hourly air quality monitoring data for five other pollutants (carbon monoxide (CO), oxides of nitrogen (NO_x), sulfur dioxide (SO_2), ozone (O_3) and hydrocarbons (total hydrocarbons, non-methane hydrocarbons and methane) are determined using specific and prescribed analyzers in the automated air monitoring stations); and meteorological data were collected for five years (2007–2011) using automated fixed and mobile air monitoring stations. The air monitoring stations are equipped with ambient air analyzers, Combined Wind Vane, Anemometer (Koshin Denki Kogyo Co, Ltd. Model KVS 501), Thermo hygrometer (Koshin Denki Kogyo Co, Ltd. Model HT-010), Solar Radiation Meter (Koshin Denki Kogyo Co, Ltd. Model SR-010) and Data Logging System (Horiba, Ltd. Model Special). Data Logging systems at Federal and Provincial EPAs retrieve the ambient air quality data from air monitoring stations through data processing software.

Seasons have been specified as winter (December–February), spring (March–May), summer (June–August), and fall (September–November). Seasonal average has been calculated in order to find out the variation of $PM_{2.5}$ mass concentration in various seasons.

Back Trajectory Modeling

Backward air trajectories (48 hours) were determined by using the Hybrid-Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model which has been developed by the US National Oceanic and Atmospheric Administration's (NOAA) Air Resources Laboratory (ARL). Archived three-dimensional meteorological data is used by HYSPLIT model to compute the trajectories. Gridded Meteorological Data Archives from Global Data Assimilation System (GDAS) of National Center for Environmental Prediction (NCEP)/National Center for Atmospheric Research (NCAR) was used to calculate the back trajectories. The trajectories were computed for a height of 500 m AGL, 1000 m AGL and 1500 m AGL for different time durations. Label interval was set to be six hours to track the path of trajectory. The back trajectories were calculated for Pakistan with a buffer zone including some part of China, India, Afghanistan, Iran and Arabian Sea.

RESULTS AND DISCUSSION

Spatial and Temporal Variation of $PM_{2.5}$

Spatial and temporal variation in mass concentration of $PM_{2.5}$ in different cities gives an insight of the factors affecting the pollution levels with respect to geographical location and time. Fig. 2(a) provides the average annual and seasonal $PM_{2.5}$ mass concentration in Islamabad during

2007–2011. In Islamabad, average annual $PM_{2.5}$ mass concentrations were 81.1 ± 48.4 µg/m^3, 93.0 ± 49.9 µg/m^3, 47.8 ± 33.2 µg/m^3, 79.0 ± 49.2 µg/m^3 and 66.1 ± 52.1 µg/m^3 during 2007 to 2011 respectively. The highest hourly average concentrations were observed as 303 µg/m^3 during December 2007, 495.0 µg/m^3 during November 2008, 259.8 µg/m^3 during September 2009, 456.0 µg/m^3 during October 2010 and 379.0 µg/m^3 during January 2011. Major sources of air pollution in Islamabad are rapid urbanization, vehicular and industrial emissions, construction activities, emissions from brick kilns located on the outskirts of Islamabad (Qadir *et al.*, 2012; Siddique *et al.*, 2012) and forest fires in the Margalla Hills. There were about eighty wildfire incidents in the Margalla Hills during summer months of 2006–2010 (The Express Tribune, 2011) contributing to the $PM_{2.5}$ pollution during summer months. Some dust storms also contributed to the $PM_{2.5}$ burden in Islamabad city during the monitoring period. A dust storm of 148 km/h had impacted Islamabad on July 14, 2007 (Pakistan Weather Portal, 2013); and another storm with an intensity of 130 km/h struck the city on June 23, 2010 (GEO Pakistan, 2010). Advection of air pollution from nearby city Rawalpindi also increases the pollution level in Islamabad. Biomass combustion for space heating in the suburban areas of Islamabad increases the mass concentration of $PM_{2.5}$ during winter season. Moreover, Margalla Hills also restricts the dispersion of pollutants leading to buildup of pollutant levels in Islamabad.

Fig. 2(b) provides a comparison of annual and seasonal $PM_{2.5}$ mass concentration among Islamabad, Lahore, Peshawar and Quetta during summer 2007–spring 2008 (during this period all the four cities have overlapping data). Data shows that Lahore is the most polluted city among all the urban environments with the highest $PM_{2.5}$ mass concentration in each of the seasons. Furthermore, the average annual and seasonal $PM_{2.5}$ mass concentrations in all the cities, during the whole year, remained above the Pakistan NEQS.

The annual average $PM_{2.5}$ mass concentration in Islamabad has been observed to be 81.2 ± 47.4 µg/m^3 and the highest

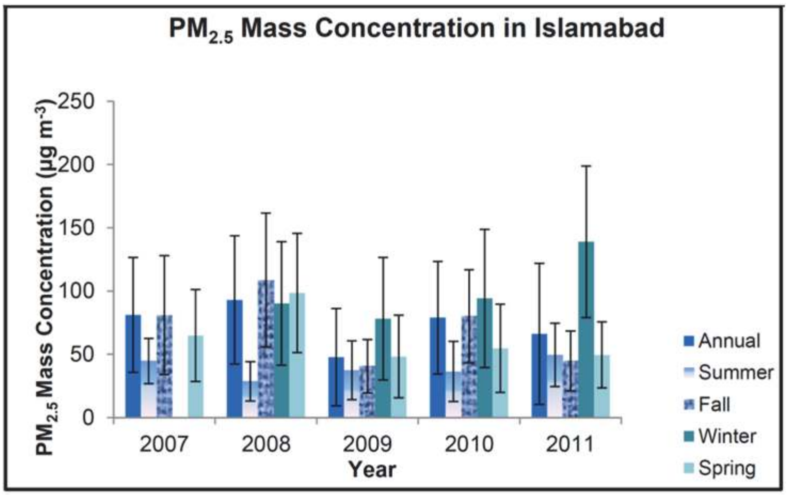

Fig. 2(a). Annual and Seasonal Average $PM_{2.5}$ Mass Concentration (µg/m^3) in Islamabad during 2007–2011 (±1 standard deviation is also shown in the figure).

