



## Seasonal Variations of Atmospheric Particulate Matter and its Content of Heavy Metals in Klang Valley, Malaysia

Rasheida E. Elhadi<sup>1</sup>, Ahmad Makmom Abdullah<sup>1\*</sup>, Abdul Halim Abdullah<sup>2</sup>,  
Zulfa Hanan Ash'aari<sup>1</sup>, Md Firoz Khan<sup>3</sup>

<sup>1</sup> *Environmental pollution Control Technology, Department of Environmental Sciences, Faculty of Environmental Studies, University Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia*

<sup>2</sup> *Department of Chemistry, Faculty of Sciences, University Putra Malaysia, 43400 UPM Serdang, Malaysia*

<sup>3</sup> *Centre for Tropical Climate Change System, Institute of Climate Change, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia*

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### ABSTRACT

The composition of atmospheric particulate matter (PM<sub>10</sub>) can help to identify its potential sources and possible impact on human health. This study investigates the seasonal variations and sources of heavy metals in PM<sub>10</sub> in the Cheras area. PM<sub>10</sub> samples were collected on a 24-h basis using a high-volume air sampler in June during the southwest monsoon and in December during the northeast monsoon of 2014. Selected hazardous trace metals, viz., As, Pb, Cu, Ni, Cd, Co, Mn, Zn, Fe, Cr, V and Ba, were measured by inductively coupled plasma mass spectrometry (ICP-MS). The result showed that the mean concentrations of PM<sub>10</sub> were 207.63 ± 7.82 and 138.32 ± 4.67 during the southwest and northeast monsoons, respectively. The heavy metal concentrations during the southwest monsoon followed the order Ba > Fe > Cu > V > Zn > Pb > Mn > Cr > As > Ni > Cd > Co, while those during the northeast monsoon followed the order Zn > Fe > Ba > Cu > Pb > V > Cr > As > Mn > Ni > Cd > Co. The results of the enrichment factors (EFs) showed that the major trace metals mainly originated from anthropogenic sources. A correlation analysis indicated that pairs of trace metals from similar sources were suspended in PM<sub>10</sub> in the ambient air. The source apportionment by principal component analysis (PCA) and cluster analysis (CA) suggested that vehicle exhaust and brake and tire wear, industrial emissions and the re-suspension of dust, as well as oil combustion, were the most dominant sources of PM<sub>10</sub> in this study.

**Keywords:** Particulate matter; PM<sub>10</sub>; Heavy metals; Seasonal change; Enrichment factors; Meteorological factors; Multivariate modeling.

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### INTRODUCTION

Particulate matter (PM) in the atmosphere is among the most important pollutants affecting the ambient air pollution level. Moreover, it affects severely the human respiratory system (Das *et al.*, 2015). Recent studies on the exposure to particulate matter (PM) with an aerodynamic diameter of 10 µm or less (PM<sub>10</sub>) have identified a variety of health-related problems, which include deterioration in lung function, chronic pulmonary disease, heart disease and premature death along with a rise in mortality (Capasso *et al.*, 2015; Xie *et al.*, 2015; Zúñiga *et al.*, 2016).

Heavy metal compositions of the respirable size fraction of particulate matter (PM) are increasingly being monitored,

due to the fact that several metals have been connected to adverse human health outcomes (Freitas *et al.*, 2010; Garcia *et al.*, 2011). Several epidemiological studies revealed that heavy metals in the airborne particles constitute serious problems because of their threat to human health (Andersen *et al.*, 2007; Liu *et al.*, 2009; Mavroidis and Chaloulakou, 2010).

Recent times have witnessed an unbelievable increase in heavy metal concentrations in the atmospheric aerosols, which vary considerably, mainly as a result of extensive anthropogenic activities, wild forest fires and secondary sources (Khan *et al.*, 2016b; Shah *et al.*, 2006; Wahid *et al.*, 2014). To strategically address this issue and enhance the quality of ambient air, it is crucial to study and determine the extent to which various emission sources contribute to the ambient particulate composition. Both natural and anthropogenic sources are responsible for releasing heavy metals into the atmosphere. Among the natural emissions, it has been discovered that re-suspended surface dust is a major contribution. On the other hand, the main sources of

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\*Corresponding author.

Tel.: +601126250360; Fax: 603-89438109  
E-mail address: amakmom@upm.edu.my

heavy metals originating from anthropogenic sources include fossil fuel combustion emission from automobiles and industrial activities, which are responsible for the bulk of toxic metals in urban areas (Zereini *et al.*, 2005; Hao *et al.*, 2007). Because of this, there has been close attention paid to the potential effects of the metallic constituents of the atmospheric aerosols on human health and on different sections of the environment (Shah and Shaheen, 2010). It has been observed that the manner in which heavy metals in the airborne particulates are distributed exhibit considerable changes in relation to the meteorological conditions worldwide (Khan *et al.*, 2010; Shah and Shaheen, 2010; Yusup and Alkarkhi, 2011).

Assessing the extent of the contributions from various sources to the atmospheric particulate has always been a big challenge to environmental scientists. However, it has been reported in literature that several ways were tried to cope with the problem in the past (Qin and Oduyemi, 2003). Nevertheless, the most frequently used methods apply multivariate statistics; using PCA and CA are considered effective tools for identifying the sources and understanding how heavy metals are distributed in the atmospheric particulate matter (Shah *et al.*, 2012).

From the findings on land use studies and the consensus of the earlier researches, indications are that there is a serious air quality problem in Peninsular Malaysia, usually in the highly urbanized areas. This is especially the case in terms of the suspended particulate matter and lead (Pb) in the ambient air in the vicinity of crowded roadsides. The major causes of such problems were motor vehicles, industrial emissions and open burning (Afroz *et al.*, 2003; Azmi *et al.*, 2010). It should be noted that earlier short-term studies were conducted in this area (Sulaiman *et al.*, 2005; Tahir *et al.*, 2009; Ismail *et al.*, 2011; Yusup and Alkarkhi, 2011), revealing an increase of the toxic metals over a specific period of time. The above studies mainly discussed the sources of the pollutants and alteration of the chemical compositions in the particle phase over a certain period. However, a systematic sampling of PM<sub>10</sub> in an urban area during the southwest and northeast monsoon can improve the understanding of the factors involved in the changes in the PM<sub>10</sub> level and its chemical composition.

The main objective of this study to measure the PM<sub>10</sub> concentration in two specific seasons, namely the southwest and northeast monsoon, for a certain period in an urban area in Cheras. This current study will make a seasonal change of the pollutants and source apportionment using enrichment factors, cluster analysis and principal component analysis.

## MATERIALS AND METHODS

### *Description of the Sampling Location*

Cheras is a suburb of Kuala Lumpur, the Malaysian capital city. The township is located to the southeast of Kuala Lumpur (3.0422°N, 101.7706°E). It is adjacent to Ampang and Kajang, which are two major towns within the metropolitan area. Cheras is one of the major towns in the Klang Valley region, which is highly populated, with

heavy transportation activities. The town is characterized by high temperatures in March, and the highest precipitation in November, corresponding with the northeast monsoon, which lasts until March. For this study, the sampling site is representative of the Cheras highway; hence, the location of sampling points was proximate to the roadsides (see Fig. 1).

