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Distribution and Source of Trace Elements in Marine Aerosol of Mersing, Johor, Malaysia

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Abstract

Particulate matter samples with diameter less than 10 μm (PM_{10}) were collected from the coastal area of Mersing, Johor Meteorological Station to investigate external sources of trace elements injected into the marine atmosphere. The concentrations of PM_{10} in the 47 samples for over two years of sampling period between January 2009 and December 2010 were ranged from 8 μg/m^3 to 87 μg/m^3, and a weak negative statistical correction (r=0.139) with daily rainfall were observed except for certain inter-monsoon period. While the concentration of trace elements such as Na, Ba, Al, K and Ca were classified as major metals ranging in concentration from 1 μg/m^3 to 100 μg/m^3. Other elements comprising Mg, Fe, Sr, Cr, Cu, Pb, Mn and Ni were classified as intermediate metals with concentrations ranging from 1 ng/m^3 to 1000 ng/m^3. Also detected were Co and Cd, but these were classified as minor metals with concentration levels of less than 1 ng/m^3. The fluctuate concentration levels of trace elements in PM_{10} also found at study area were directly corresponded to the source of particulate matters, but the concentration levels of particulate matters were influenced by monsoon events. Trace metal compositions in the PM_{10} and the PMF (positive matrix factorization) model have shown a significant presence of terrestrial soil sources especially the by-product of biomass burning (53%) together with terrestrial soil, crustal dust (34.9%), and mixture of anthropogenic sources (12%).

Keywords: Marine aerosol; PM_{10}; Trace metals; Trans-boundary haze; Forest fires; Soil dust

Introduction

The chemical properties of aerosols have been used by many researchers as an indicators of climate abruptions [1-6]. In fact, any chemical changes that occur in the particles during transportation in the atmosphere can also be identified through a chemical composition study [3,4,7-11]. However, the study to characterize and determine the chemical composition of aerosol is a challenging task and to some extent we are yet to develop the tool to do this. Primarily, this is due to the fact that aerosol compositions are highly variable, with a complex mixture of chemical elements. A single aerosol particle may consist of hundreds of different chemical compounds [12,13]. The size, number and chemical composition of the particles may vary due to a number of processes: nucleation (homogeneous and heterogeneous), coagulation, and adsorption/desorption [14]. After their release and evolution, the particles may be removed from the atmosphere by dry and/or wet deposition and heterogeneous chemistry processes as in-cloud scavenging or below-cloud scavenging [15,16]. Therefore, various considerations should be taken into account when identifying aerosol characteristics. This includes identifying whether sources of contaminant release are natural or anthropogenic; determining the direction and rate of air transport through the body; chemical and physical reactions; the mechanism to be used, whether by dry or wet depositions; the effects on living things on earth [17,18] and so on.

One of the most important components of aerosol is mineral dust generated from continental surfaces via long range air mass, transported by wind. In general, continental dust consists mainly of quartz, calcium-rich minerals (calcite, dolomite, gypsum), clay (kaolinite, illite), alkali feldspar, and iron oxides [19]. Therefore, the study of maritime aerosol has its own importance to the biogeochemical state of the underlying ocean. One of the most notable examples is the deposition of iron and other micronutrients from dust that simultaneously affect productivity in the marine ecosystem [20]. Dust deposition is a major factor influencing surface ocean biological productivity in high nutrient, low chlorophyll regions of the oceans [21].

Nevertheless, the haze resulting from biomass burning in Borneo, Sumatra and other parts of Southeast Asia has been occurring with increasing frequency, severity and duration over the last 20 years [22,23,25-27]. Haze episodes have been recorded in 1983, 1990, 1991, 1994, 1997, and 1998, and their causes and effects have been widely reported [28]. The last 1997 and 1998 events have been by far the most severe and they have caused considerable concern among the public and experts alike. Forest fires and the resultant haze have many diverse effects; health impacts on the exposed population being of greatest concern. Aerosols generated by biomass burning consist mainly of carbonaceous compounds (e.g., organic and inorganic carbon) and lower concentrations of various inorganic components [29]. This inorganic fraction is mainly formed by insoluble dust and ashes. The main constituents of the soluble salts are potassium, ammonium, sulphate and nitrate [18]. The $PM_{2.5}$ fraction are also predominantly incorporates metals such as Al, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd, Pb, etc [27]. Meanwhile, coarse mode aerosol contains primarily sea salt and/or mineral dust derived from mechanical erosion of soils and desert sands. Coarse mode mineral dust aerosols are rich in crustal elements like Ca, Si, Al, Fe, while sea-salt aerosols by sea spray mechanism are enriched in Na^+ and Cl^-, with significant amounts of Mg^{2+}, K^+, SO_4^{2-} and Ca^{2+} [9,30].

In recent years, atmospheric researchers have paid special attention to determining the origins and sources of aerosol based on their chemical contents and other natural components as its fingerprints.

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This topic should be stressed further in the global climate change era with emphasize on the identification of the substances that have a significant effect on natural toxicity or radioactivity, and their impacts on marine ecosystems and natural environments. In this study, we aim to investigate external sources of aerosol in the Southeast Asian region and their injection into the marine atmosphere of the southern South China Sea. While the fate of air pollutants has always been a main focus of similar studies, less attention has been given to measuring the amount of the source deposits in Malaysian waters.

Materials and Methods

Sampling site

The study was carried out over a period of two years between January 2009 and December 2010. The aerosol sampling station used, the Mersing Meteorological Station (owned by the Malaysian Meteorological Department) is located on the east coast of Peninsular Malaysia, about 130 km northeast of Johor Bahru and approximately 5 km north of Mersing town, Johor (2°27' N; 103°50' E) (Figure 1). This station is placed on top of the highest ground in the area (approx. elevation is 43.6 m above sea level) and is adjacent to southern South China Sea coastal waters (approx. 300 m eastward). It was selected for its suitability in tracing airborne particulate sources and variations as well as haze monitoring for Malaysian maritime air regions.

Aerosol Sampling

The aerosol samples containing particulate matter with diameters of less than 10 μm (as PM_{10}) were collected from the Mersing Meteorological Station using a high-volume air sampler with a PM_{10} separator at the inlet (Sierra-Andersen Model 1200 PM_{10}) with a flow rate of about 1.13 m^3/min which is equal to 1631.04 m^3/day. The sampling time was set at 24 h to collect a sufficient mass of PM_{10} for chemical analysis. Filter samples were dried in a desiccator for at least 24 h before and after exposure. The weight of PM_{10} samples were determined gravimetrically onto glass fibre (Whatman EPM 2000) filter papers by AND GR-200 microbalance. Filters were wrapped in aluminum foil and stored at 4 °C for further analysis.

Digestion procedure

Collected marine aerosols were measured for Al, Ca, Fe, Mn, K, Ba, Na, Mg, Sr, Co, Pb, Cu, Ni, Cd and Cr using the inductively coupled plasma (ICP) mass spectrometry (MS) after total digestion with a mixture of HNO_3-HCl-H_2O_2-HF in a Teflon beaker as published by Sabuti and Mohamed [5]. All reagents used in this research were of analytical grade.