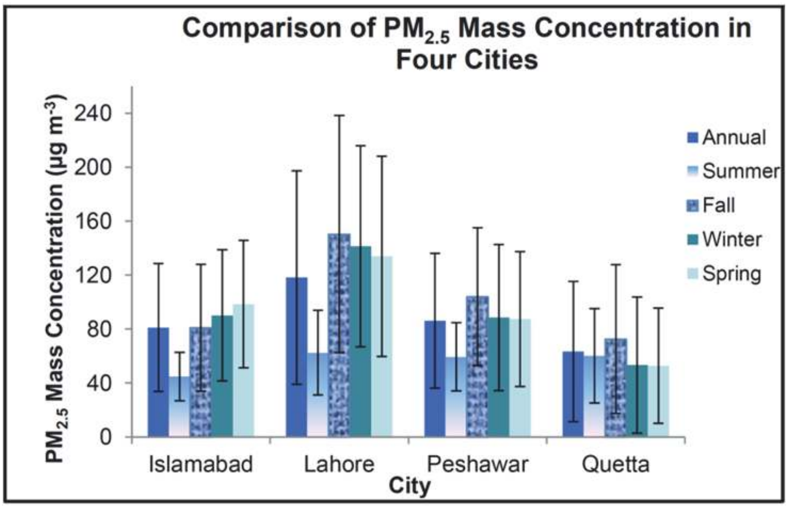

Fig. 2(b). Comparison of Annual and Seasonal Average $PM_{2.5}$ Mass Concentration (µg/m^3) in Islamabad, Lahore, Peshawar and Quetta during Summer 2007–Spring 2008 (±1 standard deviation is also shown in the figure).

seasonal average $PM_{2.5}$ mass concentration for Islamabad was 98.5 µg/m^3 during spring 2008. Fuel burning for space heating in surrounding rural areas is a source of sulfate particulates. In addition, atmospheric dispersion of $PM_{2.5}$ is decreased during winter season due to lower mixing depth and lower wind speed. Winter fog is another phenomenon restricting the dispersion of pollutants away from the city. Lower concentration of $PM_{2.5}$ in summer than in winter may also be explained by the heavy monsoon rainfall during the months of July and August (Sadiq and Qureshi, 2010).

The annual average $PM_{2.5}$ mass concentration for Lahore was observed to be 118.3 ± 79.1 µg/m^3 and the highest seasonal averaged $PM_{2.5}$ mass concentration of 150.5 ± 87.9 µg/m^3 was observed during Fall, 2007. Primary sources of $PM_{2.5}$ in Lahore are diesel emission, biomass burning, coal combustion, two-stroke vehicles and industrial activities (Dutkiewicz *et al.*, 2009; Stone *et al.*, 2010). Crop residues burning in Punjab, a region spanning northwestern India and eastern Pakistan during October and November to prepare fields for the next year's cultivation increases the concentration of $PM_{2.5}$ in ambient air during fall (Haq *et al.*, 2014; Singh and Kaskaoutis, 2014). Peak values of $PM_{2.5}$ in winter season may be attributed to primary emissions from combustion-related sources, which increase during the winter season for space heating purposes. Lodhi *et al.* (2009) has also documented about four times higher $PM_{2.5}$ mass concentration during winter than those observed in other seasons. Mixing height of about 250 m during night time and 1000 m during day time in winter season (Husain *et al.*, 2007) may be another factor for accumulation and increase in concentration of pollutants within this area. Lower mixing height traps the pollutants within a particular area restricting the dispersion of pollutants to other areas. Long-lasting episodes of stagnation accompanied by fog in Lahore are quite favorable for increased formation and accumulation of sulfate particulates (Biswas *et al.*, 2008). Wet deposition mechanism is also more pronounced during summer season (Sadiq and Qureshi, 2010).

The annual average $PM_{2.5}$ mass concentration for Peshawar was found to be 86.2 ± 50.0 µg/m^3 and the highest seasonal $PM_{2.5}$ mass concentration observed during fall, 2007 was 104.1 ± 51.1 µg/m^3. Vehicular and industrial emissions are the primary sources for $PM_{2.5}$ mass concentration in Peshawar (Khan *et al.*, 2008). Moreover, increased number of brick kilns situated in and around the city (Jan *et al.*, 2013) has led to high level of $PM_{2.5}$ as these kilns use low quality of coal and tyre burning as fuel. Wood burning during winter season for space heating has a great influence on the ambient mass concentration of $PM_{2.5}$ during winter season (Sandradewi *et al.*, 2008). High $PM_{2.5}$ mass concentration during winter may be attributed to the atmospheric stability under low temperature and lower inversion layer. Such conditions lead to accumulation of $PM_{2.5}$ which has a higher residence time as compared to PM_{10}. The scavenging effect of monsoon rainfall may also be considered a factor for less $PM_{2.5}$ mass concentration in summer than in winter. High $PM_{2.5}$ concentration during fall season may be attributed to burning of agricultural residue (Ali *et al.*, 2014). The annual average $PM_{2.5}$ mass concentration in Quetta was found to be 63.3 ± 52.0 µg/m^3 and the highest seasonal average $PM_{2.5}$ mass concentration was observed to be 72.7 ± 55.2 µg/m^3 during Fall, 2007. Major sources of air pollution in Quetta include emissions from motor vehicles, increased usage of two-stroke vehicles, dairy farms diesel-driven heavy vehicles, stone crushing, coal-fired power plants, industrial activities and brick kilns (Faiz *et al.*, 1996; Muhammad *et al.*, 2006; Quetta District Government, 2011). The topography of Quetta is favorable for accumulation of $PM_{2.5}$ and its precursor gases subsequently leading to photochemical formation of secondary $PM_{2.5}$ as well. Quetta remains engulfed by a thick layer of smog during winter season (Quetta District Government, 2011). High winter $PM_{2.5}$ concentrations are largely due to a combination of meteorology and an increase in primary combustion sources for space heating purposes. During summer months, low pressure prevails over Quetta due to which dust storms generated in deserts of Iran are transported to Quetta (Muhammad *et al.*, 2006). Higher $PM_{2.5}$ concentrations during summer 2007 may also be attributed to dust storm arriving in Quetta on July 23, 2007 (Wikipedia, 2013).

All of these cities are under the influence of extensive anthropogenic activities leading to higher $PM_{2.5}$ mass concentration. Higher $PM_{2.5}$ mass concentration during the fall season is due to burning of agricultural residue in the surrounding areas during the months of September, October and November (SUPARCO, 2009). Mansha *et al.* (2011) has also reported higher $PM_{2.5}$ mass concentrations during fall and winter seasons as compared to summer. In winter season, elevated levels may be associated with increased coal and biomass burning for heating purposes within the cities and nearby rural areas. Inversion layers suppress the vertical dispersion and diffusion of pollutants which leads to elevated levels of pollutants during winter season. The difference between winter and summer $PM_{2.5}$ mass concentration seems to be due to a combination of increased burning of fossil fuel i.e., coal for space heating purposes in winter season; and meteorological conditions i.e., shallow planetary boundary layer (Husain *et al.*, 2007), low precipitation level in cities other than Quetta and comparatively stable conditions.

Diurnal Profile of $PM_{2.5}$

Annual Averaged Diurnal Profile

Fig. 3 shows the integrated average hourly diurnal profile of $PM_{2.5}$ mass concentration in Islamabad, Lahore, Peshawar and Quetta during 2007–2011. Diurnal profiles of all four cities follow a similar pattern of variation – in general, the first peak occurs between 7.00 a.m. to 10.00 a.m.; and the second maximum in $PM_{2.5}$ mass concentration occurs during about 9:00 p.m. to 1:30 a.m. This diurnal pattern shows that even the lowest levels are above the Pakistan hourly NEQS for $PM_{2.5}$. The daytime peaks of $PM_{2.5}$ mass concentration correlate well with the traffic pattern.

