

## Research Article

# Temporal Variation and Concentration Weighted Trajectory Analysis of Lead in PM<sub>10</sub> Aerosols at a Site in Central Delhi, India

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Ambient levels of lead (Pb) in PM<sub>10</sub> were studied at a site in Central Delhi for the period of one year during day and night. The annual mean concentration of lead has been observed as 625 and 1051 ng/m<sup>3</sup> during day and night time, respectively. The seasonal averaged concentrations of Pb have followed the order winter > postmonsoon > summer > monsoon. Highest levels of lead have been observed in winter with 31% samples exceeding the CPCB-NAAQS value as 1000 ng/m<sup>3</sup>. Lead levels during winter have been found to be 5.7 times higher than in monsoon, which might be attributed to prevailing meteorological conditions and more biomass burning. The low levels of Pb during summer might be attributed to its higher dispersion in the atmosphere. A sharp rise of Pb during postmonsoon might be linked to the local nonpoint sources, more biomass burning, and shifting of boundary layer. However, the higher concentrations of lead were observed during night time in all the seasons of the year as compared to those of the day time. To identify the potential source regions of Pb, Concentration Weighted Trajectories (CWT) have been plotted which showed higher influence of local sources during winter and postmonsoon while showing distant sources during summer.

## 1. Introduction

Among the various air pollutants, particulate matter plays an important role due to its effects on different atmospheric processes such as visibility, atmospheric chemistry, radiative balance, and human health [1, 2]. Trace metals associated with particulate matter may increase manyfold, by natural or anthropogenic sources [3–5]. At elevated concentrations, certain trace metals (Hg, Pb, Sn, Cd, etc.) show toxic effects on living beings [2, 6, 7]. Lead (Pb), as a well-known neurotoxin, shows high level of toxicity on living beings [2] and is the only metal listed in the EPA-NAAQS (<http://www.epa.gov/air/criteria.html>). Due to its adverse effects on human health, especially on pregnant women and children's growth and intelligence, study of Pb has been a topic of interest. Anthropogenic activities play a major role for the emission of lead into the atmosphere [8]. Once it enters into the atmosphere, it can be deposited on the surface and resuspended to reenter the atmosphere. Thus, it has high potential to remain in

the environment [9, 10]. This could increase human exposure to Pb causing adverse health effects [11].

Lead is released into the atmosphere mainly in the gaseous form during high temperature anthropogenic processes such as metal processing, fuel combustion, nonferrous metal production, or waste incineration [12]. This further condenses in the atmosphere and forms smaller particles [13]. These particles are too small to sediment at a significant rate and so remain suspended in atmosphere for longer duration of time and transported over vast distances. They can readily penetrate into the lungs and affect the human respiratory system. The main species of Pb are chlorides, oxides, and sulfates, mainly released by oil combustion (PbO), nonferrous metal production (PbSO<sub>4</sub> and PbO), coal combustion, and incineration (PbCl<sub>2</sub>) [14]. Lead and its compounds can be absorbed into the human body by inhalation of dust, aerosol, fume, and vapor, with the degree of absorption dependent on particle size and solubility. Once absorbed, lead binds strongly to red blood cells and is then deposited in bone,



Delhi ( $76^{\circ}50'E-77^{\circ}23'E$ ,  $28^{\circ}12'N-28^{\circ}53'N$ )

FIGURE 1: Location of sampling site in Central Delhi.

accumulating there. Relatively high levels of Pb have been observed in the blood of people living in industrial areas [15].

Over the past decade, a large number of studies have established a link between airborne particles and human mortality [16]. The chemical composition of particulates, especially the presence of toxic chemicals or metals, is a more appropriate parameter for the assessment of particulate health effects rather than their mass concentrations [17]. Several studies on Pb in aerosols [8, 18, 19] have been reported. However, few studies have been focused on ambient levels of Pb during day and night in different seasons and their long range transport. The previous studies of Pb were generally focused on different study sites, namely, traffic, industrial, and commercial areas with 24-hour mean concentration, while the understanding of long-term study of Pb and its day and night time variation in typical residential areas of Delhi is found to be fairly low. The major sources of Pb pollution in Delhi include industrial units (metal processing, chemical processing, paint, etc.), coal-fired power plants, waste incineration (municipal solid waste, medical waste, wood, etc.), and resuspended dust by anthropogenic and vehicular activities [12, 20]. As such, land-use patterns in Delhi are overlapping and essentially mixed, which means that residential areas are likely to be impacted by a host of different sources that needs to be delineated. Therefore, this study aims to present a holistic view of day and night time variation of Pb in  $PM_{10}$  at a site in Central Delhi.