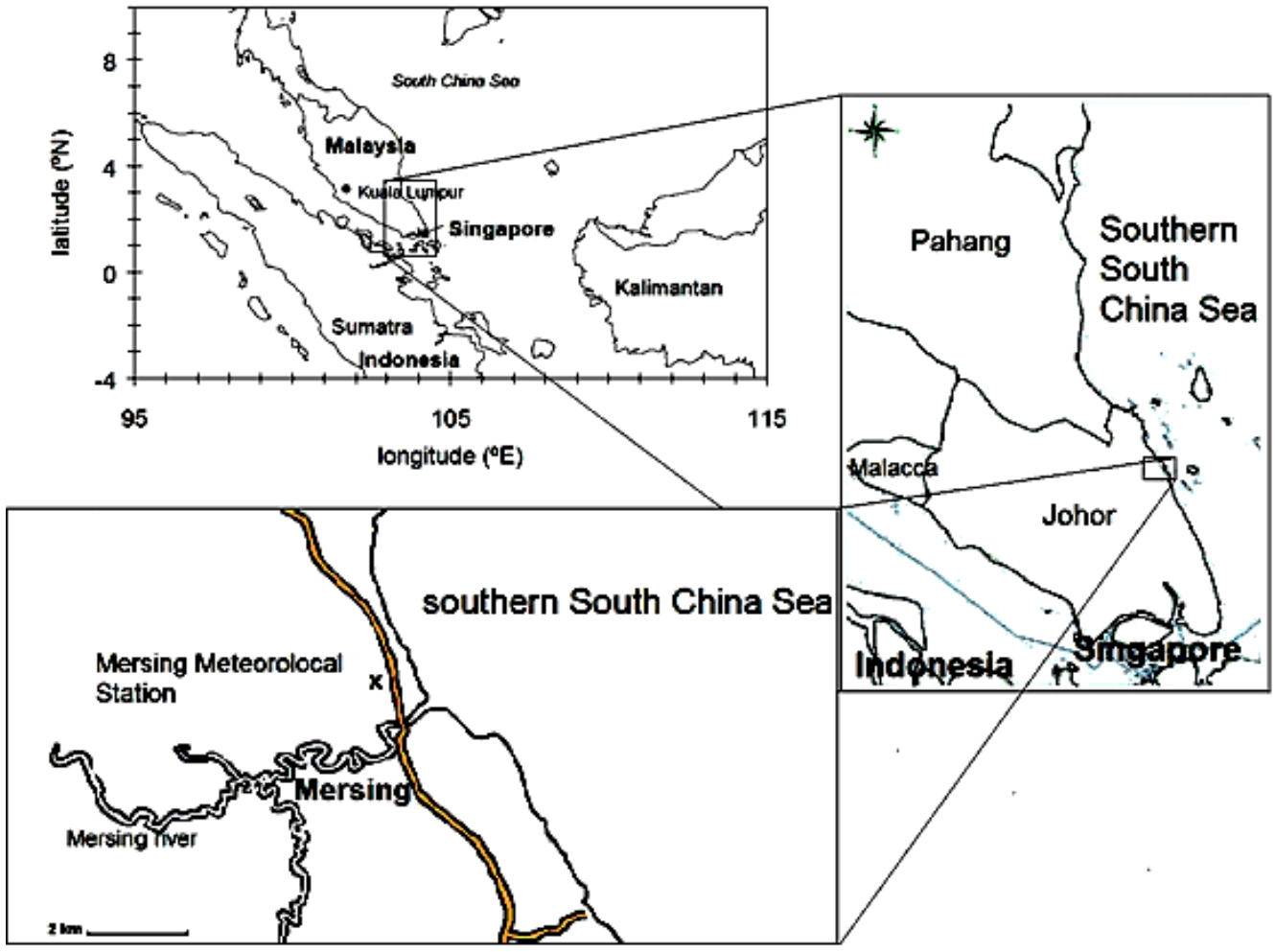

Figure 1: Location of Mersing Meteorological Station (marked with X) which was equipped with automated weather observation systems and high volume air sampler PM_{10}.

Analysis of metals

Working standards were prepared by diluting the PerkinElmer Pure Plus Multi-element Calibration Standard 3 solutions (CL-7-173YPY1) and calibration using the PerkinElmer Inc. ELAN 9000 ICP-MS (PerkinElmer Inc. USA). The detection limits (LODs) for all target analytes were calculated based on three times the standard deviation of blank (air filter blank). The value of LODs ranged from 0.01 (Pb) to 1.09 (Fe) µg/L reported in the instrument manual was used during the calculation. The precision and accuracy of the extraction procedures were evaluated using standard reference materials (SRM) of NIST SRM 1646a. The SRM samples in triplicate were processed following the same procedure as samples, and analyzed using ICP-MS. The obtained total concentrations were compared with the certified values and the recovery of total elements from the SRM samples were satisfactory with average mean ranging from 64% to 94%.

Results and Discussion

Variation of PM_{10} concentrations and meteorological parameters in Mersing

About 47 samples of PM_{10} collected at Mersing Meteorological Station during the two year sampling period between January 2009 to December 2010 had a mean concentration value of 28 ± 17 µg/m^3 and rangedfrom 8 µg/m^3 on 3 January 2009 to 87 µg/m^3 on 15 April 2009 (Table 1). Meanwhile, the annual mean concentrations of PM_{10} for years 2009 and 2010 in Mersing were 30 ± 14 µg/m^3 and 24 ± 15 µg/m^3, respectively. These values were distinctly lower than in other major cities in Malaysia such as in the Klang Valley [31,32], Johor Bahru, Kota Kinabalu, Kuantan and Nilai [33]. Published results from the inter-comparison studies of major Asian cities in most cases revealed high levels of PM_{10} and $PM_{2.5}$, especially during the dry season, and often exceeded USEPA annual standards [34-37]. It has recently been quite common for these major cities to obtain a higher range of PM_{10} concentrations; up to 150 µg/m^3, especially during episodic haze phenomena [38-40]. In this case, the Malaysian Department of Environment (DOE), through its concession company Alam Sekitar Malaysia (ASMA) Berhad, has been monitoring PM_{10} levels at 51 monitoring stations across Malaysia as part of the Malaysian Continuous Air Quality Monitoring (MCAQM) programme since 1996 [41]. Past research has also used DOE data to establish a clear link between seasonal monsoon variations with levels of PM_{10} across the country [31].

Figure 2b shows that the air quality in the east coast of Johor was generally clean with an Air Pollutant Index (API) value of less than 50 as an indication of good air quality status [40]. Nevertheless, 9–13% of the sampling period recorded showed a higher concentration of PM_{10} (Figure 2b), which is above the Recommended Malaysian Air Quality Guideline (RMAQG) with limited healthy level by European Union (50 µg/m^3) [41,42]. In recent years many studies have confirmed the effect of trans-boundary particulate matter on Malaysian air quality