Night-time $PM_{2.5}$ high concentration is due to a combination of heavy duty vehicles movement in the city and the development of night time inversion layer (diurnal meteorological changes). During winter season, high night time concentration of $PM_{2.5}$ may also be attributed to the increased use of biomass burning (fuelwood) for heating

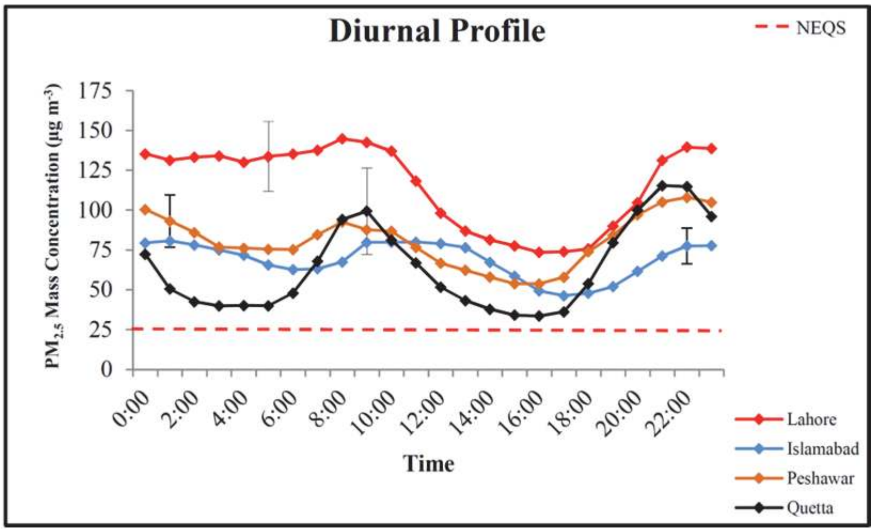

Fig. 3. Integrated Average Diurnal Profile of $PM_{2.5}$ Mass Concentration (μg/m^3) in Islamabad, Lahore, Peshawar and Quetta for 2007–2011 (±1 standard deviation is also shown in the figure).

purposes. The early morning elevation in $PM_{2.5}$ mass concentration is owing to an increase of the traffic density.

Seasonal Averaged Diurnal Profile

The seasonally averaged diurnal profile has been determined for each city separately in order to find out any variation in diurnal pattern of $PM_{2.5}$ mass concentration during different seasons. Fig. 4 shows the seasonal averaged diurnal profiles for Islamabad, Lahore, Peshawar, and Quetta. In general there is a bimodal distribution in the diurnal $PM_{2.5}$ seasonal variation. The level of $PM_{2.5}$ concentration has a peak during the morning associated with increase in the traffic movement (i.e., mobile emissions and dust entrainment); and a second peak associated with rush hour traffic and reduction in the planetary boundary layer height. Midnight high values of $PM_{2.5}$ may be attributed to heavy duty vehicles carrying goods into and through the city. The seasonally averaged diurnal profile shows that the concentration of $PM_{2.5}$ is highest during winter season in Islamabad, Lahore, and Peshawar associated with usage of coal for heating purposes and long-lasting fog.

The seasonally averaged diurnal profile of $PM_{2.5}$ in Quetta (Fig. 4(d)) is different from other cities' $PM_{2.5}$ pattern in a way that the $PM_{2.5}$ mass concentration in Quetta during the winter season is lower than the other seasons. Quetta, being at high altitude (1680 m, MSL), has increased $PM_{2.5}$ mass concentration owing to high usage of biomass burning (fuel wood combustion) for space heating purpose in winter compared to other cities. However, meteorological conditions in Quetta vary with precipitation in winter due to western disturbance and no summer monsoon rainfall (Chaudhary, 1992). Winter rainfall may be considered as a factor contributing towards lower $PM_{2.5}$ mass concentration in winter than summer season. During summer months, stagnation and dry weather conditions prevail in Quetta leading to enhanced accumulation of $PM_{2.5}$ and its precursor gases within the city. The dust storms generated in the desert of Iran are another major contributor towards the pollution in Quetta during summer (Muhammad *et al.*, 2006).

Effect of Meteorology on $PM_{2.5}$

The $PM_{2.5}$ mass concentration in Islamabad, Lahore, Peshawar and Quetta has been correlated with meteorological variables in order to find out any possible association between ambient $PM_{2.5}$ mass concentration and meteorology in these cities.

$PM_{2.5}$ and Temperature

Fig. 5(a) shows the seasonal correlation of $PM_{2.5}$ with temperature in Islamabad. The figure shows that $PM_{2.5}$ has a negative correlation ($p \leq 0.01$; r = –0.2) with temperature in winter season and a positive association ($p \leq 0.01$; r = 0.07) with temperature during summer season. About 4% of the variance in $PM_{2.5}$ in winter and 0.4% variance during summer can be explained by its linear relationship with temperature. An increase in temperature causes decrease in nitrate particulates (Aw and Kleeman, 2003; Jacob and Winner, 2009) and formation of higher sulfate particulate concentrations due to faster SO_2 oxidation (Kleeman, 2008; Liu *et al.*, 2009). Fig. 5(b) gives the relationship between $PM_{2.5}$ and temperature in Lahore. The figure shows that 26% of the variance in $PM_{2.5}$ can be explained by temperature in winter months. During winter, $PM_{2.5}$ has a strong correlation with temperature ($p \leq 0.01$; r = –0.5). Fig. 5(c) shows that $PM_{2.5}$ has a negative correlation ($p \leq 0.01$; r = –0.1) with temperature in winter season and a positive correlation ($p \leq 0.01$; r = 0.1) during summer months. Negative correlation observed between $PM_{2.5}$ and temperature during winter in Islamabad, Lahore and Peshawar suggests that thermal inversion layers and fog may play a major role in elevation of $PM_{2.5}$ levels. Positive correlation between $PM_{2.5}$ and temperature may also be an indication of increased agricultural and biogenic emissions of ammonia and oxides of nitrogen (Tai *et al.*, 2010). Raja *et al.* (2010) has also reported low temperature as a significant factor contributing to the formation of ammonium nitrate particles in Lahore.