As Pb is considered as one of the criteria air pollutants, having severe health implication after long term exposure, this study is important to assess the mean levels of Pb during different seasons and to identify the regional source areas contributing to Pb in atmospheric  $PM_{10}$  aerosols.

## 2. Methodology

**2.1. Sampling Site.** The samples were collected at the terrace of main building of National Physical Laboratory, New Delhi. Delhi is among the highly polluted cities in the world and lies in north-central India ( $76^{\circ}50'E-77^{\circ}23'E$ ,  $28^{\circ}12'N-28^{\circ}53'N$ ) about 216 m above mean sea level and 1400 km away from the sea (Figure 1). Delhi has a population of around 16.7 million people as per 2011 census. Intense industrial activities, large population, and rise in motor vehicle usage are posing

TABLE 1: Optimized instrumental conditions used for the determination of lead.

Parameters	Pb
Lamp current (mA)	10.0
Wavelength (nm)	283.31
Slit width (mm)	2.7/1.05
Burner type	Air/acetylene
Fuel ( $C_2H_2$ ), lit/min	2.5
Oxidant (air), lit/min	10.0

adverse environmental impact in the region [3, 4]. The length of roadways has covered 31 thousand kms in the city. The registered vehicular strength of the city has grown up to 7.4 million till 2011. The sampling site is covered by dense forest of an area 7800 hectares (known as “green lung” of the city) in south, IARI farm field in west, and major commercial and residential areas in north and east directions, respectively. This shows that sampling site represents the ambient quality of air, with no direct source of emission.

**2.2. Sample Collection.** The samples ( $n = 106$ ) were collected at the roof-top of National Physical Laboratory approximately 15 m above the ground level. A  $PM_{10}$  sampler (Envirotech APM 460 BL) was used for the collection of 12 hourly samples from 7 am to 7 pm (day time) and 7 pm to 7 am next day (night time) in every month throughout the year 2011. Each day 2 samples were collected with average of 8–10 samples every month. The samples were collected on Whatman Quartz fiber filters (rectangular  $20.3\text{ cm} \times 25.4\text{ cm}$ ) at a flow rate of  $0.9$  to  $1.4\text{ m}^3/\text{min}$ . These samples were refrigerated till their further analysis.

**2.3. Analysis.** After completion of sampling, two circular portions of the filter (diameter 40 mm each) were cut using a stainless steel punch. These portions of the filter were extracted in 40 mL of 5% nitric acid (Merck) by keeping them in ultrasonic bath for a period of one hour [37]. The field blanks and reagent blanks were also extracted and analyzed in similar manner. Recovery experiments were conducted with recovery efficiency for Pb above 97%. The extracted samples were analyzed by using Atomic Absorption Spectrometer (Perkin Elmer AAnalyst 400). The calibration was done by using certified reference standard solutions (Merck, Germany) covering concentration range of lead. A three-point calibration curve was linear with  $R^2 > 0.99$  and was used for quantification. The minimum detection limit for Pb was  $0.018\text{ mg/L}$ . To ensure analytical precision, a standard of known concentration was analyzed in triplets to check %RSD, which was within 2% for the measurement of lead. Optimized instrumental parameters for analysis of lead have been given in Table 1.

## 3. Results and Discussion

**3.1. Day and Night Time Variation of Lead.** High mean concentrations of Pb have been found in night time as compared

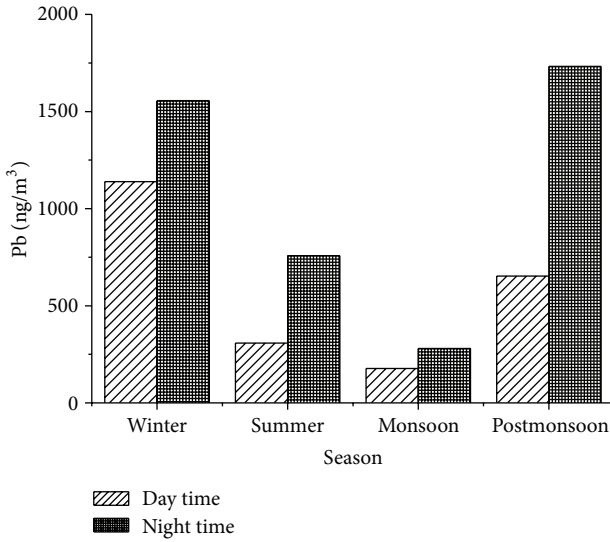


FIGURE 2: Day and night time variation of Pb in PM<sub>10</sub> during different seasons.