### *Sampling Collection*

The collection of suspended atmospheric particulate matter (PM<sub>10</sub>) was conducted using a high volume air sampler (Anderson, model B/MV2000HX, USA) in an urban area of Cheras, Malaysia. The sampling was performed on a regular 24-h cycle in June during the southwest monsoon and in December during the northeast monsoon in 2014. A total of 16 PM<sub>10</sub> samples were collected on glass fiber filters (20.4 cm × 25.4 cm) at a flow rate of 1.13 m<sup>3</sup>min<sup>-1</sup> (IO-2.1, 1999) for each season.

### *Analyses of the Samples*

PM<sub>10</sub> mass was defined gravimetrically before the analysis (IO-2.1, 1999). Selected heavy metals were then evaluated using a mixture of nitric acid (MerkSuprapure 65%) and hydrochloric acid (MerkSuprapure 37%) for extraction of the metals from the filters (IO-3.1, 1999). The extracted solution was filtered by washing with double-distilled water and refrigerated in a pre-cleaned strong polyethylene bottle for later analysis (Beceiro-González *et al.*, 1997; Shah and Shaheen, 2010). This was followed by the treatment of the filter and reagent blanks. Selected heavy metals were identified by Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Perkin Elmer Elan 9000) based on IO-3.2, 1999. The calibration of the ICP-MS was performed with standard multi-element solutions.

### *Validation of the Analytical Procedures*

As part of quality assurance (QA) and quality control (QC), the levels of the PM<sub>10</sub> composition were corrected from the reagent and filter blank samples, which were similarly treated with the procedure applied to the exposed filters. The recovery (%) of the heavy metals used Standard Reference Material (NIST SRM1648a for Urban Particulate Matter) acquired from the National Institute of Standards and Technology (NIST), USA. Results exhibiting the recoveries of all the metals measured were in the range of 79%–95%. The detection method limits were 0.3 for As, 0.12 ng m<sup>-3</sup> for Pb, 0.3 ng m<sup>-3</sup> for Cu, 0.13 ng m<sup>-3</sup> for Ni, 0.02 ng m<sup>-3</sup> for Cd, 0.05 ng m<sup>-3</sup> for Co, 0.93 ng m<sup>-3</sup> for Mn, 0.10 ng m<sup>-3</sup> for Zn, 0.4 ng m<sup>-3</sup> for Fe, 0.03 ng m<sup>-3</sup> for Cr, 0.02 ng m<sup>-3</sup> for V and 0.03 ng m<sup>-3</sup> for Ba.

### *Source Apportionment Procedures*

#### *Enrichment Factors (EF)*

The EF can be used to determine the difference between heavy metals caused by human activities and those originating from natural processes to establish the degree of anthropogenic influence. EFs were typically taken as double ratios of the target metal and a reference metal in aerosols and earth crust. The reference element has to be

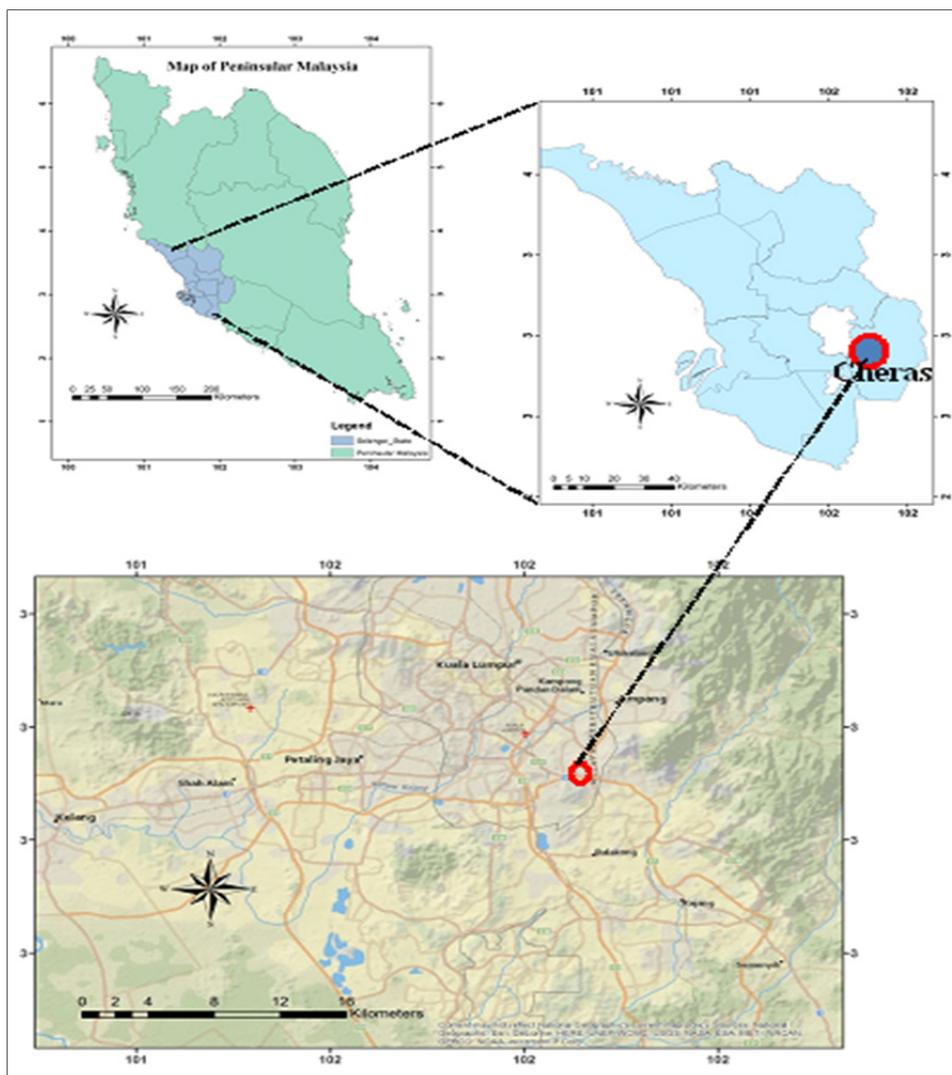


Fig. 1. Map of study area indicating sampling site.

one that is clearly from a single source. Normally, Na, K, Al, Mg, Ca, Mn or Fe is utilized as the reference. In this study, EFs were evaluated with Fe as the reference, employing the following relationship:

$$EF = \frac{[X/Fe]_{\text{aerosol}}}{[X/Fe]_{\text{crust}}} \quad (1)$$

where  $[X/Fe]_{\text{aerosol}}$  and  $[X/Fe]_{\text{crust}}$  refer respectively to the ratios of mean concentrations of the target element and Fe in atmospheric particulate matter and continental crust. The EFs were measured the origin of earth crust mean abundance of the elements provided in the CRC handbook (Lide, 2005).

#### Principal Component Analysis (PCA) and Cluster Analysis (CA)

Multivariate modeling such as PCA and CA were applied to the dataset with the XLSTAT 2015 software. The PCA was performed using varimax normalized rotation on the dataset and the application of the CA to the standardized matrix of samples, following Ward's method,

and the outcomes are presented in a dendrogram. The Kaiser-Meyer-Olkin (KMO) measure is used first, prior to the execution of the PCA, in order to evaluate the appropriateness of the PCA and to determine whether the samples are adequate. Proceeding to the next level should only be done if the KMO value is 0.5 or above (Lawrence *et al.*, 2013; Jamhari *et al.*, 2014). In the current study, the KMO was found to be greater than 0.5. Thus, the dataset for the present study was adequate for PCA analysis.