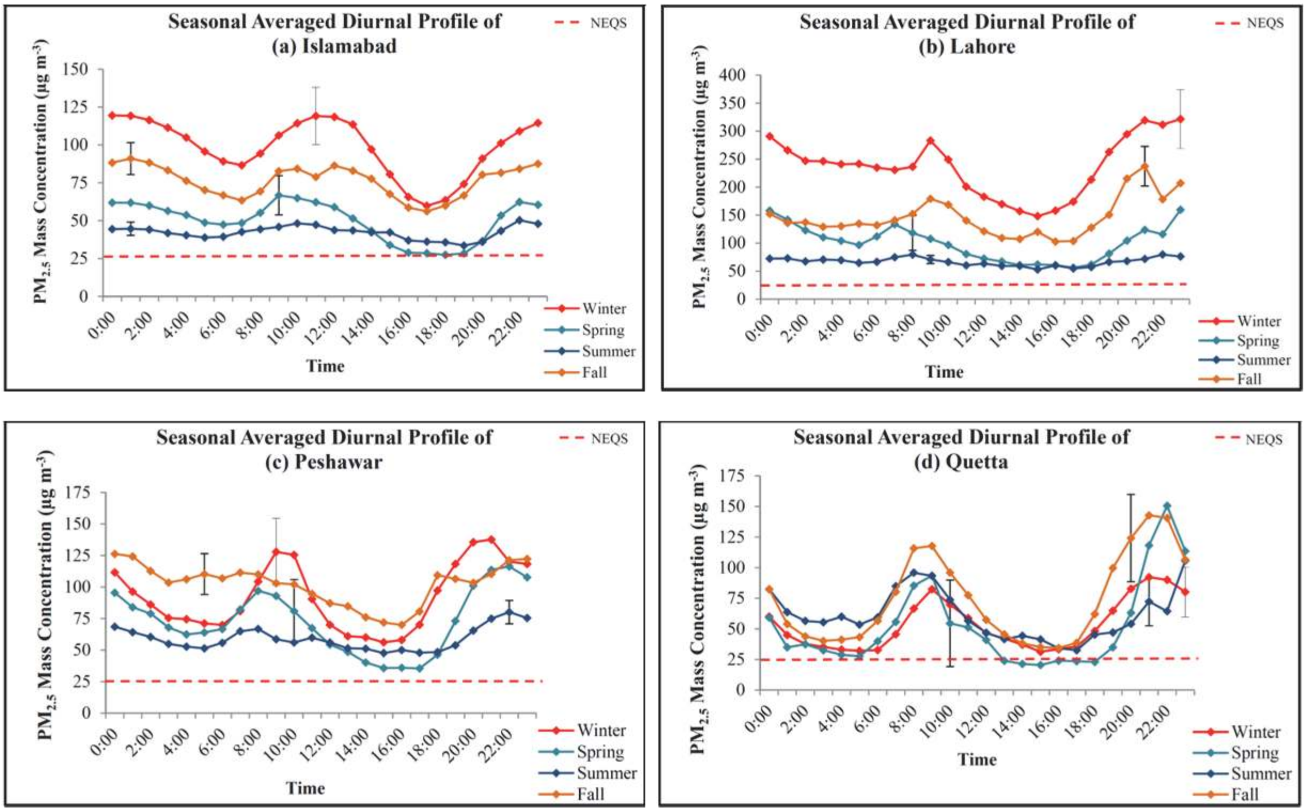

Fig. 4. Seasonal Averaged Diurnal Profile of $PM_{2.5}$ Mass Concentration in (a) Islamabad; (b) Lahore; (c) Peshawar; and (d) Quetta (±1 standard deviation is also shown in the figure).

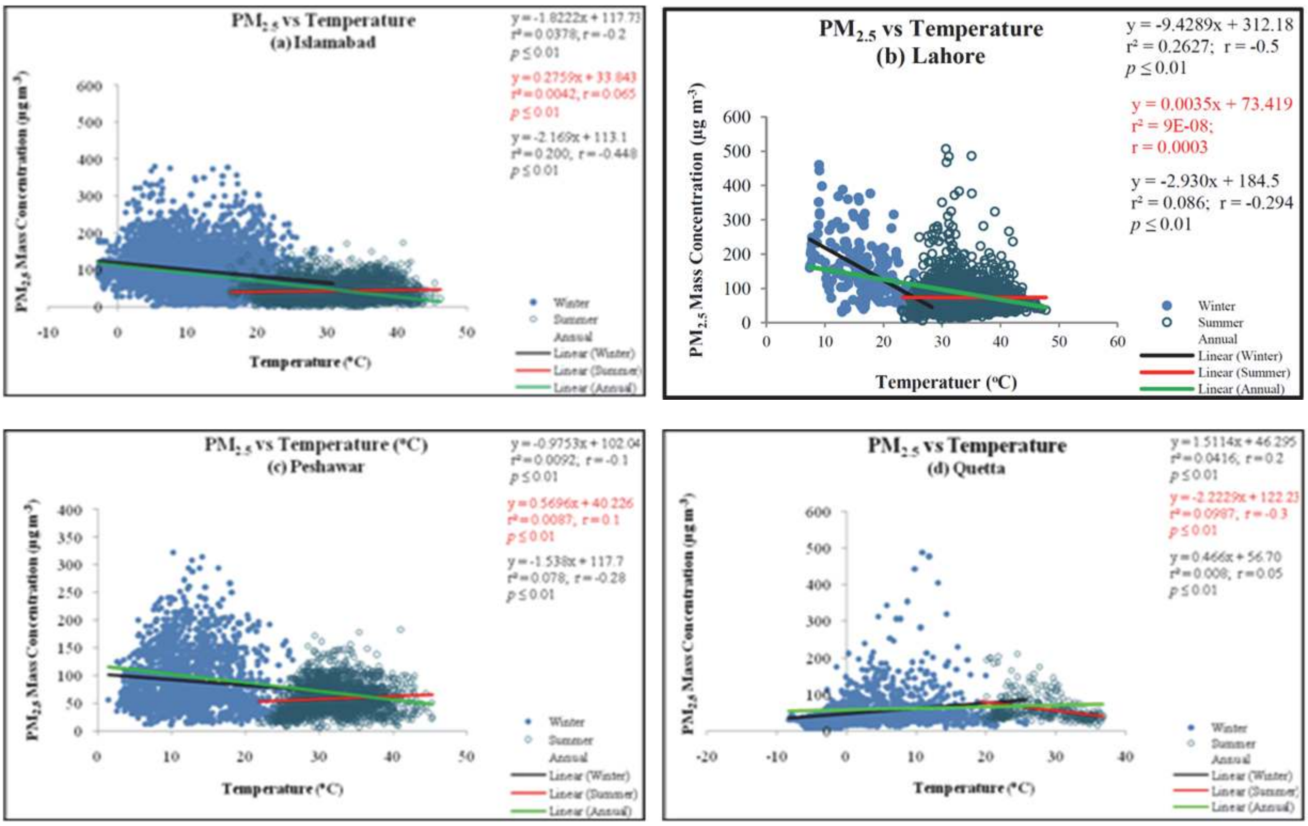

Fig. 5. Effect of Temperature on $PM_{2.5}$ Mass Concentration (μg/m^3) during 2007–2011 in (a) Islamabad; (b) Lahore; (c) Peshawar; and (d) Quetta.