to day time in all seasons of the year 2011. The high concentration of Pb during night time (93 ng/m<sup>3</sup>) as compared to day time (71 ng/m<sup>3</sup>) was also reported in Milan, Northern Italy [38]. Although, Pb is mainly contributed by biomass burning and industrial sources, higher concentration observed during night time may also be attributed to stable atmosphere and shifting of boundary layer. The mean level of Pb during day and night time in winter and summer seasons has been 1139 and 1556, and 307 and 757 ng/m<sup>3</sup>, respectively. The postmonsoon period shows significant variation of Pb during day and night time whereas monsoon shows least variation among all seasons (Figure 2). The average day time and night time concentrations for the postmonsoon season have been found to be 652 and 1731 ng/m<sup>3</sup> while for monsoon season they were 177 and 280 ng/m<sup>3</sup>. The standard value recommended by NAAQS-CPCB, New Delhi, for Pb is 1000 ng/m<sup>3</sup> and 500 ng/m<sup>3</sup> for 24 hrs and annual mean, respectively. Only in winter season, the concentration has been found above 1000 ng/m<sup>3</sup> during day and night time. During postmonsoon period also some samples showed Pb concentration above 1000 ng/m<sup>3</sup> during night time. The annual mean concentration of Pb is increased up to 1051 ng/m<sup>3</sup> during night time and 625 ng/m<sup>3</sup> during day time.

**3.2. Seasonal Variation of Lead.** This study shows significant seasonal variation of Pb in PM<sub>10</sub> aerosols with maximum concentration in winter and minimum concentration in monsoon (Figure 3(a)). The Pb levels during different seasons have been found in the decreasing order as winter > postmonsoon > summer > monsoon. The ambient levels of Pb during winter season have been found to be 1335 ng/m<sup>3</sup>. Around 31.3% samples in winter have exceeded the NAAQS, that is, 1000 ng/m<sup>3</sup> (for 24 hrs mean). The high levels observed during winter may be due to atmospheric stability and lower inversion layer, which are very

frequent in this season. The more fossil fuel and biomass burning during this period may also contribute to higher ambient concentration of Pb during this season [39, 40]. The source strength and meteorological conditions highly influence the accumulation of Pb in the atmosphere. The shifting of atmospheric boundary layer towards the surface of earth reduces dispersion of Pb particles, resulting in its elevated concentration (Figure 3(b) plotted using data from <http://ready.arl.noaa.gov/READYamet.php>). In addition, prevailing calm wind conditions also facilitated the existence of Pb in higher concentration [41]. The clear and sunny days during summer season resulted in unstable atmospheric conditions, which maximized dispersion of generated pollutants via vertical mixing in the atmosphere (Figures 3(c) and 3(d)). As a result, the ambient levels of Pb have been found to be significantly low in summer (532 ng/m<sup>3</sup>) as compared to those of the winter season. Sharp rise of Pb in postmonsoon season (1124 ng/m<sup>3</sup>), just after monsoon (235 ng/m<sup>3</sup>), indicates its possible contribution by burning of crop residues in Punjab and biomass burning near IARI farm land, which is in the vicinity of sampling site. A comparative seasonal variation shows that winter season has 1.2, 2.5, and 5.7 times higher Pb concentrations than postmonsoon, summer, and monsoon seasons, respectively [42]. It has been observed that the summer season has 2.3 times higher Pb concentration than the monsoon season.