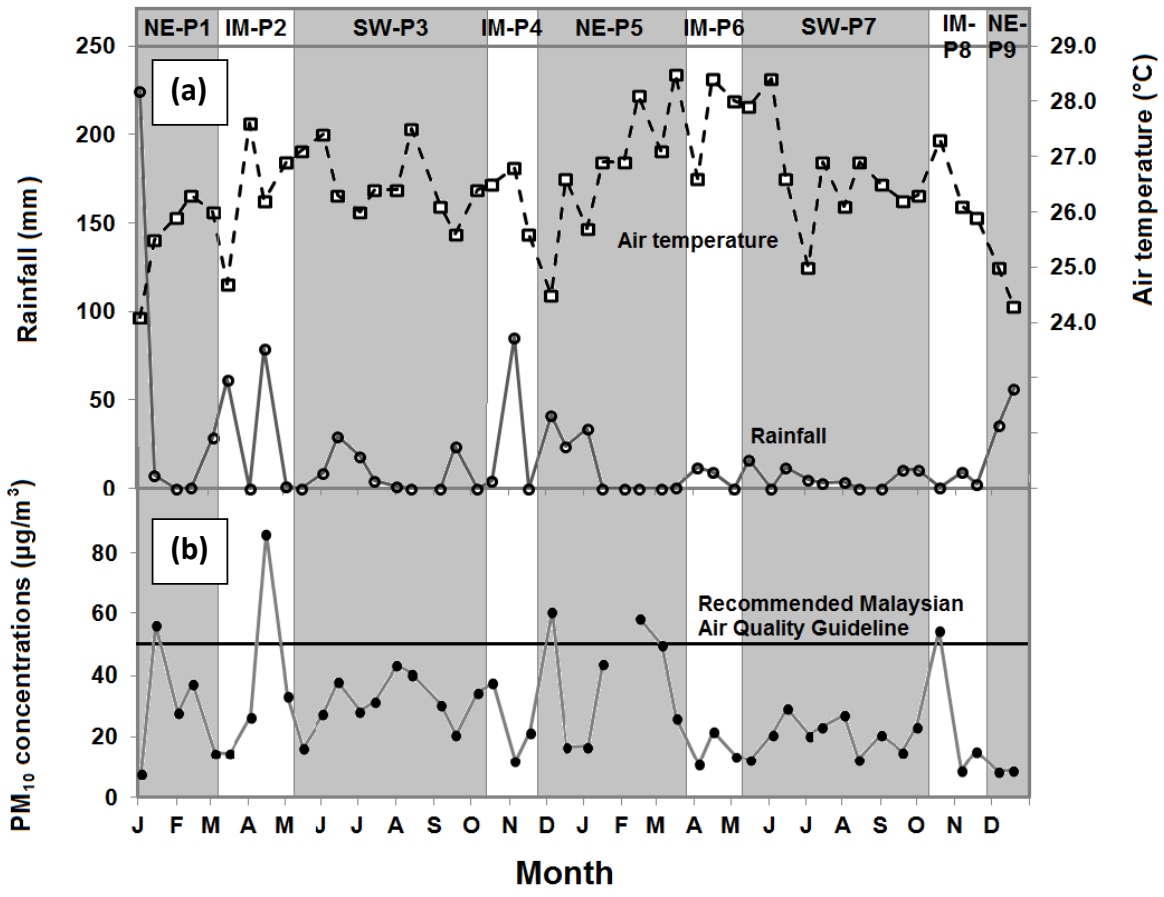

Figure 2: The temporal profile of (a) air temperature and rainfall in relation to the (b) PM_{10} concentrations as an indicator for the Mersing atmosphere synoptic pattern.

[31,41,43,44]. The highest PM_{10} values were recorded during the most serious haze event between August and September 1997 by all monitoring stations during the Sumatran forest fires [43].

According to the study by Juneng et al. [31], the annual PM_{10} fluctuations in Peninsular Malaysia were normally higher during the southwest monsoon (May-September). This feature has been confirmed to have been due to regional low level winds that were predominantly southerlies and southwesterlies which brought a dry season to the southwestern part of Peninsular Malaysia with a maximum PM_{10} concentration between May and September [31]. Somehow the annual cycle of PM_{10} concentrations in Mersing from the collected data showed a bimodal temporal variation with PM_{10} peaks during both monsoons in January-March and in May-September (Figure 2a). The bimodal characteristic of PM_{10} concentration corresponds to northern sources by easterly and northeasterly winds during the northeast monsoon, but the southern source brought during southwest monsoons are predominantly by southerly and southwesterly winds [45].

Figure 2 illustrates the daily air temperature and rainfall that identify the connection between these parameters with the variation of PM_{10} and its relation to regional synoptic patterns that validate atmospheric changes. The annual rainfall amount for 2009 and 2010 were 926.4 mm and 453.8 mm, respectively [46]. Whereas the mean air temperature for 2009 and 2010 were 26.2 and 26.7, respectively (Table 1). In general, the PM_{10} concentrations over Malaysia show distinctive seasonal variations and are strongly governed by rainfall regimes following the fluctuation of the Inter-tropical Convergence Zone (ITCZ) [32]. A weak negative statistical correlation (y=-0.3133x+27.099; r=0.139) between PM_{10} concentration and daily rainfall is validation for higher concentrations during the dry period of the southwest monsoon as compared to lower concentrations during the wet period of the northeast monsoon (Figure 3). Rashid and Griffiths [47] analyzed a single site in Kuala Lumpur and concluded that there is a marked seasonal variation in coarse particulate matter, with concentrations being the highest during dry periods and lowest in the wet periods. In this paper, the observed lower PM_{10} concentration in 2010 was accompanied by relatively warmer and drier months than in 2009. The results in Figure 2 showed no indication or clear pattern for rainfall and temperature effects on PM_{10} concentrations in this region.

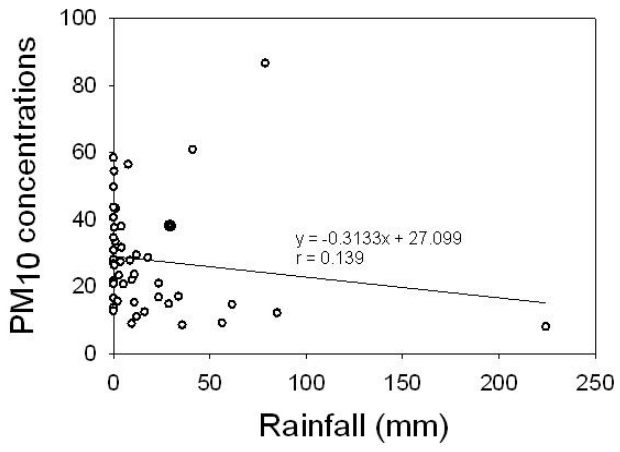

Figure 3: The statistical correlation between daily rainfall and PM_{10} concentrations at the Mersing Meteorological Station for 2009-2010.

Figure 2 shows each monsoon section according to their respective period such as the northeast monsoon (NE), the southwest monsoon (SW) and the inter-monsoons (IM). Northeast monsoon (NE-P1 & P5, November-March) conditions usually prevailed around November every year as winds over the region become progressively northeasterly. According to the National Environment Agency [48], the presence of the monsoon trough, coupled with the convergence of winds and strong solar heating over southern ASEAN continued to enhance convective activity and rainfall over the region. In contrast, weather conditions became drier in northern ASEAN due to the transition towards the traditional dry season. The low level jet bifurcates over the northern region and Indo-China, which results in a divergence zone and minimal precipitation. The ITCZ is located at its southern-most location south of the equator, indicating a dry period over the Indo-China region [31].