#### Other Statistical Analysis

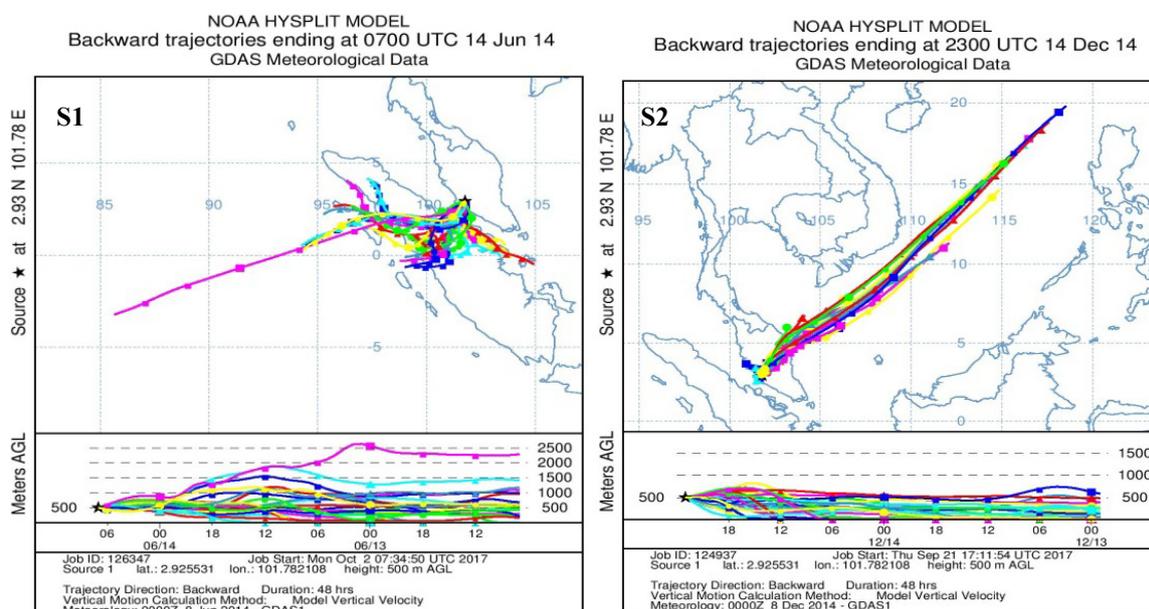
This study summarizes the  $PM_{10}$ , its composition and the meteorological data using XLSTAT2015. The correlation analysis and the related all-visual plotting were made using Excel 2013 and XLSTAT2015.

## RESULTS AND DISCUSSION

The average  $PM_{10}$  levels of the southwest and northeast monsoon seasons are shown in Table 1, and their seasonal spread is shown in Fig. 2 for comparison purposes. Maximum

**Table 1.** Shows the seasonal fluctuations in atmospheric heavy metals levels ( $\text{ng m}^{-3}$ ) for the duration of the study period.

Element	Southwest monsoon (S1) (n= 16)			Northeast monsoon (S2) (16)		
	Mean $\pm$ SD	Max	Min	Mean $\pm$ SD	Max	Min
PM <sub>10</sub>	207.63 $\pm$ 7.82	217.24	196.35	138.32 $\pm$ 4.67	144.54	130.79
As	6.84 $\pm$ 1.57	10.16	5.6	11.37 $\pm$ 2.42	15.07	8.24
Pb	24.26 $\pm$ 5.65	36.63	18.43	21.69 $\pm$ 5.43	31.62	14.79
Cu	103.78 $\pm$ 16.16	125.28	79.18	36.91 $\pm$ 14.59	53.18	17.8
Ni	3.18 $\pm$ 1.15	5.67	2.02	3.50 $\pm$ 1.10	5.12	2.21
Cd	1.75 $\pm$ 0.72	2.83	0.83	0.47 $\pm$ 0.10	0.62	0.34
Co	0.22 $\pm$ 0.07	0.35	0.14	0.26 $\pm$ 0.17	0.48	0.1
Mn	19.84 $\pm$ 4.39	25.83	14.23	10.53 $\pm$ 1.81	12.37	6.87
Zn	54.69 $\pm$ 7.02	70.84	48.9	500.58 $\pm$ 88.01	618.38	349.84
Fe	629.11 $\pm$ 280.74	1087.63	331.21	195.59 $\pm$ 58.25	306.17	141.18
Cr	13.05 $\pm$ 2.52	17.06	9.81	11.38 $\pm$ 3.67	17.58	5.89
V	54.40 $\pm$ 13.77	68.82	34.1	18.57 $\pm$ 4.27	23.07	13.12
Ba	729.29 $\pm$ 44.41	767.3	662.83	111.29 $\pm$ 8.73	124.07	99



**Fig. 2.** Backward trajectories to the selected study area in the Klang Valley during Southwest (S1) and Northeast monsoon (S2).

PM<sub>10</sub> levels were detected for the southwest monsoon, while lower levels were observed during the northeast monsoon (Juneng *et al.*, 2011; Yusup and Alkarkhi, 2011; Khan *et al.*, 2015a). The highest average PM<sub>10</sub> was indicated during the southwest monsoon at 207.63  $\mu\text{g m}^{-3}$ , whereas, the lowest mean PM<sub>10</sub> was indicated during the northeast monsoon at 138.32  $\mu\text{g m}^{-3}$  (Table 1). The mean PM<sub>10</sub> levels discovered in the southwest monsoon were higher than 150  $\mu\text{g m}^{-3}$  for 24 h Recommended Malaysian Air Quality Guidelines (RMAQG), while the levels were higher than WHO standards (50  $\mu\text{g m}^{-3}$ ) during the northeast monsoon (WHO, 2000). The PM<sub>10</sub> concentration during the southwest monsoon (dry season) over the Klang Valley is particularly high due to the contribution of biomass burning from regional sources. Also, high PM<sub>10</sub> concentrations may be due to high automobile emissions where high amounts of particulates are produced and pollute the atmosphere

(Azmi *et al.*, 2010; Juneng *et al.*, 2011). Moreover, the high concentrations of PM<sub>10</sub> during the southwest monsoon may be due to lack of precipitation (2.31 mm) and high temperature (29.18°C) (Juneng *et al.*, 2011; Yusup and Alkarkhi, 2011).

Back trajectory analysis using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model indicated that there were high numbers of biomass fire hotspots during both seasons (Fig. 2). The southwest monsoon season prevailing (from June till September) every year is frequently associated with the generation of haze episodes in Peninsular Malaysia due to biomass burning from Sumatra, Indonesia (Sahani *et al.*, 2014; Khan *et al.*, 2016a). The analysis revealed that the southwest monsoon wind from Sumatra reaches the study area within 48 h. Therefore, the high concentration of PM<sub>10</sub> during the southwest monsoon (June) was considered as an indicator

of the source of aerosol during this season, whereas during the northeast monsoon season (December till March), aerosols were mostly associated with the northeast monsoon winds coming from the South China Sea. This corresponds with the rainy season and therefore makes little contribution to the biomass burning generated from the east (Azmi *et al.*, 2010; Khan *et al.*, 2016a).