**3.3. Concentration Weighted Trajectory (CWT) for Lead.** The receptor based method is widely used for long term measurements of pollutants. For unknown sources of pollutants, hybrid models that incorporate wind trajectories can be used to resolve sources locations. CWT is a type of receptor model which has been used in this study to locate regional source areas potentially affecting the receptor site. CWT analysis is useful to identify pollution sources especially for long-term environment measurements. The sources either local or regional contributing to lead (Pb) in PM<sub>10</sub> at an urban site in Central Delhi were investigated using CWT [43]. TrajStat: GIS-based software has been used for CWT ([http://www.meteoinker.com/Documents/Wang\\_TrajStat\\_Manuscript.pdf](http://www.meteoinker.com/Documents/Wang_TrajStat_Manuscript.pdf)).

In the CWT method, each grid cell is assigned a weighted concentration by averaging the sample concentrations that have associated trajectories crossing that grid cell, as follows:

$$C_{ij} = \frac{1}{\sum_{l=1}^M \tau_{ijl}} \sum_{l=1}^M C_l \tau_{ijl}, \quad (1)$$

where  $C_{ij}$  is the average weighted concentration of Pb in the grid cell ( $i, j$ ),  $l$  is the index of the trajectory,  $M$  is the total number of trajectories,  $C_l$  is the concentration observed at sampling location (receptor site) on arrival of trajectory  $l$ , and  $\tau_{ijl}$  is the residence time (time spent) of trajectory  $l$  in the grid cell ( $i, j$ ). Thus, the weighted concentration obtained at each grid cell represented that this concentration can be expected at the receptor site. A high value for  $C_{ij}$  implies that air parcels traveling over the grid cell ( $i, j$ ) would be, on average, associated with high concentrations at the receptor site.