During the time period mentioned, a large number of forest fires were detected, mostly in the Indo-China region (e.g., Thailand, Myanmar, Laos, etc.) to the north of the Southeast Asian region [48,49,31]. Anthropogenic sources from Indo-China and Eastern China could impact the South China Sea regions during this time [50,51]. For the combined years 2009 and 2010, the mean PM_{10} concentration during the northeast monsoon (NE-P1 & P-5) in Mersing was 30.97 ± 19.69 $\mu g/m^3$ (Table 2). This value is comparatively higher than during the southwest monsoon and inter-monsoon events despite a relatively high mean daily rainfall of (32.34 ± 58.54 mm) and temperature (26.04 ± 1.34°C) values.

There was an emergence of hotspots with thin smoke plumes in the east coast of central Sumatra from late January until mid-February, 2009 [48] and mid-February, 2010 [49]. Smoke plumes and haze were also recorded in Miri, Sarawak during early and mid-March, 2010 [49]. However, wind directions suggest a possible weaker influence of a Sumatran source on the Peninsular Malaysia region during this period of time. Therefore, the high PM_{10} concentration during the late winter-spring is considered to be associated with the southward shift of the ITCZ and the emergence of hotspot counts and extensive biomass burning from the north, especially from Indo-China and the Philippines [31,48,49]. More smoke plumes with unhealthy air quality were detected in Myanmar, Laos and Thailand during the second half of the northeast monsoon in both years [48,49] and some peaks in Figure 2a were subsequently associated with smoke plumes and extensive forest burnings. Meanwhile a surge in rainfall activities such in early March 2009, end of December 2009 and November-December 2010 in Figure 2b effectively brought down the concentrations of airborne particulates.

Meanwhile, southwest monsoon conditions prevailed in the region in June with intensifying sub-tropical high pressure systems in the southern hemisphere leading to the strengthening of southeasterly winds south of the equator and the establishment of southwesterly winds in the region north of the equator [49]. This also led to the migration of the ITCZ rain belt by several degrees latitude northwards [32]. Generally dry weather conditions except for occasional shower activities affected the southern ASEAN region, including Mersing (Figure 2b). The rainfall received for most of the southern ASEAN region was also below normal with less than 50% of the normal rainfall [49]. Mersing also recorded a mean of far below normal; only 7.34 ± 8.73 mm during this period with the onset of the traditional dry season (Table 2).

Forest fires in southern Thailand, parts of Borneo and Sumatra, particularly in the fire-prone province of Riau, central and southern

Year	Parameter	Unit	N	Minimum	Maximum	Mean ± SD
2009	PM_{10}	µg/m³	18	7.9	60.7	29.83 ± 14.48
	Rainfall	mm	18	0.0	224.6	25.28 ± 52.80
	Temperature	°C	18	24.1	27.6	25.28 ± 1.00
2010	PM_{10}	µg/m³	23	8.4	58.3	23.59 ± 14.54
	Rainfall	mm	24	0.0	56.4	9.24 ± 14.10
	Temperature	°C	24	24.3	28.5	26.73 ± 1.13
Both	PM_{10}	µg/m³	47	7.9	85.5	27.84 ± 16.51
	Rainfall	mm	47	0.0	224.6	18.00 ± 36.83
	Temperature	°C	47	24.1	28.5	25.46 ± 1.04

N: Number of samples; SD: Standard deviation

Table 1: Descriptive statistical analysis of PM_{10} concentrations, rainfall amount and air temperature at Mersing Meteorological Station in years 2009-2010 (Data is a summary from Appendix A).

Period	Parameter	Unit	N	Minimum	Maximum	Mean ± SD
NE-P1 & P5	PM_{10}	µg/m³	14	7.9	60.7	30.97 ± 19.69
	Rainfall	mm	14	0.0	224.6	32.34 ± 58.54
	Temperature	°C	14	24.1	28.5	26.04 ± 1.34
SW-P3 & P7	PM_{10}	µg/m³	20	12.3	43.2	25.86 ± 8.88
	Rainfall	mm	20	0.0	29.4	7.34 ± 8.73
	Temperature	°C	20	25.0	28.4	26.60 ± 0.79
IM-P2 & P6 and IM-P4 & P8	PM_{10}	µg/m³	9	8.8	54.3	20.88 ± 13.78
	Rainfall	mm	9	0.0	61.6	10.60 ± 19.73
	Temperature	°C	9	24.7	28.4	26.69 ± 1.22

N: Number of samples; SD: Standard deviation

Table 2: Descriptive statistical analysis of PM_{10} concentrations, rainfall amount and air temperature at Mersing Meteorological Station at different periods (northeast monsoon [NE], southwest monsoon [SW] and the intermonsoons [IM]) (Data is a summary from Appendix A).

Kalimantan increased significantly and the biomass burning extended southward [48,49]. Also, according to Juneng et al. [31], there was a sudden upswing in the number of forest fires over southern ASEAN when the ITCZ reached the northern-most latitude. When the southwesterly winds prevailed in June, it affected neighboring southwest Peninsular Malaysia. The southerly and southwesterly winds also transported pollutants to the northern region thus causing PM_{10} to increase in this region during this period (Figure 2a). Soleiman et al. [52] analyzed three severe haze episodes in the Klang Valley area and concluded that the maximum PM_{10} coincided with the southwesterly winds. The long-range transportation of haze across the Malacca Straits to the southwest of the Malaysian Peninsular was also promoted by a rising number of forest fire occurrences in Sumatra [53]. In addition to seasonal variations, El Niño modulations may amplify the impact of haze and the concentration of pollutants in the region during the dry season of the southwest monsoon [31].

From the issued report by the National Environment Agency [48,49], the dry weather conditions in late May led to an increase in hotspot activities in Sumatra. Scattered hotspots with localized smoke plumes were detected mostly in northern and central Sumatra. Hotspots with moderate to thick smoke haze were most often detected in the Riau province. Transboundary smoke haze from these persistent hotspot clusters in the Riau province affected parts of Peninsular Malaysia on several occasions during the month. Mersing and other southern parts of Peninsular Malaysia and Singapore also experienced slightly hazy conditions when the smoke haze from South Sumatra was transported northwards by the prevailing shift in direction from southerly to southwesterly on several days in a month [48,49].

Between the two year period, the 2009 southwest monsoon (SW-P3) was recorded as having an approximately 34% higher mean PM_{10} concentration with mean 31.17 ± 8.40 µg/m³ than that in 2010 (SW-P7) with mean 20.55 ± 5.76 µg/m³ (Table 3). Based on the records as presented in Figure 2a, 2009 experienced more haze events compared to 2010. A high hotspot counts and sometimes thick smoke haze were detected mostly in the Riau province on 25 May 2009, 26 May 2009 and 10 June 2009 [48]. Consequently, the air quality in Selangor, Malaysia was in the unhealthy range on 12 June 2009 and in Port Klang, the Air Pollutant Index reached a high of 136 [54]. Meanwhile in 2010, the records showed relatively low hotspot counts in the region [49]. It has been explained that due to frequent shower activities in the region, hotspot activities in the entire ASEAN region continued to remain subdued in the southwest monsoon 2010 (SW-P7). Many parts of the southern ASEAN region received above normal rainfall later in October (Figure 2b).