Table 1 presents the statistical distribution of heavy metals in PM<sub>10</sub> based on the average concentration. The highest metal mean concentration in the PM<sub>10</sub> during the southwest monsoon season was recorded for Ba at 729.29 ± 44.41 ng m<sup>-3</sup> with maximum and minimum values of 767.3 and 662.8 ng m<sup>-3</sup>, respectively, followed by Fe > Cu > Zn > V > Pb > Mn > Cr > As > Ni > Cd > Co ng m<sup>-3</sup>. However, during the northeast monsoon, Zn showed the highest average levels (500.6 ± 88.01 ng m<sup>-3</sup>), followed by Fe > Ba > Cu > Pb > V > Cr > As > Mn > Ni > Cd > Co ng m<sup>-3</sup>. Commonly, vehicle exhaust and non-exhaust emissions along the road were considered to be the probable contributors to the higher levels of heavy metals in PM<sub>10</sub> (Mansha *et al.*, 2012; Wahid *et al.*, 2014). Ba, remarkably, originated from vehicle exhaust (combustion of gasoline and diesel) and brake wear (Pant and Harrison, 2013; Khan *et al.*, 2016a).

Furthermore, the results showed the concentrations of most heavy metals were higher during the southwest monsoon than the northeast monsoon, except for Zn, As, Ni and Co. These results are consistent with the results obtained by Pengchai *et al.* (2009) and Yusup and Alkarkhi (2011). They reported that Zn correlated positively with relative humidity. However, Zn, As, Ni and Co have not shown any specific pattern with changes in seasonal or meteorological conditions (Yusup and Alkarkhi, 2011). Also, the effect of highway roads characterised by the movement of heavy-duty vehicles has been identified as one of the major contributing factors to the high airborne concentration of heavy metals polluting the atmosphere in Malaysia (Ismail *et al.*, 2011; Wahid *et al.*, 2014).

The average metal concentrations in the particulate matter were seen to be within the limits proposed by international agencies. WHO guideline values and USEPA standards for atmospheric Pb, Mn, Cr, Ni, As and Cd are 500, 150, 110, 20, 6.6 and 6 ng m<sup>-3</sup>, and 1500, 500, 100, 20, 6 and 6 ng m<sup>-3</sup>, respectively (ATSDR, 2002; WHO, 2008). In the course of the current study, the average concentrations of Pb, Mn, Cr, Ni and Cd were below the range permitted by WHO guidelines and USEPA standards. However, the mean level of As exceeded the proposed WHO and USEPA standards. This high concentration of As most likely originated from traffic emission (Pérez *et al.*, 2010; Song and Gao, 2011). The results obtained were in line with the findings (Khan *et al.*, 2016a). The concentrations of heavy metals were well below the WHO guidelines, except the As concentration, which was close to the WHO guideline value and USEPA standard.

Table 2 shows the average levels of airborne heavy metals compared to those recorded in various other locations worldwide.

The observed concentrations of As, Cd, Mn, Zn, Fe, V

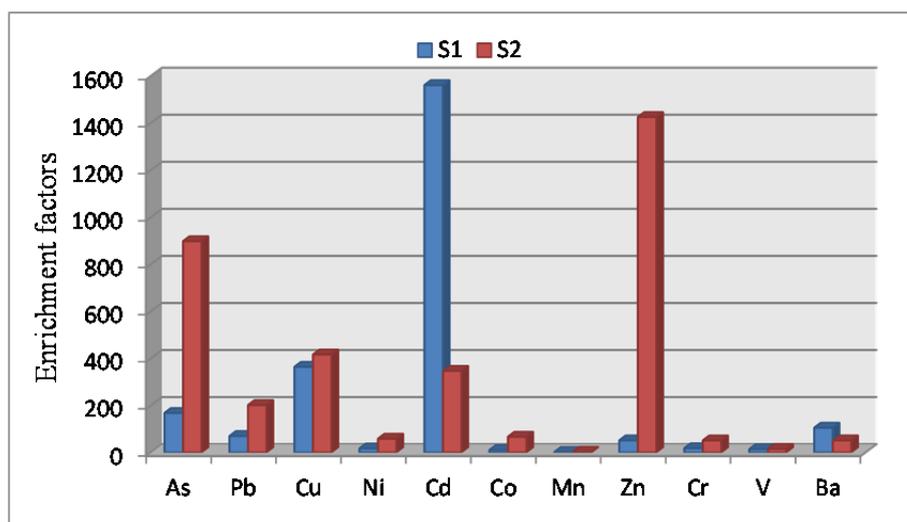
and Ba in the atmospheric particles of the study area were significantly in excess of those recorded for Lodz City, Poland. Conversely, lower to almost similar levels were reported for Pb, Ni, Co and Mn. However, Ni, Fe and Mn are shown to be lower during the northeast monsoon for the study area than for Lodz in the winter season (Bem *et al.*, 2003). In comparison with Erdemli, Turkey (Koçak *et al.*, 2004), lower levels have been recorded for Pb and Fe (northeast monsoon), while the mean levels of Cu, Cd, Mn, Zn and Fe during the southwest monsoon were noticeably higher in the area under study. The present concentrations of Cu, V and Zn (northeast monsoon) are considerably higher compared to the recorded atmospheric particles of Qingdao, China, (Hao *et al.*, 2007). Some of the heavy metals revealed significantly higher levels in the urban particulates of the current study in comparison with those for Izmir, Turkey (Yatkin and Bayram, 2007). The current levels of As, V and Zn (northeast monsoon) were significantly higher compared with those in Al-Hashimya, Jordan (Al-Momani *et al.*, 2005). The observed levels of some heavy metals in this study, such as Pb, Cu, Mn, Zn, Fe and Ba, were higher than those reported for Chiang Mai and Lamphun, Thailand. Higher PM<sub>10</sub>, Cu and Fe but lower Ni, Cd, Mn and Zn concentrations were observed in comparison with those recorded for urban coastal locations in Malaysia (Yusup and Alkarkhi, 2011). The average levels of Pb, Cu, Ni, Cd, Co, Mn, Zn, and Fe in this study were significantly below those for Islamabad, Pakistan (Shah and Shaheen, 2010). The atmospheric concentrations of PM<sub>10</sub>, Pb, Cu, Ni, Cd, Mn, Zn and Fe over Cheras Highway were much in excess of levels determined for semi-urban areas in Malaysia (Wahid *et al.*, 2014). The individual seasonal fluctuations in the levels of the heavy metals show no specific patterns at different locations worldwide due to the considerable differences in the atmosphere, climatic and anthropogenic conditions, particularly an increase in vehicular emissions in locations 100 m away from the sampling site.

EFs make it possible to differentiate an anthropogenic source from one of natural origin and therefore can also assist in determining the extent of the contamination (Han *et al.*, 2006). Then heavy metal enrichment factors have a certain level of unpredictability relative to the natural differences in the composition of the earth's crust. So only metals with enrichment factors greater than 5 can be considered as enriched in atmospheric particulate matter and related to sources besides the local soil (Hien *et al.*, 2001). Normally, enrichment factors close to unity indicate a crustal origin, whereas those higher than 10 are viewed as having a primarily non-crustal source.

The EF values of several metals are presented in Fig. 3, which indicates that the seasonal fluctuations in the enrichment factors of Mn are insignificant and mainly around two, thus suggesting that these metals are mostly of crustal origin. On the other hand, the EFs of As, Pb, Cu, Ni, Cd, Co, Mn, Zn, Cr, V and Ba recorded higher EFs, which indicate that these originated from anthropogenic sources; particularly, automobile emissions contribute more significantly to these metals (Hao *et al.*, 2007; Tahir *et al.*, 2009; Pey *et al.*, 2010). The high enrichment factors indicated

**Table 2.** Comparison the mean levels of PM<sub>10</sub> (µg m<sup>-3</sup>) and heavy metals levels (ng m<sup>-3</sup>) in the atmospheric particulate matter measured during the present with previous studies that reported from different areas around the world.