Similar positive correlation of $PM_{2.5}$ and temperature during summer and a negative correlation during winter season have been found by Barmpadimos *et al.* (2012). However, the overall $PM_{2.5}$ mass concentration trend with temperature is negative for Islamabad, Lahore, and Peshawar. Tiwari *et al.* (2013) have also reported a negative correlation of $PM_{2.5}$ with temperature which depends on composition of the particulate matter. This is perhaps due to semi-volatile components such as nitrates and organics which are expected to decrease as they shift from the particle phase to gas phase at higher temperature (Sheehan and Bowman, 2001; Aw and Kleeman, 2003; Dawson *et al.*, 2007; Tsigaridis and Kanakidou, 2007; Kleeman, 2008).

Fig. 5(d) shows the relationship between $PM_{2.5}$ and temperature in Quetta. The regression analysis shows that about 4% of the variation in $PM_{2.5}$ is associated with temperature in winter season and 10% of the $PM_{2.5}$ variation is associated with temperature in summer months. Quetta, being a high elevation site, the association between $PM_{2.5}$ and temperature is unlike other cities. Here, $PM_{2.5}$ is positively correlated (r = 0.2) with temperature in winter season, whereas, these two variables are negatively correlated (r = –0.3) during summer. The correlation between $PM_{2.5}$ and temperature is statistically significant ($p \leq 0.01$) during both the seasons. Negative correlation observed between $PM_{2.5}$ and temperature during summer suggests that there may be more abundance of nitrate particles which get converted from particle to gas phase due to high summer temperatures (Dawson *et al.*, 2007). The positive correlation of $PM_{2.5}$ with temperature during winter may be due to the fact that the winter precipitation in the form of rainfall and snow works as a scavenger for $PM_{2.5}$ and perhaps plays an important role in decrease of $PM_{2.5}$ mass concentration with decrease in temperature during winter months justifying the positive relationship between these two variables.

$PM_{2.5}$ and Solar Radiation

Fig. 6(a) shows the correlation between $PM_{2.5}$ and solar radiation in Islamabad. The regression analysis provides the information that about 0.4% of variation of $PM_{2.5}$ during summer is associated with solar radiation and 0.7% variation in $PM_{2.5}$ during summer months depends on solar radiation. There is a correlation of r = 0.06 between $PM_{2.5}$ and solar radiation in winter and a correlation of r = 0.1 during summer. Fig. 6(b) shows that $PM_{2.5}$ has a significant ($p \leq 0.05$) correlation with solar radiation (r = –0.24) during winter season in Lahore, however, it does not have a good association (r = 0.03) with solar radiation during summer season. The regression line shows that 6% of the $PM_{2.5}$ variation is associated with solar radiation in winter and a negligible fraction of about 0.1% has dependence on solar radiation. Fig. 6(c) shows that $PM_{2.5}$ is negatively correlated ($p \leq 0.01$; r = –0.2) with solar radiation during winter season but there is a weak association (r = 0.06) between $PM_{2.5}$ and solar radiation during summer months. About 3% of the variation in $PM_{2.5}$ in winter may be explained by

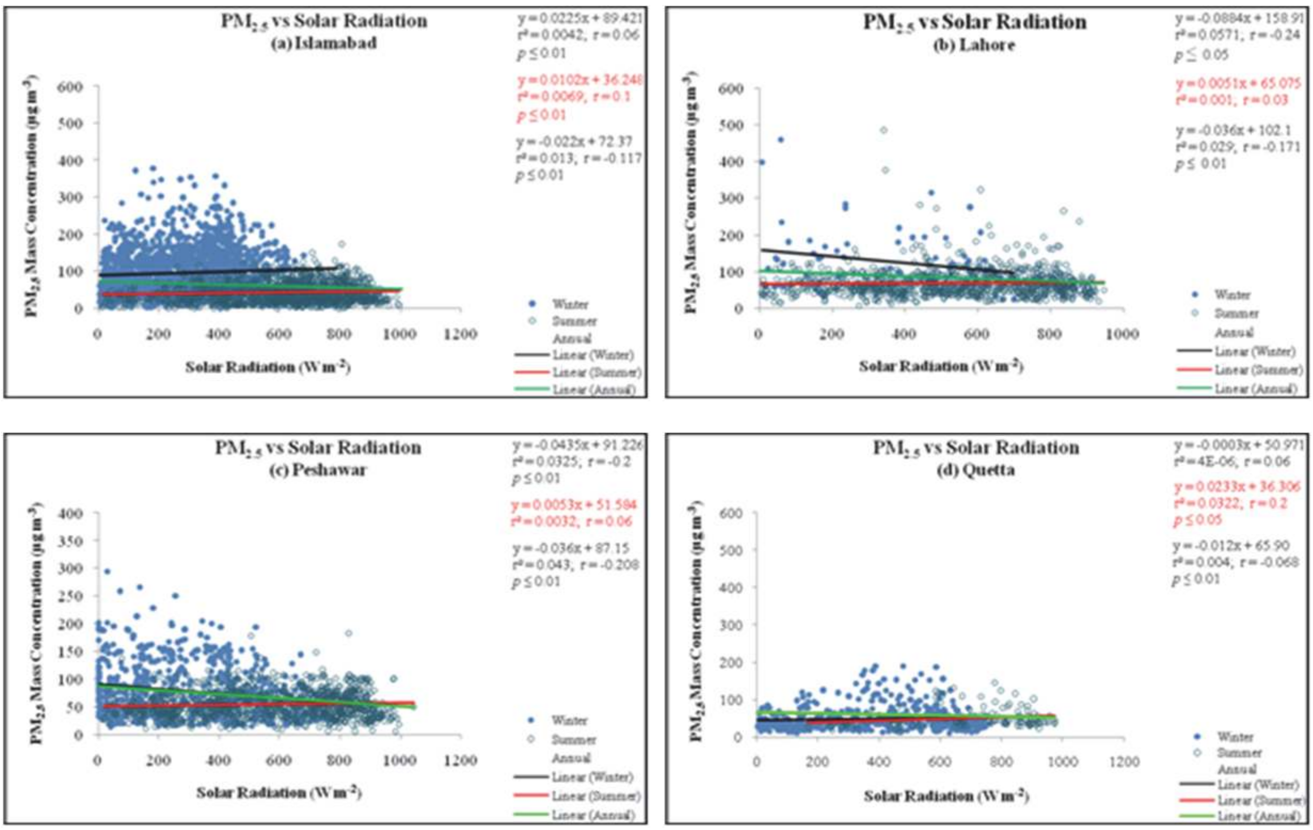

Fig. 6. Effect of Solar Radiation on $PM_{2.5}$ Mass Concentration ($\mu g/m^3$) during 2007–2011 in (a) Islamabad; (b) Lahore; (c) Peshawar; and (d) Quetta.

its linear relationship with solar radiation and 0.3% of $PM_{2.5}$ variation during summer is associated with solar radiation which is almost negligible. Fig. 6(d) shows that there is positive correlation of $PM_{2.5}$ and solar radiation during both winter and summer in Quetta. $PM_{2.5}$ has a statistically insignificant correlation (r = 0.06) with solar radiation during winter season and a good correlation ($p \leq 0.05$; r = 0.2) during summer season with 3% of variation in $PM_{2.5}$ caused by solar radiation.