The interchange between the northeast and southwest monsoons occurs from mid-March 2009 onwards; commonly referred to as the inter-monsoon (IM-P2 & P6), this is where prevailing winds gradually weaken and turn from northeasterly to light and variable in the ASEAN region [48]. Rainfall is predominantly characterized by afternoon thunderstorms and a high humidity over the region reduces the biomass burning over central Sumatra [31]. In this period, the traditional dry season continued to prevail over the northern ASEAN region of Cambodia, Laos, Myanmar, Thailand and Vietnam. Dry weather conditions continued to prevail and hotspot activities remained elevated, particularly during the first half of April 2009 [48]. However, our results showed markedly high PM_{10} concentrations during March-May 2009 (IM-P2) with mean 40.25 ± 31.80 µg/m³, as opposed to rather conservative PM_{10} concentrations during April-May 2010 (IM-P6) with mean 15.50 ± 5.72 µg/m³ (Table 4). Later in May, increased shower activities in the northern ASEAN region helps to ease hotspot counts.

Period	Parameter	Unit	N	Minimum	Maximum	Mean ± SD
SW-P3	PM_{10}	µg/m³	10	16.4	43.2	31.17 ± 8.40
	Rainfall	mm	10	0.0	29.4	8.52 ± 11.12
	Temperature	°C	10	25.6	27.5	26.52 ± 0.62
SW-P7	PM_{10}	µg/m³	10	12.3	29.3	20.55 ± 5.76
	Rainfall	mm	10	0.0	16.2	6.16 ± 5.84
	Temperature	°C	10	25.0	28.4	26.68 ± 0.95

N: Number of samples; SD: Standard deviation

Table 3: Descriptive statistical analysis of PM_{10} concentrations, rainfall amount and air temperature at Mersing Meteorological Station during the southwest monsoon [SW] of 2009 [P3] and 2010 [P7] (Data is a summary from Appendix A).

Period	Parameter	Unit	N	Minimum	Maximum	Mean ± SD
IM-P2	PM_{10}	µg/m³	4	14.5	86.5	40.25 ± 31.80
	Rainfall	mm	4	0.0	78.8	35.35 ± 40.85
	Temperature	°C	4	24.7	27.6	26.35 ± 1.24
IM-P4	PM_{10}	µg/m³	3	12.0	37.8	23.80 ± 13.04
	Rainfall	mm	3	0.1	85.0	29.73 ± 47.90
	Temperature	°C	3	25.6	26.8	26.30 ± 0.62
IM-P6	PM_{10}	µg/m³	3	10.9	21.9	15.50 ± 5.72
	Rainfall	mm	3	0.0	12.0	7.20 ± 6.35
	Temperature	°C	3	26.6	28.4	27.67 ± 0.95
IM-P8	PM_{10}	µg/m³	3	8.8	54.3	26.20 ± 24.57
	Rainfall	mm	3	0.4	9.5	4.03 ± 4.82
	Temperature	°C	3	25.9	27.3	26.43 ± 0.76

N: Number of samples; SD: Standard deviation

Table 4: Descriptive statistical analysis of PM_{10} concentrations, rainfall amount and air temperature at Mersing Meteorological Station during the 2009 northeast to southwest inter-monsoon [IM-P2], 2009 southwest to northeast inter-monsoon [IM-P4], 2010 northeast to southwest inter-monsoon [IM-P6], and 2009 southwest to northeast inter-monsoon [IM-P8] (Data is a summary from Appendix A).

Meanwhile, the interchange between southwest to northeast monsoon (IM-P4 and P8) conditions with light and variable winds prevailed in the region in the first half of November before transitioning to the northeast monsoon season around mid-November. The transition marked the start of the rainy season in the southern ASEAN region and the dry season in the northern ASEAN region [49]. Hotspot activities in other parts of the southern ASEAN region were also subdued by the prevailing wet conditions which resulted in lower PM_{10} concentrations (Figure 1a), except for isolated hotspots detected in the Riau province of Sumatra in late November during a brief dry interval [48]. Our record showed relatively low concentrations for both in 2009 (IM-P4) and 2010 (IM-P8) with 23.80 ± 13.04 μg/m^3 and 26.20 ± 24.57 μg/m^3, respectively (Table 4).

Trace element compositions in PM_{10} at Mersing

The temporal variations of 15 trace elements measured in this study also showed fluctuations (Appendix A). Thus, seasonal changes of metals in aerosol is categorized into: i) Northeast monsoon NE-P1 and NE-P5; ii) Southwest monsoon SW-P3 and SW-P7; iii) Inter-monsoon IM-P2 and IM-P6; and iv) Inter-monsoon IM-P4 and IM-P8 (Table 5). The identification of particulate matter sources in the study is allocated based on the dominance of major element compositions in the aerosol. The concentrations of trace elements were found to have different signatures and variations according to time-interval classifications (monsoon and inter-monsoon periods), so that the nature and characteristics of each trace element can be explained. In this study, ambient samples refers to a baseline of atmospheric conditions set at a PM_{10} concentration of less than 20 μm/m^3 as the reference to each time-interval classification.

During the northeast monsoon (NE-P1 and NE-P5), trade winds from the northeast towards the east coast of Peninsular Malaysia as well as the southern part of the South China Sea is marked with higher concentrations of Ca, Fe, Mn, Mg, Ni and Cd than the ambient value at a ratio of 1.40 ± 0.11, 1.40 ± 0.19, 1.26 ± 0.18, 2.50 ± 0.42, 1.40 ± 0.29 and 1.15 ± 0.20, respectively (Table 6). Most of these metals are abundant and part of the important elements in terrestrial soils, and by-product of biomass burning and smoke plumes [11,55-58]. Thus the presence of elements such as Ca, Fe, Mn and Mg may coincide with the transfer of long-range air mass transport brought within the northeasterly winds during the monsoon. This indicates the presence of terrestrial soil dust and forest fires particles in our PM_{10} samples, brought via atmospheric transport from northern Asia during the northeast monsoon. A study by Lin et al. [51] has also shown that the input of anthropogenic sources from Indo-China and eastern China has reached a major part of the South China Sea.

The northeast monsoon was also characterized by the higher presence of anthropogenic sources associated to emissions of forest fires and fossil fuel burning such as Cu, Ni and Cd (Table 5) compared to ambient sources. Anthropogenic trace metals such as Pb, Cd, Zn,