	Season	PM <sub>10</sub>	As	Pb	Cu	Ni	Cd	Co	Mn	Zn	Fe	Cr	V	Ba	Reference
Cheras, Malaysia	Southwest monsoon	207.6	6.84	24.26	103.78	3.18	1.75	0.22	19.84	54.69	629.1	13.05	54.4	729.3	Present study
	Northeast monsoon	138.3	11.37	21.69	36.91	3.5	0.47	0.26	10.53	500.6	195.6	11.38	18.57	111.3	
Lodz City, Poland	Summer	-	0.2	34.8	-	10.4	-	0.18	7.24	28.3	353	4.32	0.97	-	(Bem et al., 2003)
	Winter	-	1.4	41.2	-	0.59	3.52	0.31	16.7	71.7	677	4.11	1.35	7.47	
Qingdao, China	Summer	-	-	63.9	14.2	6.3	1.3	-	44.4	204	1541	-	10.4	-	(Hao et al., 2007)
	autumn	-	-	166	25	10	1.7	-	72	200	2997	-	4.6	-	
Izmir, Turkey	Summer	73.4	-	115	36.6	14.5	1.5	-	31.8	285	949	26.9	12.8	21	(Yatkin and Bayram, 2007)
	Winter	89.5	-	184	58	18.1	1.6	-	24.8	294	874	25.5	15.5	21.6	
Al-Hashimya, Jordan	Summer	-	3.45	81.8	7.74	11.5	4.71	11.7	52.3	215	1351	14.18	12.62	-	(Al-Momani et al., 2005)
	Winter	-	1.23	97.1	13.5	6.49	2.89	10.3	48.3	209	1896	4.10	13.20	-	(Pengchai et al., 2009)
Chiang Mai and Lamphun, Thailand	Summer	77	-	40	20	10	-	-	30	80	590	10	-	10	
	Winter	26	-	10	10	10	-	-	10	300	110	0	-	0	
Urban coastal location/ Malaysia	Summer	70.7	-	-	20	50	20	-	30	1910	330	-	-	-	(Yusup and Alkarkhi, 2011)
	Winter	58	-	-	30	30	10	-	20	2840	260	-	-	-	
Islamabad, Pakistan	Summer	-	-	68	159	27	3	41	71	1255	2482	16	-	-	(Shah and Shaheen, 2010)
	autumn	-	-	118	174	26	6	38	81	2032	3705	34	-	-	
Semi-urban Area, Malaysia	Summer	-	-	-	-	-	-	-	-	-	-	-	-	-	(Wahid et al., 2014a)
	Winter	-	-	23.1	6.08	0.05	0.39	-	4.43	84.5	172	-	-	-	



**Fig. 3.** Seasonal variations of enrichment factors of different heavy metals during the (S1) Southwest and (S2) Northeast monsoons.

for these metals suggest that their presence in atmospheric particulates was at levels too excessive to be due to expected crustal weathering processes. As such, they were mainly of anthropogenic origin (Yaroshevsky, 2006; Shah and Shaheen, 2010). The bulk of the elements, including Cd and Ba, displayed the maximum enrichment factors during the southwest monsoon, while Zn and As revealed the highest EFs during the northeast monsoon.

The local meteorology related variables are presented in Table 3 and Fig. 4. As shown in Table 3, seasonal changes in meteorological parameters for the duration of the study period are discussed in detail. The relative average temperature during the southwest monsoon was 29.18°C. Average rainfall during this season was 2.31 mm, with a maximum value of 10 mm and minimum value of zero, while the average values for wind speed and relative humidity recorded were 3.69% and 87%, respectively. However, all meteorological parameters were higher during the northeast than the southwest monsoon. Such meteorological conditions are typical of regions in the tropics. All these parameters were shown to be highly variable, as indicated by the wide ranges. It was likely that the conditions during the dry season would have an influence on the general distribution of the particulates (Shah and Shaheen, 2010; Yusup and Alkarkhi, 2011). The prevailing winds with the strongest intensity observed during the southwest monsoon and northeast monsoon were southwesterly and northeasterly (Fig. 4).

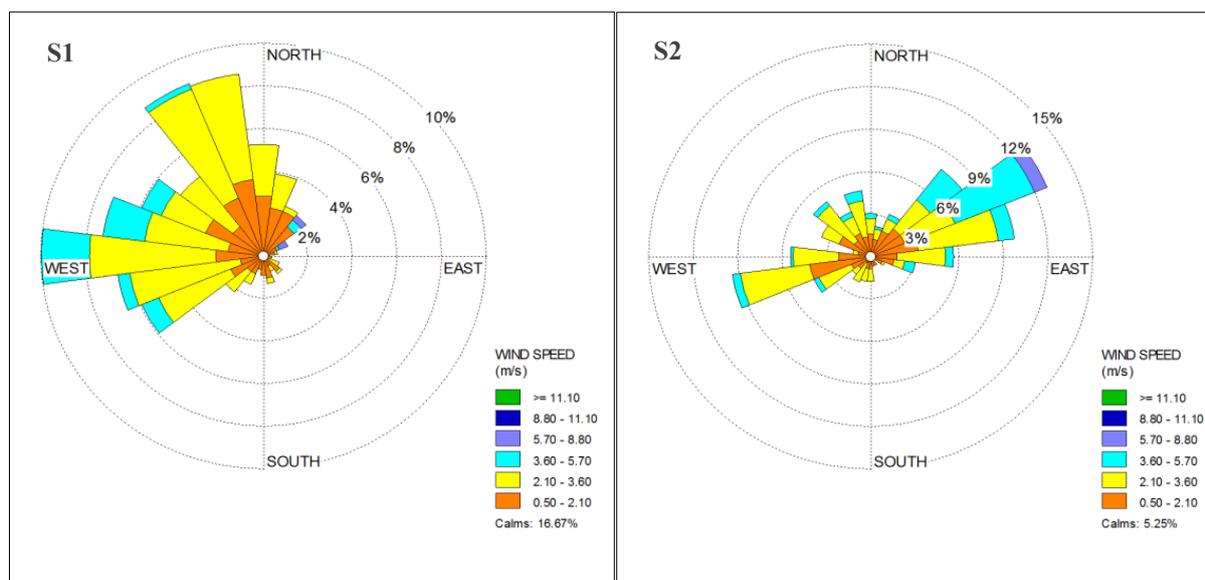
The relationships between matrixes of selected parameters concentration in air were studied to determine how various parameters were related. Table 4 presents the correlation of the results for the parameters selected. In the case of the southwest monsoon season, As exhibited a very significant positive relationship with, Cu, Ni, Cd, Zn and Fe and a moderate correlation with Mn and V, whereas during the northeast monsoon, As showed a strong positive relationship with V and a moderate relationship with Mn. Also, among the metals, Pb revealed significant and strong correlations