$PM_{2.5}$ and Wind Speed

Wind speed and mixing depth have strong effect on particulate matter (Jacob and Winner, 2009). High wind speed results in dispersion of $PM_{2.5}$ mass concentration and stagnation leads to accumulation of $PM_{2.5}$ in a particular area. The stagnant conditions caused by low wind speed i.e., high pressure systems deteriorate the ambient air quality (Liao *et al.*, 2006; Leibensperger *et al.*, 2008). Figs. 7(a), 7(b), 7(c) and 7(d) show the effect of wind speed on level of $PM_{2.5}$ in Islamabad, Lahore, Peshawar and Quetta respectively. It has been observed that there is a statistically significant ($p \leq 0.01$) negative correlation between $PM_{2.5}$ and wind speed during winter and summer seasons in all the cities. In Islamabad, $PM_{2.5}$ has a negative correlation with wind speed in both the seasons (winter: r = –0.3 and summer: r = –0.17). About 10% of the $PM_{2.5}$ variation in winter and 3% of $PM_{2.5}$ variation is associated with the linear relationship of $PM_{2.5}$ with wind speed in Islamabad. Lower correlation between $PM_{2.5}$ and wind speed shows the contribution of local emission sources towards the ambient concentration of $PM_{2.5}$. $PM_{2.5}$ has a correlation of r = –0.5 during winter months and a correlation of r = –0.2 in summer in Lahore. About 30% of $PM_{2.5}$ variation in winter and about 4% of $PM_{2.5}$ variation is due to wind speed in Lahore city.

In Peshawar, $PM_{2.5}$ has a negative correlation of r = –0.3 with wind speed during winter months with dependence of about 10% of $PM_{2.5}$ variation on wind speed. There is a correlation of r = –0.2 between $PM_{2.5}$ and wind speed in summer season and 4% of $PM_{2.5}$ variation is associated with wind speed in the city. $PM_{2.5}$ and wind speed have a negative correlation of r = –0.13 in winter and a correlation of r = –0.25 in summer months in Quetta. About 1% of $PM_{2.5}$ variation in winter and 6% variations in summer are explained by its relationship with wind speed in Quetta. Correlation of $PM_{2.5}$ with wind speed is stronger in winter than in summer in all cities except Quetta. It indicates the contribution of local sources to high $PM_{2.5}$ mass concentration in Quetta during winter season. As Quetta is situated in a valley, there is stagnation in the air leading to accumulation of $PM_{2.5}$ mass concentration. Islamabad and Peshawar have also mountains which hinder the transport of $PM_{2.5}$. DeGaetano and Doherty (2004) has also shown a negative correlation of wind speed with fine particulate matter.

$PM_{2.5}$ and Vapor Pressure

Fig. 8(a) represents the correlation between $PM_{2.5}$ and

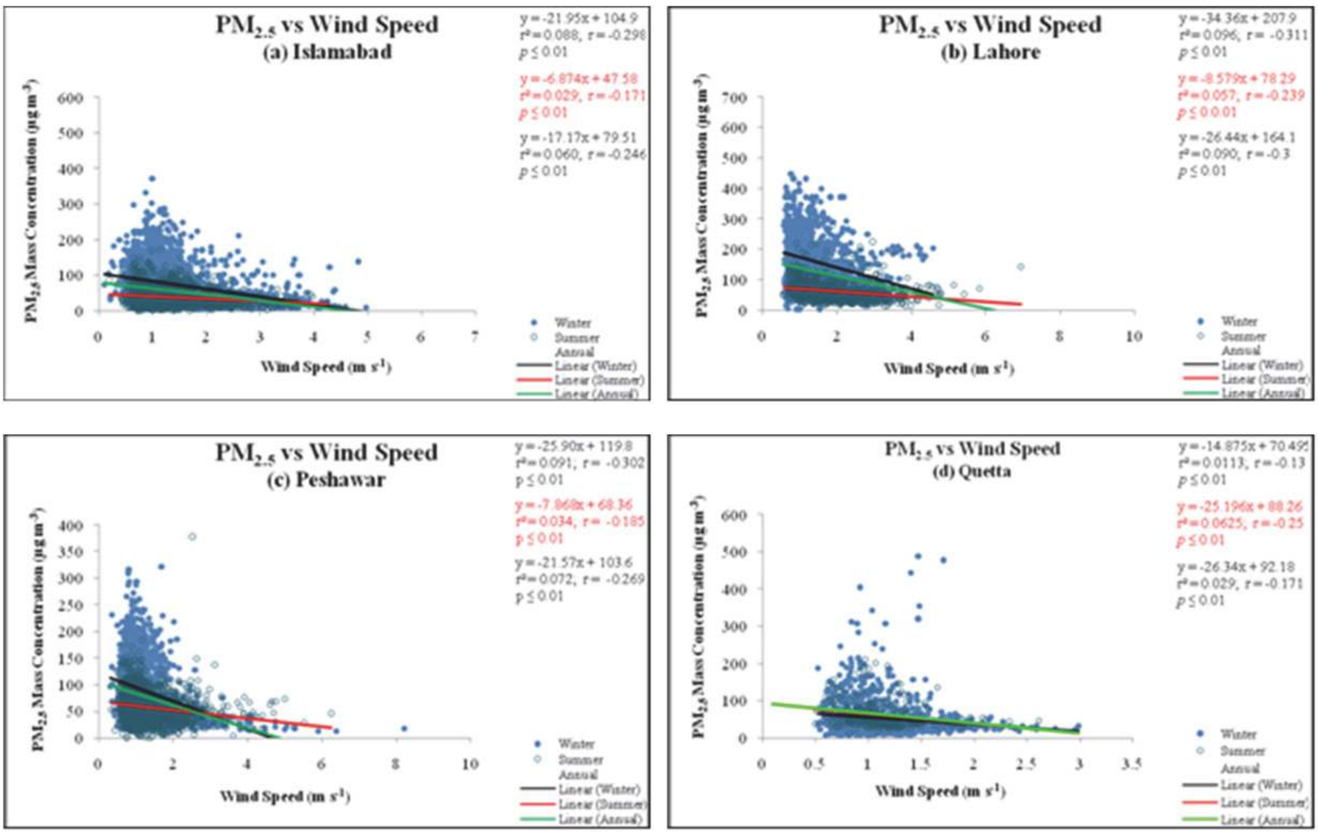

Fig. 7. Effect of Wind Speed on $PM_{2.5}$ Mass Concentration (µg/m^3) during 2007–2011 in (a) Islamabad; (b) Lahore; (c) Peshawar; and (d) Quetta.