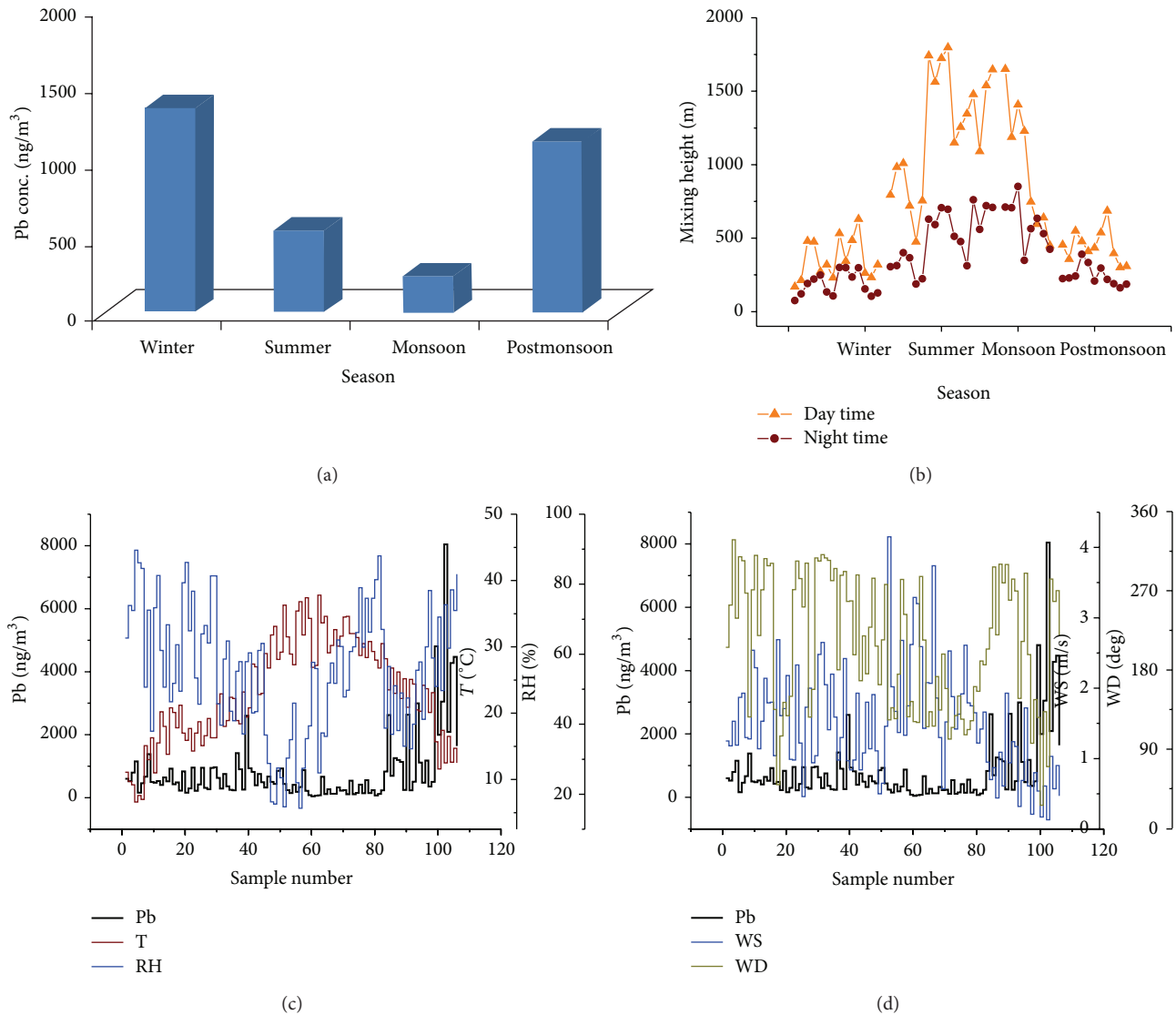


FIGURE 3: (a) Seasonal variation of Pb during different seasons. (b) Average mixing height during different seasons. (c) Variation of Pb with temperature and relative humidity. (d) Variation of Pb with wind speed and wind direction.

Back air trajectory calculations were done by using HYSPLIT model (<http://www.arl.noaa.gov/ready/hysplit4.html>). To observe the role of long range transport, 5-day isentropic air mass back trajectory arriving at receptor site was plotted at 500 m agl (above ground level). The Pb concentrations were estimated at 12-hour interval (day and night basis). Therefore, two back trajectories for each sampling day were generated, at 02 ut and 14 ut (universal time) for day and night time, respectively. The Pb levels at receptor site were added to the corresponding trajectories, so the trajectories could be selected according to the measured data.

This method helped to determine the relative significance of potential sources. Based on the analysis of the whole trajectory data set, the most frequent wind directions were west, north-west, and south-east, thus suggesting that the sampling site might be under influence of several source regions.

Figure 4 shows the concentration weighted trajectory during winter, summer, and monsoon and postmonsoon. The air masses, reaching the sampling site coming from different areas throughout the year 2011, significantly contribute to the quality of air at the sampling site. The main areas from which the air masses were coming to the sampling site included Northern Gangetic plain, Thar Desert, industrial areas of U P, Arabian Sea, and Bay of Bengal. The CWT showed that the air masses were approaching the sampling site coming from the hot sandy region of northern India and Thar Desert (India and Pakistan) during summer. The low levels of Pb during this period may be attributed to winds arriving from the regions having lesser extent of anthropogenic and industrial activities [41]. During monsoon, the air reached the site coming from Arabian Sea via Northern Gangetic plain of India, whereas, in winter, it covered the major industrial area of north east