during the southwest monsoon with Cu, Ni, Cd, Zn and Fe. A moderate correlation was revealed with Mn and a negative correlation with Ba. These metals are well known for their association with vehicular emissions (Wu *et al.*, 2007; Yu *et al.*, 2013; Khan *et al.*, 2016a). However, during the northeast monsoon, Pb showed a strong positive relationship with Ni, Mn, Zn, Fe and Cr and only a moderate correlation with Cd. These metals were directly associated with vehicular emissions (Khan *et al.*, 2016a). Cu showed moderate correlations during the southwest monsoon with Ni, Cd, Zn and Fe and a moderate negative correlation with Cr. For the northeast monsoon, Cu exhibited a moderate positive relationship with V only. During the southwest monsoon, a number of strong correlations were detected for Ni–Cd, Ni–Zn, Ni–Fe, Cd–Zn, Cd–Fe and Zn–Fe and moderate correlations for Ni–V, Cd–V, Co–Zn, Co–Fe and Mn–Zn. A negative correlation was revealed for Ba–Ni, Ba–Cd, Ba–Mn and Ba–Zn. However, for the northeast monsoon, there was evidence of a strong positive relationship between Ni and all the metals except Co, V and Ba, while Cd showed a moderate correlation with Mn, Zn, Fe and Cr. Also, Co revealed no significant correlations during the northeast monsoon, while Mn showed a moderate correlation with Zn and Cr. Zn was shown to be strongly and positively related to Fe and Ba and had a moderate relationship with Cr, suggesting that they might originate from the same source. However, Fe showed a moderate correlation with Cr and a moderate negative relationship with V.

The correlation coefficient matrix between the selected heavy metals, PM<sub>10</sub> and meteorological parameters is presented in Table 5. During the southwest monsoon, PM<sub>10</sub> showed no significant correlation with the mean temperature, while As, Fe and Ba showed a moderate to weak negative relationship. Furthermore, Pb, Cu and Zn demonstrated a moderate correlation, whereas Cr and Ba showed a negative correlation, with rainfall. Rainfall typically decreases the level of particles as a result of the wash-out processes of

**Table 3.** Seasonal variations in meteorological parameters during the study period.

Parameters	Southwest monsoon (S1)			Northeast monsoon(S2)		
	Mean $\pm$ SD	Max	Min	Mean	Max	Min
Temperature ( $^{\circ}$ C)	28.56 $\pm$ 0.62	30	28	26.49 $\pm$ 1.10	27.8	24.8
Rainfall (mm)	2.31 $\pm$ 3.64	10	0	7.81	37.4	0
Wind speed	3.69 $\pm$ 0.71	5	3	3.11 $\pm$ 1.15	5	1.9
Humidity (%)	87.01 $\pm$ 3.86	93	82	93.59 $\pm$ 3.53	98.7	87.6

**Fig. 4.** Wind rose diagram of study area during the study period (blowing from) during the Southwest (S1) and Northeast monsoons (S2).

the atmospheric particles (Azmi *et al.*, 2010; Khan *et al.*, 2010; Shah and Shaheen, 2010).  $PM_{10}$  and wind speed revealed a high negative relationship, while a moderate weak correlation was found between Ba and Co. A moderate correlation was shown between As, V and humidity, while a weak correlation was observed between Pb, Cu, Ni, Cd and humidity. For the northeast monsoon, Cu and Cr showed a high to moderate positive relationship with temperature, while As and V showed a weak relationship with temperature. Ba showed a moderate positive relationship with rainfall. Conversely, a negative correlation was shown between  $PM_{10}$ , As, Fe, Cr, Pb, Ni, Cd, Zn and rainfall. There was a strong positive correlation between Cd, As, Ni, Mn, Cu and wind speed, but only a weak correlation was shown between V and wind speed. Thus, wind speed had a greater effect on the concentration of particulates in the atmosphere. It was found that humidity and Mn, Cd, Ni, Cu and Pb were negatively correlated. It is also common for high humidity to be related to the frequency of rainfall, and this decreases the presence of particles because of the wash-out processes of the atmospheric aerosols.

PCA and CA were used to identify the role played by the seasonal variations in the identification of sources of selected heavy metals. Table 6 showed the PC loadings of the dataset for the two seasons, and the related eigenvalues and variations, while the CA results are presented in Fig. 5. During the southwest monsoon, three PCs with significant

eigenvalues of more than 1 were obtained, showing 85.89% of the total variance. PC1 accounts for 59.26%, with the highest loading for As, Pb, Ni, Cd, Zn, Fe and Cu and moderate loading for Ba and Mn. These metals could be from vehicle exhaust and non-exhaust emissions (brake wear) (Fabretti *et al.*, 2009; Gietl and Klemm, 2009; Pérez *et al.*, 2010; Pey *et al.*, 2010; Amato *et al.*, 2011; Mansha *et al.*, 2012). PC2 explains 13.84% of the total variability and shows a higher loading for Cr and a moderate one for Co, which could be from non-exhaust (tire wear) and industrial emissions (Zereini *et al.*, 2005; Apeagyei *et al.*, 2011; Song and Gao, 2011). PC3 accounts for 12.79% of the total variability with a 1.53 eigenvalue. It contains strong loadings of V and moderate loadings of Cr. The manifestation of PC3 was believed to be from exhaust emissions (oil combustion), re-suspension dust and tire wear (Pey *et al.*, 2010; Amato *et al.*, 2011; Apeagyei *et al.*, 2011; Khan *et al.*, 2016b).

The identification of sources during the northeast monsoon, which is the wet period, was expected to be considerably different from the dry season. The clustering pattern (Fig. 5) and PC loadings of the selected metals for the northeast monsoon were not much different in comparison with the southwest monsoon. During this season, three PCs were obtained with 84.46% of the cumulative variance. The PC1 with 46.1% of the total variance exhibited higher loadings for Pb, Ni, Cd, Mn, Zn, Fe and Cr, which could have

**Table 4.** Person correlation matrix of particulate matter (PM<sub>10</sub>) and heavy metals levels during the study period.