vapour pressure for Islamabad. $PM_{2.5}$ has a negative correlation with vapour pressure ($p \leq 0.01$; r = –0.2) during winter months, however, it has a positive correlation with vapour pressure ($p \leq 0.01$; r = 0.05) during summer season. In winter season, about 4% of $PM_{2.5}$ variation is associated with vapour pressure whereas, about 0.3% of $PM_{2.5}$ variance is associated with its linear relationship; with vapour pressure during summer season. Fig. 8(b) shows the correlation of $PM_{2.5}$ with vapour pressure during both the seasons in Lahore. There is a negative correlation ($p \leq 0.01$; r = –0.23) between $PM_{2.5}$ and vapour pressure during winter season and a positive correlation (p ≤ 0.01; r = 0.07) during summer months. Regressions analysis shows that about 5% of the variation in $PM_{2.5}$ mass concentration is associated with vapour pressure in winter, whereas, only about 0.5% $PM_{2.5}$ variation is caused by vapour pressure during summer. Particulate matter and relative humidity are negatively correlated in Southwestern U.S. indicating the primary source origin (Wise and Comrie, 2005). High relative humidity and low temperature in presence of excess ammonia give a significant rise to formation of ammonium nitrate particles (Pitchford, *et al.*, 2009) in Lahore (Raja *et al.*, 2010).

Fig. 8(c) gives the correlation of $PM_{2.5}$ and vapour pressure in Peshawar. $PM_{2.5}$ has statistically significant ($p \leq 0.01$) positive correlation with vapour pressure during both the seasons i.e., r = 0.36 in winter and r = 0.22 in summer season. About 13% of the variation in $PM_{2.5}$ is related to its linear relationship with vapour pressure during winter months and about 5% of $PM_{2.5}$ variation is associated with vapour pressure in summer season. Fig. 8(d) shows that $PM_{2.5}$ has a positive correlation (r = 0.05) with vapour pressure during winter season, and a negative correlation (p ≤ 0.05; r = –0.12) during summer season in Quetta. As winter months are more humid, the positive correlation between these two variables during winter show that the anthropogenic emissions like nitrates and sulfates are contributing more towards the $PM_{2.5}$ pollution (Tai *et al.*, 2010). The negative correlation of $PM_{2.5}$ with vapour pressure shows the contribution of dust particles and elemental and organic carbon to $PM_{2.5}$ level during summer months (Wise and Comrie, 2005; Tai *et al.*, 2010). Regression analysis shows that there is weak association of $PM_{2.5}$ variation with vapour pressure i.e., 0.3% in winter and 1% in summer.

Linear Regression Analysis

Linear regression analysis has been conducted to determine the meteorological variables that explain the most variance in the overall data. Summary of linear regression analysis is given in Table 1. The regression analysis provides the extent to which meteorological conditions affect the concentration of $PM_{2.5}$ in the atmosphere. The regression analysis shows that the four meteorological variables accounted for 20% variance in $PM_{2.5}$ mass concentration in Islamabad. Meteorology seems to affect the $PM_{2.5}$ mass concentration in Islamabad, Lahore and Peshawar more during winter than in summer season. The meteorological factors accounted for

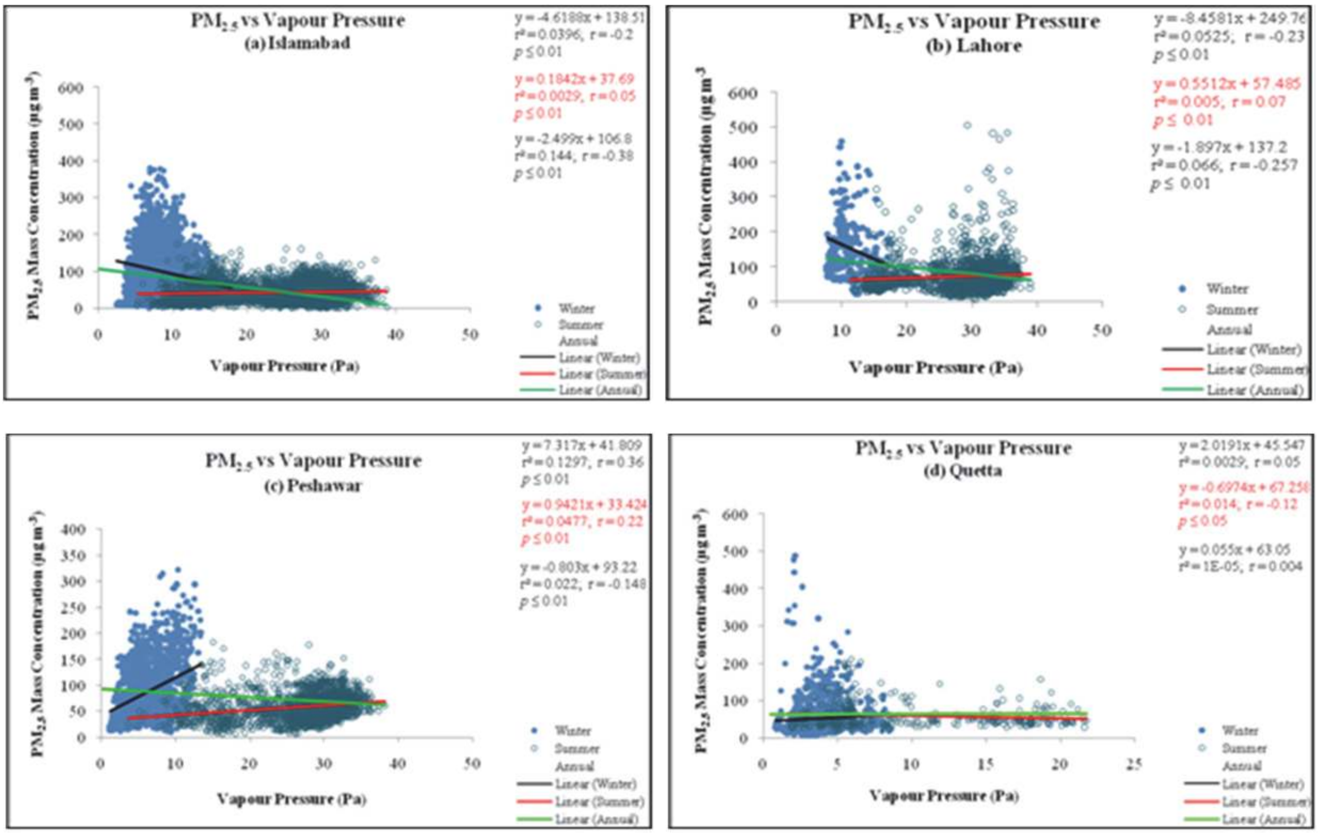

Fig. 8. Effect of Vapour Pressure on $PM_{2.5}$ Mass Concentration (µg/m^3) during 2007–2011 in (a) Islamabad; (b) Lahore; (c) Peshawar; and (d) Quetta.

Table 1. Linear Regression Analysis of $PM_{2.5}$ and Meteorological Variables for Islamabad, Lahore, Peshawar, and Quetta.