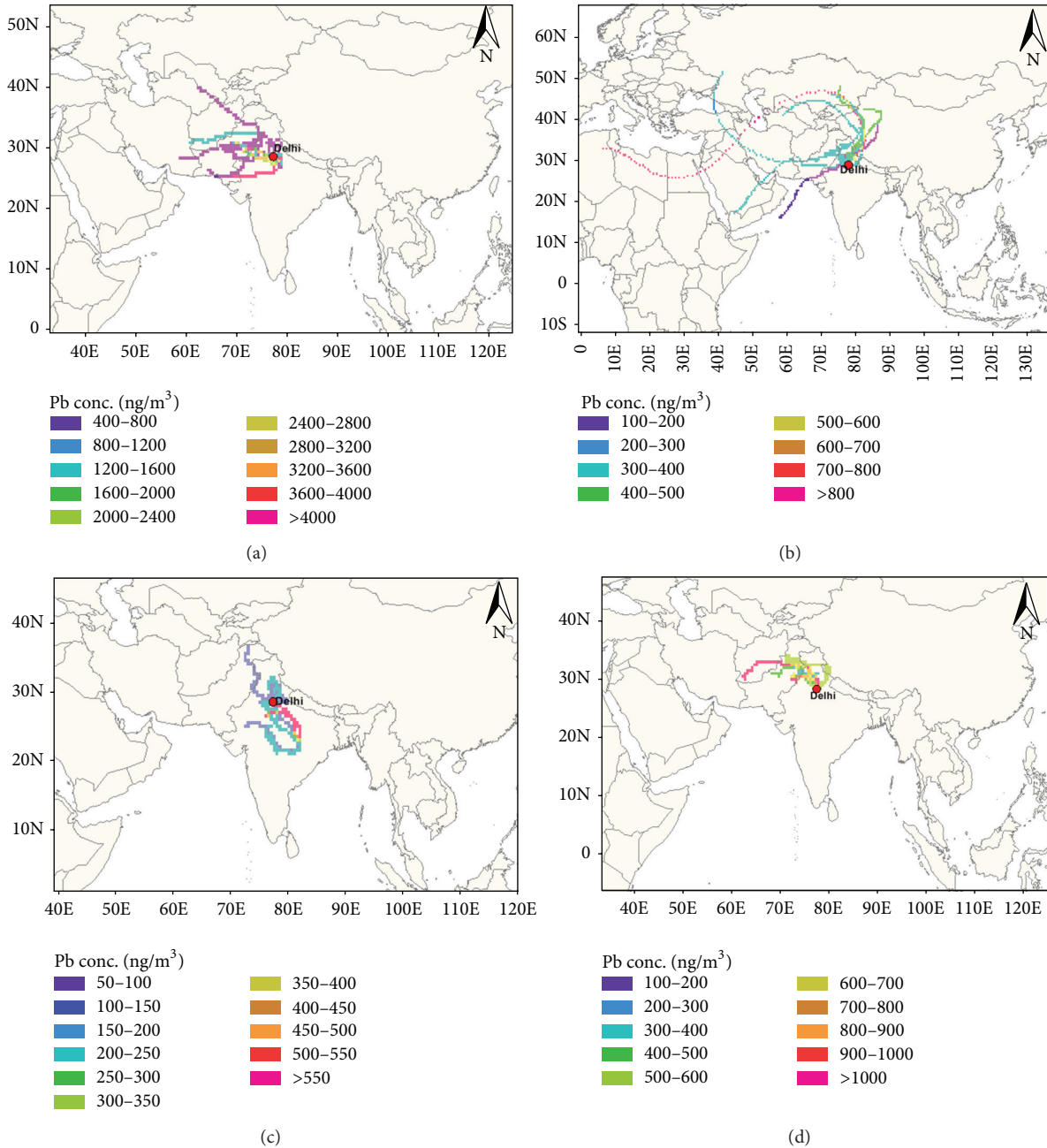


FIGURE 4: (a) Concentration weighted trajectory of Pb during winter season. (b) Concentration weighted trajectory of Pb during summer season. (c) Concentration weighted trajectory of Pb during monsoon season. (d) Concentration weighted trajectory of Pb during postmonsoon season.

Pakistan and north west India. The trajectory showed that the air masses arriving at the site came from the hot sandy areas of Pakistan via industrial areas in India like, Ludhiana, Firozpur, and Kapurthala. Air coming from industrial area might contribute to enhanced levels of lead. The air masses reached the site via highly populated areas having high density of roadways and vehicular emissions which might have contributed to higher concentration of lead. The postmonsoon season has been identified with highly confined potential source regions located in NW direction to the sampling site.

These regions act as a major contributor of Pb at the receptor site.

3.4. Comparison with Other Studies. Table 2 gives a comparison of Pb reported by various workers at different sites studied worldwide [8, 19, 21–36]. But very few studies have reported ambient levels of Pb on day and night time basis in  $\text{PM}_{10}$ . This study reveals that the mean concentration of Pb in night time is higher than the day time in all seasons of the year 2011. The annual mean concentration of Pb ( $835 \text{ ng/m}^3$ ) is substantially

TABLE 2: Comparison of ambient levels of lead with other studies worldwide.