Element	N	NE-P1&P5	N	IM-P2&P6	N	SW-P3&P5	N	IM-P4&P8	N	Ambient
Al	13	48.8 – 67.6	13	42.7 – 64.7	21	36.4 – 75.4	6	48.1 – 64.3	14	48.8 – 68.2
		(59.4 ± 5.6)		(56.6 ± 7.2)		(61.9 ± 8.4)		(55.9 ± 6.8)		(60.2 ± 6.0)
Ca	11	0.67 – 2.77	7	0.58 – 1.37	13	0.25 – 1.73	3	0.97 – 1.37	13	0.58 – 1.62
		(1.41 ± 0.66)		(1.03 ± 0.24)		(1.29 ± 0.38)		(1.15 ± 0.20)		(1.01 ± 0.26)
Fe	11	0.04 – 0.52	8	0.05 – 0.34	18	0.00 – 0.88	3	0.05 – 0.24	11	0.10 – 0.41
		(0.15 ± 0.02)		(0.17 ± 0.09)		(0.25 ± 0.18)		(0.14 ± 0.10)		(0.20 ± 0.09)
Mn‡	13	3.9 – 18.0	13	2.2 – 13.2	21	3.9 – 16.9	6	3.0 – 12.1	14	3.9 – 13.2
		(11.5 ± 4.0)		(8.1 ± 3.1)		(10.9 ± 3.1)		(8.3 ± 3.2)		(9.1 ± 2.1)
K	13	27.4 – 44.5	13	21.9 – 44.8	20	18.7 – 46.7	6	27.8 – 35.8	14	27.4 – 44.8
		(36.0 ± 5.4)		(34.5 ± 5.9)		(36.6 ± 5.9)		(32.1 ± 2.9)		(36.0 ± 5.4)
Ba	13	47.7 – 85.3	13	56.0 – 79.0	21	50.1 – 108.2	6	56.0 – 75.7	14	61.6 – 95.1
		(66.2 ± 10.3)		(68.5 ± 7.3)		(68.4 ± 13.0)		(64.9 ± 7.2)		(71.6 ± 8.4)
Na	13	73.4 – 114.4	13	60.5 – 106.2	21	46.8 – 107.2	6	67.8 – 91.0	14	73.4 – 106.2
		(92.9 ± 11.3)		(85.3 ± 11.8)		(91.9 ± 12.7)		(82.9 ± 8.5)		(91.6 ± 9.4)
Mg	11	0.02 – 0.88	4	0.02 – 0.22	8	0.04 – 0.34	2	0.04 – 0.22	7	0.02 – 0.27
		(0.35 ± 0.34)		(0.12 ± 0.10)		(0.20 ± 0.12)		(0.13 ± 0.13)		(0.14 ± 0.10)
Sr	8	0.01 – 0.28	6	0.03 – 0.30	12	0.01 – 0.41	3	0.03 – 0.25	11	0.01 – 0.36
		(0.14 ± 0.10)		(0.19 ± 0.10)		(0.28 ± 0.12)		(0.16 ± 0.12)		(0.16 ± 0.12)
Co‡	13	0.07 – 0.60	13	0.03 – 0.52	21	0.17 – 1.04	6	0.36 – 0.52	14	0.07 – 0.55
		(0.39 ± 0.16)		(0.33 ± 0.13)		(0.44 ± 0.20)		(0.42 ± 0.06)		(0.37 ± 0.14)
Pb‡	11	0.1 – 38.2	7	0.7 – 19.2	8	3.1 – 29.1	4	0.7 – 19.2	8	3.0 – 38.2
		(11.6 ± 10.4)		(7.3 ± 7.2)		(17.8 ± 8.4)		(9.9 ± 9.1)		(14.5 ± 13.4)
Cu‡	13	16.5 – 38.6	13	9.6 – 28.8	21	7.7 – 103.8	6	10.6 – 28.8	14	15.4 – 38.2
		(20.7 ± 5.9)		(17.5 ± 5.3)		(25.0 ± 19.6)		(19.4 ± 6.5)		(19.2 ± 5.7)
Ni‡	11	0.1 – 26.7	9	0.0 – 6.6	19	0.4 – 19.5	4	0.2 – 6.6	10	0.1 – 16.7
		(6.1 ± 8.1)		(2.2 ± 2.1)		(5.0 ± 4.8)		(2.4 ± 2.9)		(4.4 ± 4.9)
Cd‡	12	0.23 – 0.89	12	0.10 – 1.10	20	0.19 – 1.10	6	0.10 – 1.10	13	0.23 – 1.01
		(0.53 ± 0.24)		(0.43 ± 0.25)		(0.56 ± 0.23)		(0.44 ± 0.35)		(0.46 ± 0.20)
Cr‡	12	11.2 – 52.9	12	4.9 – 44.5	20	6.8 – 65.8	6	4.9 – 44.5	13	23.9 – 53.6
		(32.0 ± 9.5)		(25.1 ± 11.5)		(36.4 ± 15.2)		(23.7 ± 15.8)		(35.3 ± 9.8)

N: Number of samples; ( ): Mean value

Table 5: Descriptive statistical analysis of trace elements concentrations (μg/m^3) (except with the '‡' marks, ng/m^3) from the collected PM_{10} samples from Mersing Meteorological Station (Data is a summary from Appendix A).

Ti and As are usually associated to the significant emissions from oil and gas industries attached to the aerosol and particulate matter [59]. While other anthropogenic sources from forest fires events are also significant to elements such as K, Na, Mg, Cl, Fe, Zn, Br, Cd, Cu and Pb [11,57,60,61]. Therefore, in accordance with earlier discussions, atmospheric input from northern Asia during this period is well associated with hot spot emergence in parts of Indo-China and northern ASEAN (e.g., Thailand, Myanmar, and Laos).

Thus, elemental characterizations during the southwest monsoon (SW-P3 and SW-P7) can also be attributed to air mass transport by southerly or southwesterly winds as described above. Therefore, most airborne particulate sources can be presumed to have originated from the southern Asian region. It has been described that dry weather in southern ASEAN during this period led to an increase in hotspot counts and consequently peat bog and forest fire incidences at prone areas such as the Riau province [48,49]. The results in Table 6 show a ratio of almost all elemental concentrations that indicated a notable increase from ambient concentrations, especially Sr (1.75 ± 0:21), Mg (1:43 ± 0:25, Ca (1:28 ± 0.11), Fe (1.25 ± 0.17), Pb (1.23 ± 0.13), Cd (1:22 ± 0:21), Mn (1:20 ± 0.19), Cu (1.20 ± 0.13), and Ni (1:13 ± 0:23).

It shows that the prevailing southerly or southwesterly winds that normally emerge in June has brought some contaminants of terrestrial soil origin pollutants (i.e., Ca, Fe, Mn and Mg), biomass burning (i.e., Ca, Mn, Cd and Pb), and anthropogenic petroleum industries (i.e., Pb, Cu, Ni and Cd) from the southern Asian region. Moreover, the data showed that long-range air mass transport from southern ASEAN brings terrestrial contaminants during the southwest monsoon more intensively than during the northeast monsoon. The transport of haze smoke and particles from Sumatra across the Straits of Malacca was relatively significant to Peninsular Malaysia and Singapore [31,48,49] and therefore permits a trans-boundary issue within the South China Sea region.

In addition, there are two inter-transitional periods between two inter-monsoon events (i.e., IM-P2 and IM-P6; IM-P4 and IM-P8) that have different characteristics and are worthy of further discussion. Although both have almost the same features in terms of the prevailing weaker winds in fickle directions, there are also differences in the contents of particulate matter between them. According to Table 4, PM_{10} concentrations during IM-P2 and IM-P6 will be characterized as high with a wider range of 10.9–86.5 μg/m^3 compared to IM-P4 and IM-P8 with 8.8–54.3 μg/m^3. In fact, elemental compositions based on their concentrations were also different. For example, based on concentration ratios to ambient in Table 6, it was found that a majority of terrestrial soil origin elements such as Al, Fe, K, Ba, Na and Mg were comparatively lower in IM-P4 and IM-P8 than in IM-P2 and IM-P6. This signature was also similar to anthropogenic-bound elements such as Pb, Cu, Ni, Cd and Cr. This shows the introduction of natural and anthropogenic sources such as soil dust, sea spray, forest and biomass burning, and emissions from oil and gas industries were higher in IM-P2 and IM-P6 (northeast to southwest) than in IM-P4 and IM-P8 (southwest to northeast). However, these values were always small and negligible compared to both monsoons.