City	Season	Correlation Coefficient (R)	Variance Explained
Islamabad	Winter	0.457	21%
	Summer	0.211	4%
	Annual	0.447	20%
Lahore	Winter	0.453	20%
	Summer	0.251	6%
	Annual	0.432	19%
Peshawar	Winter	0.424	18%
	Summer	0.372	14%
	Annual	0.357	13%
Quetta	Winter	0.279	8%
	Summer	0.489	24%
	Annual	0.239	6%

approximately 19% variance in $PM_{2.5}$ mass concentration in Lahore city. About 13% variance in average $PM_{2.5}$ concentration in Peshawar is explained by the meteorological conditions. Annual average mass concentration of $PM_{2.5}$ in Quetta seems to be less affected (6%) by meteorology as compared to other cities. It has been observed that the effect of meteorology on $PM_{2.5}$ is more prominent during winter season in Islamabad (21%), Lahore (20%) and Peshawar (18%), however, Quetta (located at high elevation) city has a different scenario. Here, the meteorological parameters have a contribution of about 8% of the variance in $PM_{2.5}$ mass concentration during winter months, whereas, about 24% of variance in $PM_{2.5}$ is explained by meteorology during summer season.

Back Trajectory Analysis

The back trajectories for some of the high $PM_{2.5}$ pollution episodes have been calculated to find out the possible source regions influencing the ambient air quality of the four major cities of Pakistan (Fig. 9). Nine pollution episodes have been selected for back trajectory analysis. The trajectories for the three altitude levels studied were similar since trajectories end inside the boundary layer (500 m, 1000 m, 1500 m). So we have provided the back trajectories for only 1000m altitude. The back trajectories show that, during high $PM_{2.5}$ pollution episodes, the air masses are transported from eastern Afghanistan, eastern Iran or western India. Major pollution sources from eastern Afghanistan particularly Kabul and Jalalabad include vehicular and industrial emissions, biomass burning, use of diesel electric generators and burning of tyres (ADB and CAI-Asia, 2006). The air masses originating in western India are from the states of Gujarat, Rajasthan and Punjab, which have coal-fired power plants, industries and mechanized farming that are sources of particulate and gaseous emissions. Furthermore, two-stroke vehicles also aggravate the problem of $PM_{2.5}$ pollution to a great extent (Raja *et al.*, 2010; Guttikanda and Jawahar, 2011). Moreover, large scale crop residues burning during October and November in Punjab, a region spanning northwestern India and eastern Pakistan to prepare fields for the next year's cultivation increases the concentration of $PM_{2.5}$ in ambient air during fall in Eastern Pakistan (Singh and Kaskaoutis, 2014). Major pollution sources in Iran include vehicular emissions, stationary sources and desert storms (Torkian *et al.*, 2012).

Conclusion

This study attempts to characterize the $PM_{2.5}$ pollution in four major urban centers in Pakistan. The average $PM_{2.5}$ mass concentration is significantly higher in Lahore than the other three cities. All the four cities have average $PM_{2.5}$ concentration higher than the Pakistan NEQS. These high concentrations of $PM_{2.5}$ may be attributed to primary sources e.g., coal and fossil fuel combustion coupled with the meteorological conditions controlling the formation of secondary $PM_{2.5}$ and their dispersion within the troposphere. Analysis of high pollution episodes conducted using the NOAA HYSPLIT model indicates that the air trajectories influencing Lahore, Islamabad, Peshawar and Quetta commonly originate from western India, especially in summer as part of the prevailing monsoon circulation and eastern Afghanistan. These source areas (states of Gujarat, Rajasthan and Punjab) have high concentration of industrial activity and are likely sources of enhanced $PM_{2.5}$ concentration, in addition to the local sources. High $PM_{2.5}$ mass concentration in winter may be a result of increased local coal combustion activities due to more usage of coal and biomass for heating purposes. Furthermore, in low temperatures, nitrates may have been exchanged from gas-phase into particles which suggests that the ratio of nitrates particulate formation in winter to sulfate formation in summer is high. It may also indicate that the primary NO_x emissions are more than SO_2 emissions in these cities indicating higher contribution from vehicular emissions. Surface inversion layers and fog contribute to a great extent to hinder the dispersion of $PM_{2.5}$ in urban areas of Pakistan especially in Lahore where winter fog is extensive and at its maximum. Lower mixing height in winter season may be another factor for high $PM_{2.5}$ in winter as it does not allow dispersion of pollutants during prolonged low temperature periods. Significant daily variation has also been observed in $PM_{2.5}$ mass concentration with peaks in morning till noon and another peak with maximum values from evening to midnight. Among meteorological factors, temperature and wind speed show a negative correlation with $PM_{2.5}$ indicating the dispersion of pollutants by wind and accumulation of pollutants at low temperatures.

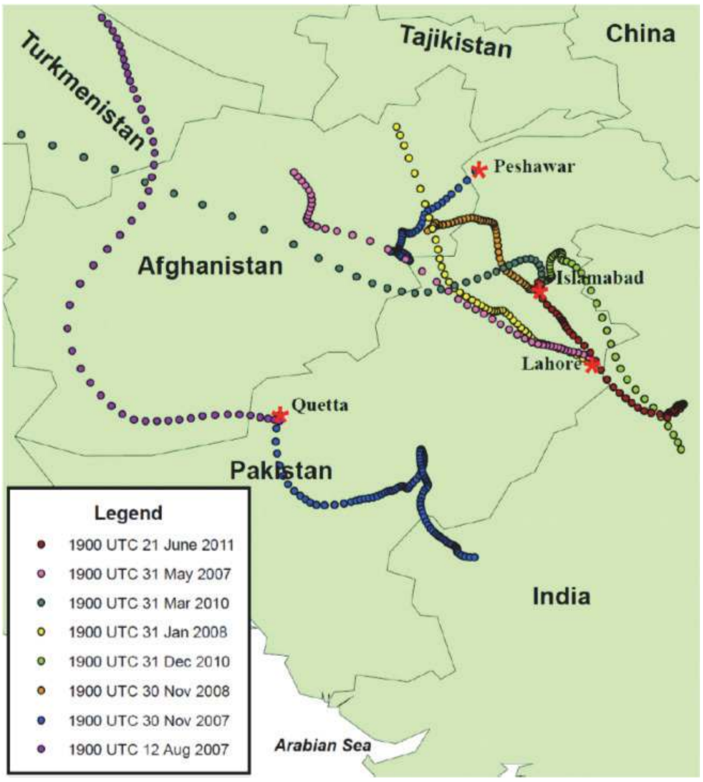

Fig. 9. Air parcel 48-hour back trajectories analysis for selected $PM_{2.5}$ and Ozone high pollution episodes during 2007–2011.

Particulate pollution has become an issue of great concern in Pakistan, and its control is a challenge for regulatory agencies. There is a need to strictly enforce the vehicular and industrial emission standards in order to control the elevated pollution levels.

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