Site	Characteristics	PM type	(ng/m <sup>3</sup> )	Reference
India				
Central Delhi, India	Urban	PM <sub>10</sub>	835	Present study
Delhi, India	Urban (residential)	PM <sub>10</sub>	270–460	[8]
Agra, India	Urban	PM <sub>10</sub>	1100	[21]
Kolkata, India	Urban	SPM	159	[19]
Mumbai, India	Urban	SPM	101	[22]
Coimbatore, India	Rural	SPM	210–620	[23]
Delhi, India	Urban	SPM	440	[24]
Asia				
Lahore, Pakistan	Urban	PM <sub>10</sub>	4400	[25]
Karachi, Pakistan	Urban	SPM	4000	[26]
Beijing, China	Rural	PM <sub>10</sub>	330	[27]
Lanzhou, China	Urban	PM <sub>2.5</sub>	635	[28]
Tehran, Iran	Urban	SPM	1020	[29]
Seoul, Korea	Urban	PM <sub>10</sub>	200	[30]
Europe				
Istanbul, Turkey	Urban	PM <sub>10</sub>	70	[31]
Lecce, Italy	Urban (urban background)	PM <sub>10</sub>	8	[32]
Vienna, Austria	Urban	PM <sub>10</sub>	10	[33]
Huelva, Spain	Urban	PM <sub>10</sub>	20	[34]
Edinburgh, UK	Urban (urban background)	PM <sub>10</sub>	10	[35]
USA				
Los Angeles, USA	Urban	PM <sub>10</sub>	2	[36]

SPM: suspended particulate matter, PM<sub>10</sub>: coarse particulate matter, PM<sub>2.5</sub>: fine particulate matter.

high as compared to previously reported Indian cities. Several studies within Indian Territory show that urban sites have higher levels of Pb than rural sites in atmospheric aerosols. Higher concentrations at urban sites may be attributed to increased population density, industries, vehicular emissions, and fossil fuel burning. In addition, coal burning which is common in India for thermal power plants, industries, and household activities also adds lead to the air. It has been noticed that the coastal cities of India have lower Pb levels than inland cities [19, 21, 22]. It might be attributed to less polluted sea breezes and prevailing high speed winds as compared to inland cities.

#### 4. Conclusion

Ambient levels of Pb were monitored for the period of one year on the day and night basis. It has been found that the Pb levels in Central Delhi are substantially high as compared to previously reported urban sites in Delhi. It has also been observed that the mean concentration of Pb is higher, when compared with the other European cities over the world. The mean Pb concentrations of PM<sub>10</sub> samples in Delhi during day and night have been observed as 625 and 1051 ng/m<sup>3</sup>, respectively. A significant seasonal variation of Pb has been observed. The winter season has 5.7 times higher Pb concentration than the monsoon in Central Delhi. The highest Pb level during winter period might be attributed

to prevailing atmospheric stability, calm wind conditions, and shifting of boundary layer. Moreover, biomass burning may also have contributed to the increased levels of Pb during winter. Lowest level of Pb during monsoon season has been found which might be due to frequent washout of the atmosphere. It has been observed that the summer season has 2.3 times higher Pb concentration than the monsoon season. The low level of Pb during summer season might be attributed to the wind arriving at the site from less polluted Himalayan regions. A sharp rise of Pb level was found in postmonsoon season, which may be due to more biomass burning and shifting of boundary layer. It has been found that the concentration of Pb is higher in night time as compared to day time throughout the year, which might be due to shifting of boundary layer in night time. To identify the potential sources of Pb during different seasons, CWT has been plotted for every season. The analysis of CWT shows that locations of potential sources are different for different seasons. The winter season has major local sources whereas summer has distant sources. It also suggests that the Pb level at the study site has potential role of nonpoints sources, natural emissions, and biomass burning.

#### Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

## Acknowledgments

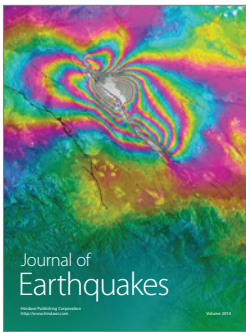
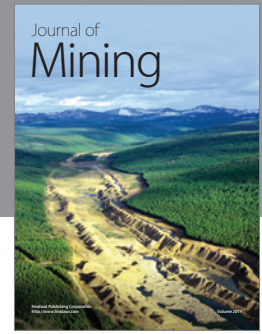
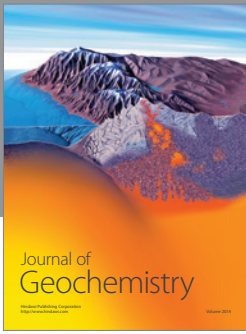
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