Variables	PM <sub>10</sub>	As	Pb	Cu	Ni	Cd	Co	Mn	Zn	Fe	Cr	V	Ba
<b>Southwest monsoon (S1)</b>	<b>1</b>												
As	0.252	<b>1</b>											
Pb	0.255	<b>0.961</b>	<b>1</b>										
Cu	-0.089	<b>0.731</b>	<b>0.754</b>	<b>1</b>									
Ni	0.161	<b>0.925</b>	<b>0.877</b>	<b>0.689</b>	<b>1</b>								
Cd	0.161	<b>0.925</b>	<b>0.877</b>	<b>0.689</b>	<b>1</b>	<b>1</b>							
Co	-0.191	0.293	0.436	0.048	0.163	0.163	<b>1</b>						
Mn	<b>0.587</b>	<b>0.593</b>	<b>0.638</b>	0.378	0.463	0.463	0.358	<b>1</b>					
Zn	0.294	<b>0.827</b>	<b>0.93</b>	<b>0.599</b>	<b>0.797</b>	<b>0.797</b>	<b>0.571</b>	0.492	<b>1</b>				
Fe	<b>0.658</b>	<b>0.778</b>	<b>0.763</b>	<b>0.571</b>	<b>0.791</b>	<b>0.791</b>	<b>0.587</b>	0.492	<b>0.745</b>	<b>1</b>			
Cr	0.471	-0.348	-0.432	-0.683	-0.525	-0.525	0.178	0.072	-0.407	-0.201	<b>1</b>		
V	0.354	<b>0.514</b>	0.275	0.07	<b>0.503</b>	<b>0.503</b>	-0.27	0.15	0.105	0.304	0.235	<b>1</b>	
Ba	-0.172	-0.515	-0.603	-0.375	-0.618	-0.618	-0.198	-0.693	-0.653	-0.421	0.47	0.062	<b>1</b>
<b>Northeast monsoon (S2)</b>	<b>1</b>												
PM <sub>10</sub>	<b>1</b>												
As	0.09	<b>1</b>											
Pb	-0.468	-0.059	<b>1</b>										
Cu	0.364	<b>0.566</b>	0.068	<b>1</b>									
Ni	-0.15	0.337	<b>0.825</b>	0.433	<b>1</b>								
Cd	0.321	0.468	<b>0.546</b>	0.58	<b>0.875</b>	<b>1</b>							
Co	0.05	-0.453	0.257	-0.322	-0.127	-0.126	<b>1</b>						
Mn	<b>0.458</b>	0.377	<b>0.708</b>	0.324	<b>0.835</b>	<b>0.625</b>	-0.465	<b>1</b>					
Zn	-0.213	-0.02	<b>0.825</b>	0.03	<b>0.731</b>	<b>0.61</b>	0.254	<b>0.508</b>	<b>1</b>				
Fe	<b>0.627</b>	-0.328	<b>0.794</b>	0.094	<b>0.712</b>	<b>0.617</b>	0.423	0.355	<b>0.768</b>	<b>1</b>			
Cr	0.481	0.197	<b>0.861</b>	0.326	<b>0.851</b>	<b>0.554</b>	0.088	0.67	<b>0.663</b>	<b>0.655</b>	<b>1</b>		
V	0.175	<b>0.867</b>	-0.424	<b>0.529</b>	-0.117	0.068	-0.337	-0.039	-0.365	-0.64	-0.13	<b>1</b>	
Ba	0.353	0.019	-0.438	0.415	-0.207	-0.097	-0.405	-0.058	-0.738	-0.292	-0.29	0.164	<b>1</b>

Values in bold (Correlation is significant at the 0.05 level (2-tailed)).

**Table 5.** Person correlation matrix of particulate matter PM<sub>10</sub>, heavy metals levels and meteorological parameters during the study period.

Parameters	PM <sub>10</sub>	As	Pb	Cu	Ni	Cd	Co	Mn	Zn	Fe	Cr	V	Ba
<b>Southwest monsoon (S1)</b>													
Temperature (°C)	0.03	<b>-0.43</b>	-0.31	-0.39	-0.32	-0.32	-0.06	0.21	-0.18	<b>-0.40</b>	-0.01	-0.51	<b>-0.52</b>
Rainfall (mm)	0.17	0.27	<b>0.56</b>	<b>0.61</b>	0.33	0.33	0.02	0.35	<b>0.53</b>	0.09	<b>-0.78</b>	<b>-0.49</b>	-0.64
Wind speed (m s <sup>-1</sup> )	<b>-0.75</b>	-0.23	-0.21	-0.01	-0.25	-0.25	<b>0.47</b>	-0.36	-0.15	0.29	0.19	-0.27	<b>0.54</b>
Humidity (%)	0.03	<b>0.64</b>	<b>0.48</b>	<b>0.41</b>	<b>0.47</b>	<b>0.47</b>	0.00	0.16	0.15	0.35	-0.01	<b>0.60</b>	0.00
<b>Northeast monsoon (S2)</b>													
Temperature (°C)	-0.20	<b>0.45</b>	0.29	<b>0.70</b>	0.45	0.29	-0.10	0.35	-0.03	0.12	<b>0.67</b>	<b>0.41</b>	0.32
Rainfall (mm)	<b>-0.46</b>	<b>-0.47</b>	<b>-0.50</b>	-0.02	<b>-0.64</b>	<b>-0.70</b>	-0.21	-0.39	<b>-0.60</b>	<b>-0.43</b>	<b>-0.45</b>	-0.10	<b>0.53</b>
Wind speed (m s <sup>-1</sup> )	0.24	<b>0.75</b>	0.30	<b>0.70</b>	<b>0.72</b>	<b>0.85</b>	<b>-0.47</b>	<b>0.71</b>	0.23	0.18	0.39	<b>0.42</b>	0.22
Humidity (%)	-0.11	-0.39	<b>-0.45</b>	<b>-0.56</b>	<b>-0.56</b>	<b>-0.62</b>	0.17	<b>-0.68</b>	-0.24	-0.24	-0.30	-0.18	-0.23

Values in bold (Correlation is significant at the 0.05 level (2-tailed)).

**Table 6.** Principal component loadings of heavy metals during the study period.

Parameter	Southwest monsoon (S1)			Northeast Monsoon (S2)		
	PC1	PC2	PC3	PC1	PC2	PC3
As	<b>0.953</b>	-0.062	0.219	0.151	<b>0.872</b>	-0.43
Pb	<b>0.969</b>	0.065	-0.012	<b>0.929</b>	-0.222	0.02
Cu	<b>0.757</b>	-0.332	-0.261	0.289	<b>0.752</b>	0.174
Ni	<b>0.947</b>	-0.232	0.136	<b>0.957</b>	0.246	0.092
Cd	<b>0.947</b>	-0.232	0.136	<b>0.792</b>	0.388	0.035
Co	0.369	<b>0.834</b>	-0.126	0.09	<b>-0.704</b>	-0.296
Mn	<b>0.648</b>	0.435	0.068	<b>0.754</b>	0.385	0.191
Zn	<b>0.913</b>	0.238	-0.138	<b>0.876</b>	-0.272	-0.276
Fe	<b>0.836</b>	0.232	0.154	<b>0.823</b>	-0.407	0.232
Cr	-0.481	<b>0.58</b>	<b>0.64</b>	<b>0.874</b>	0.072	-0.015
V	0.32	-0.309	<b>0.89</b>	-0.275	<b>0.812</b>	-0.472
Ba	<b>0.688</b>	-0.093	0.342	-0.403	0.443	<b>0.754</b>
Eigenvalue	7.11	1.66	1.53	5.53	3.33	1.27
Variability (%)	59.26	13.84	12.79	46.11	27.75	10.60
Cumulative variance (%)	59.26	73.10	85.89	46.11	73.87	84.46

Values in bold represent high and moderate factor loading.

emanated from vehicle exhaust and non-exhaust (brake wear) emissions into the local atmosphere (brake wear) (Wählin *et al.*, 2006; Mansha *et al.*, 2012). PC2 accounted for 27.75% of the variance, which contained strong positive loadings of As, V and Cu, most likely from re-suspension dust, oil combustion and industrial emissions (Fabretti *et al.*, 2009; Hieu and Lee, 2010; Amato *et al.*, 2011). A considerable variance of 10.60% was associated with PC3 (Ba), which emanated into the atmosphere from non-exhaust factors (brake wear) (Apeageyi *et al.*, 2011; Harrison *et al.*, 2012). PCA produced six PCs both for the southwest and the northeast monsoons and explained more than 84.46% of the cumulative variance of the data. Vehicle exhaust and non-exhaust (brake wear) emissions were revealed by PC1 in both seasons and PC3 during the northeast monsoon, while the contributions from non-exhaust (tire wear) and industrial emissions were demonstrated through PC2 during the southwest monsoon. Most of the re-suspended dust, oil combustion and industrial emissions were evidenced by PC3 during the southwest monsoon and PC2 during the northeast monsoon.