Application of multivariate statistical methods for source apportionment

The application of the Positive Matrix Factorization (PMF) using EPA PMF 3.0 software to our trace metals data obtained during the two year sampling has shown some of the features which have already been found on the basis of regional PM_{10} fluctuations. Positive matrix factorization (PMF) is a multivariate factor analysis tool that can be interpreted by factor contributions and factor profiles [62]. According to Abdul Rahman et al. [58], the inclusion and exclusion of species and samples can significantly influence PMF results. Therefore, time series of elemental concentrations of the PMF model were used to identify values that appeared abnormal when compared to the overall data. Only variables with less than 20% of missing values were considered in the analysis. In this study, about 7% of the dataset were excluded from the model in order to avoid unrealistic apportionment.

Trace elements	NE-P1&P5	IM-P2&P6	SW-P3&P5	IM-P4&P8
Al	0.99 ± 0.17	0.95 ± 0.17	1.03 ± 0.17	0.93 ± 0.14
Ca	1.40 ± 0.11	0.93 ± 0.11	1.28 ± 0.11	1.14 ± 0.08
Fe	1.40 ± 0.19	0.95 ± 0.19	1.25 ± 0.17	0.70 ± 0.09
Mn	1.26 ± 0.18	0.87 ± 0.14	1.20 ± 0.19	0.91 ± 0.11
K	1.00 ± 0.13	1.02 ± 0.14	1.02 ± 0.13	0.89 ± 0.12
Ba	0.92 ± 0.11	1.00 ± 0.13	0.96 ± 0.12	0.91 ± 0.09
Na	1.01 ± 0.11	0.95 ± 0.12	1.00 ± 0.13	0.91 ± 0.11
Mg	2.50 ± 0.42	0.71 ± 0.14	1.43 ± 0.25	0.93 ± 0.19
Sr	0.88 ± 0.12	1.38 ± 0.14	1.75 ± 0.21	1.00 ± 0.15
Co	1.05 ± 0.15	0.70 ± 0.09	1.19 ± 0.15	1.14 ± 0.13
Pb	0.80 ± 0.17	0.26 ± 0.05	1.23 ± 0.22	0.68 ± 0.08
Cu	1.08 ± 0.11	0.83 ± 0.10	1.20 ± 0.13	1.01 ± 0.11
Ni	1.40 ± 0.29	0.45 ± 0.11	1.13 ± 0.23	0.54 ± 0.12
Cd	1.15 ± 0.20	0.91 ± 0.16	1.22 ± 0.21	0.96 ± 0.16
Cr	0.91 ± 0.10	0.75 ± 0.08	1.03 ± 0.11	0.67 ± 0.07

Table 6: The ratios of metal concentrations in all periods to ambient concentrations from the collected PM_{10} samples from Mersing Meteorological Station (Data is a summary from Table 5).

Among other considerations that should be made is to determine the number of factors involved. There are a number of methods that can be used in selecting the number of factors to retain. In general, the best approach is to go through the complete analysis several times assuming various numbers of factors thereafter making a decision on the number of factors to extract. Roscoe [63] did a study of the interpretation of compositional particulate data set, where he described the criterion for determining the number of retained eigenvectors. In this study, eigenvectors with an eigenvalue greater than one is considered a significant factor. In order to obtain a clear pattern of factor loading and interpretable results, the varimax approach (varimax rotation) to factor axes rotation is used [64]. A plot of eigenvectors as a function of factor numbers for the data set is shown in Figure 4. The scree plot shows there were four factors with eigenvalue greater than or equal to one of the data sets. After the fourth factor, the decline in the eigenvalue gradually levels off up to the fifteenth factor.

Overall, the results of the yield analysis were meaningful indicators of the sources of PM_{10}; e.g., forest fires with additional heavy fuel oil combustions, emissions from industry and fossil fuel combustions, terrestrial soil dust, and a mixture of additional/unidentified input (Figure 5). The factor compositions and the temporal variations of each source are shown, where the Mersing sampling station is explained by the abundance of sea spray in the aerosol. The Na and Mg (sea salt factors) concentrations appears in all four factors, representing continuously significant sea salt interference in aerosols. However, sea salt appears to be relatively unrelated to the total mass PM_{10} concentrations, only contributing less than 10% of the total variation of PM_{10} [58].

The first factor was determined to be forest and biomass fires particles with the addition of heavy fuel oil combustions, and the

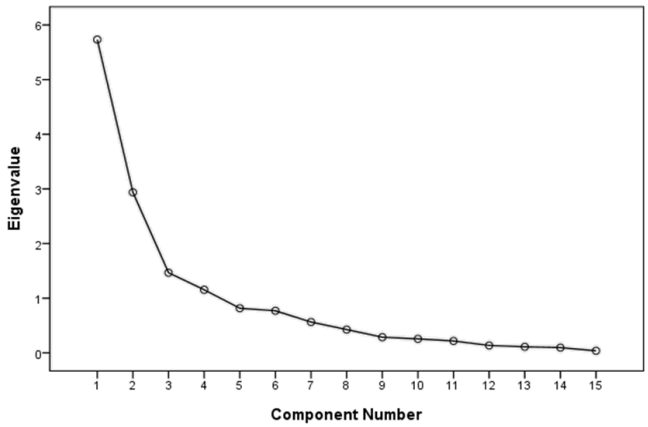

Figure 4: Eigenvalue for 2009-2010 metal concentrations in PM_{10} at Mersing Meteorological Station as a function of factor numbers.

major species contributions were Fe, Mn, Cu, Cd, Cr and Ni (Figure 5). According to Anttila et al. [61], the biomass fire episode has significantly caused K^+, Ca^{2+}, NO_3^-, Cd, Cu, Mn, Pb and Zn accompanied with gaseous O_3 and SO_2, PAH_s to elevate. Wild forest fires have also been estimated to emit considerable amounts of Cu, Mn and Zn [65]. This factor contributes as much as 53.1% to total PM_{10} variations in 2009-10 (Figure 5a). The time series plot shows that this factor contribution is relatively higher during the southwest monsoon, attributed to drier conditions. Therefore, this factor was more significant by southerly and southwesterly winds between May to October. The data from SW-P3 showed a slightly higher mean of PM_{10} concentration than SW-P7 as discussed above, which could be due to drier and warmer conditions in the sampling region (Table 3). In addition, more shower activities were recorded for SW-P3 (Figure 2b) helping to ease biomass fire particles. Meanwhile, it is highly probable that the biomass fire aerosol was accompanied by other factors, specifically from heavy fuel oil combustion (i.e., Ni and V) which is commonly associated with large marine diesel engines.

The second factor represents anthropogenic sources from industrial and fossil fuels (mainly from motor vehicles) emissions even though this factor was almost negligible to the total PM_{10} mass. The source fingerprint was characteristically high in Pb and Cd (Figure 5b), where trace metals i.e., Pb, Cd, Zn, Ti and As are typically accumulated in the anthropogenically-bound sources in the fly ash of oil shale [59]. This factor has strong peaks which are almost similar to the first factor, and were relatively higher during the southwest monsoon especially during the SW-P7 events. The same reason also due to drier and warmer environment conditions would being adopted which a highly possible. This could be the explanation for the additional anthropogenic inputs such as Cu, Ni and Cr detected in the biomass burning aerosol. Therefore one can conclude that industrial and fossil fuels emissions were reasonably similar in terms of their behavior to atmospheric changes and variability.