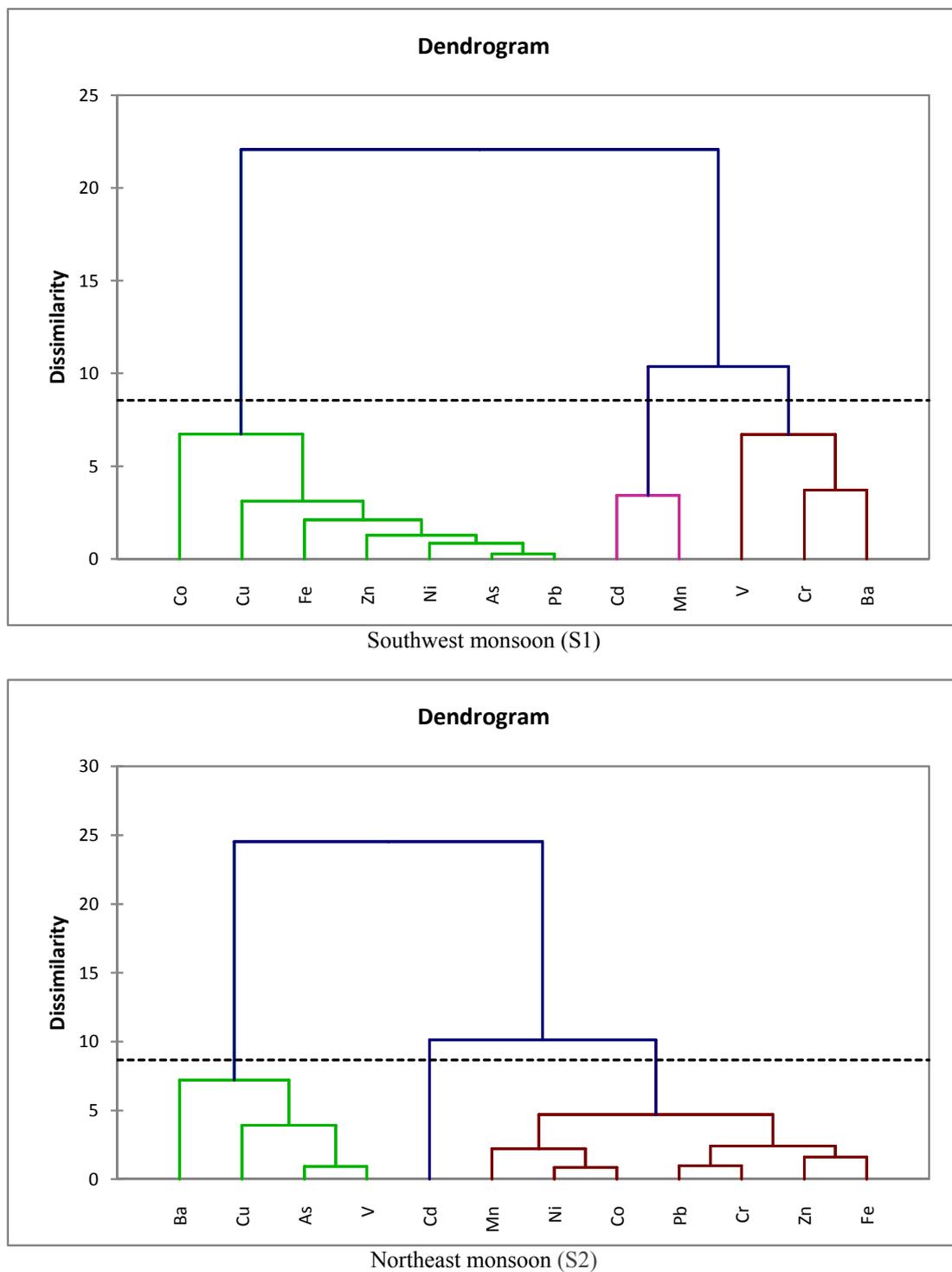
Cluster analysis was used for the source identification of heavy metals in atmospheric particulate matter (PM<sub>10</sub>), and the factors were categorized into three main groups for the two seasons on the basis of similarities among the variables and differences compared to other groups. The first group, CA1, was reported as a dendrogram, as shown in Fig. 5, and consisted of all the major heavy metals, such as Pb, As, Zn, Cd, Ni, Fe and Cu, which could have emanated from vehicle exhaust and non-exhaust emissions into the local atmosphere (Bukowiecki *et al.*, 2010; Amato *et al.*, 2011). The second group, CA2, contained Co and Mn, which most likely came from non-exhaust emission sources and industrial emission (Song and Gao, 2011), while CA3 comprised three metals, namely Ba, Cr and V, which could be associated with vehicle exhaust and non-exhaust emissions (Pey *et al.*, 2010). For the northeast monsoon, the dendrogram of the cluster analysis was also classified into three clusters.

CA1 included Fe, Zn, Cr, Pb, Cd, Ni and Mn, which could have originated from vehicle exhaust and non-exhaust emissions (Mansha *et al.*, 2012; Wahid *et al.*, 2014), while the second cluster (CA2) consisted of V, As, Cu and Ba, which revealed that exhaust and non-exhaust emissions were the main sources of these metals (Fabretti *et al.*, 2009; Pant and Harrison, 2013). The third cluster, CA3, consisted of Co probably coming from vehicle tire wear and industrial emission (Shah and Shaheen, 2010; Song and Gao, 2011). The CA, shown as a dendrogram in Fig. 5, tallied with the PCA in both the seasons. For example, the first CA class (CA1) reproduced the same PCs as PC1 during the southwest monsoon.

Another motivating fact was how PCA and CA illustrated the interrelationship between groupings by PCA and the classification by CA. For instance, CA2 reflected PC1 during the northeast monsoon and had little adjustment regarding the regrouping of the variables.

## CONCLUSION

The statistical analysis revealed that the bulk of the trace metals demonstrated either the same or a contrasting distribution arrangement, possibly due to some common source for the metals in the particulate matter. The concentration of PM<sub>10</sub> was significantly higher during the southwest monsoon compared to the northeast monsoon. Ba and Fe were the predominant metals in PM<sub>10</sub> samples during the southwest monsoon. The concentrations of trace metals during the southwest monsoon were: Ba > Fe > Cu > Zn > V > Pb > Mn > Cr > As > Ni > Cd > Co; those during the northeast monsoon were: Zn > Fe > Ba > Cu > Pb > V > Cr > As > Mn > Ni > Cd > Co. Among the trace metals, As exceeded USEPA and WHO guidelines in both the southwest and the northeast monsoon. The EF results showed that As, Pb, Cu, Ni, Cd, Co, Zn, Fe, Cr, V and Ba apparently originated from anthropogenic sources. Similarly, the observation data related to the local meteorology



**Fig. 5.** Cluster analyses of airborne heavy metals for the two seasons during the study period.

showed considerably dissimilar correlation patterns among the metals during both monsoons. The results of the source apportionment by PCA analysis showed that emissions from vehicles (exhaust, brake and tire wear and re-suspended dust), industry and oil combustion were the main pollution sources in the atmospheric particles around the study area. Due to the high concentrations of PM<sub>10</sub> and trace metals, particularly As, systematic monitoring of PM<sub>10</sub> and the

composition of selected toxic metals is highly encouraged within this study area.

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