Meanwhile, the third factor is characterized with a high loading of most of the major elements and markers for terrestrial soil and crustal dust e.g., Al, Ca, Fe and Mn, with relatively constant temporal variation contributing to 34.9% of the total PM_{10} mass (Figure 5c). There were some peaks in relation to this factor during NE-P1, IM-P2, SW-P3 and NE-P5 which could be related to highest PM_{10} peaks in Figure 2a. This aeolian dust could therefore originate largely from regions with massive arid desert areas, named Asian Dust and Australian Dust, together with the suspension of local dust. One hypothesis posits that soil dust sources from these Asian arid regions are transported to lower latitude areas [66]. This aeolian dust can be transferred a thousand kilometers from its source, which is usually an arid area [67-69]. Asian dust storms usually occur seasonally in the northern part of China during spring [70,71] and are transported by the frontal systems during the cold surges of the northeast monsoon.

A study in Taiwan has found that approximately 25–120% of monthly PM_{10} contents during spring were dominated by Asian dust enriched aerosols [72]. In addition, an attempt by Zhang et al. [73] to perform chemical analysis of Asian dust particles from the Gobi desert have shown 84% domination of crustal source trace elements as Al, Si, Ca, Ti, Fe and K from its total mass. Basically, trade winds during the northeast monsoon would bring significant sources from the north of East Asia while the southwest monsoon is related to sources from the south of East Asia. The elemental signature of this factor shows that these aeolian sources from Asian and Australian dust phenomena could be important.

The fourth and final factor may be attributed to an unknown source with a considerable mixture of soil dust component (Al, Ca, K, Ba), sea salt component (Na, Mg, Sr), and other heavy metals (Cu, Cr) (Figure 5d). This factor is a complex mixture of anthropogenic and natural emissions marked for 12% of the total PM_{10} mass. Nevertheless, its contribution to the aerosol consistently fluctuated, while the peaks could be associated with the peaks of the other three factors [74].

Conclusions

The concentrations of PM_{10} in Mersing, Johor between 2009 and 2010 was highest during the northeast monsoon, followed by the southwest monsoon. Meanwhile, the long-range air mass transport containing terrestrial dust, biomass and forest fires, fossil fuel combustions and emissions from oil and gas industries have been identified as the major anthropogenic components for aerosol in this region as showed by the PMF model. Hot and dry meteorological conditions in the northern ASEAN region during the NE monsoon and southern ASEAN during the SW monsoon will increase the extent of peat and biomass fires as a significant source of atmospheric pollution, significantly altering aerosol chemical compositions. Also, about 35% of terrestrial dust as one of the important sources to the region specifically to Malaysian waters, which containing such as iron could provide essential nutrients for ocean ecosystems and therefore contribute to biological aspects of the ocean. The role of aerosols in climate change is extremely important due to overall correlations between trends in atmospheric releases to regional meteorological conditions. A biomass and forest fires in the ASEAN region is a trans-boundary pollution issue and has a significant impact on the Malaysian ocean ecosystem as well as on distant countries. Thus, further work on aerosol inventories and chemical speciation in maritime air is needed to improve the relationship between global and regional inventories.

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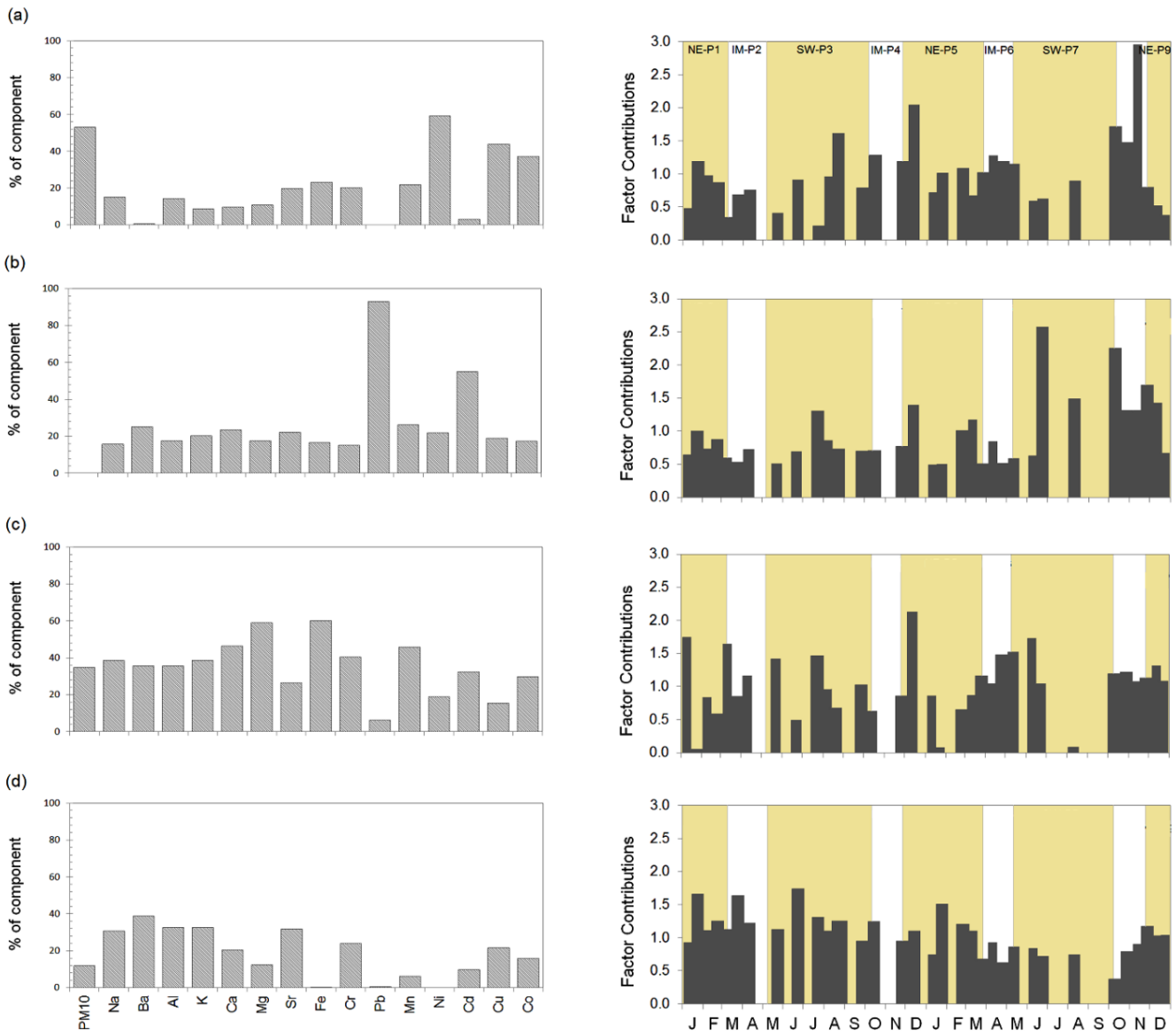

Figure 5: Source profile and time series plots for each source to the aerosols at Mersing Meteorological Station. (a) Forest fires, with additional heavy fuel oil combustions, (b) emissions from industry and fossil fuel combustions, (c) terrestrial soil dust, and (d) mixture of additional/unidentified input.

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