LONG-TERM TRENDS AND POTENTIAL ASSOCIATED SOURCES OF PARTICULATE MATTER (PM_{10}) POLLUTION IN MALAYSIA

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ABSTRACT
Particulate matter (PM_{10}) is an important pollutant particularly in urban environments in Malaysia. In addition, the level of this pollutant was also seasonally significant in most parts of Malaysia, and therefore concern of its effect towards human health is relevant and crucial. Based on a long-term series of PM_{10} measurement at 20 monitoring locations in Malaysia, this study analysed the spatial and temporal characteristics of PM_{10} from 1997 to 2015 using standard deviation ellipse and trend analyses. Satellite data and HYSPLIT model were applied to investigate the seasonal potential sources of the pollutant. Results show that annual PM_{10} average concentrations were greatly varied with large coefficient variation. In term of trend analysis, 11 monitoring sites had shown significant but small decreasing trends. Meanwhile, 7 monitoring sites had shown no significant trends and only 2 monitoring sites showed increasing trends. Trajectory analysis using the HYSPLIT model for the investigation of potential sources of pollutant has shown that high pollution levels of PM_{10} in Malaysia corresponded to the biomass burning in neighbouring countries. During the southwest monsoon, high PM levels were observed in the central and southern parts of Peninsular Malaysia and Malaysian Borneo, which corresponded to the biomass burning in Indonesia. Based on the long-term analysis, PM_{10} pollution in Malaysia was characterised by transboundary pollution as well as local sources, especially in urban areas. Despite the recognition of small but significant decreasing trends of PM_{10} pollution over long-term period, special attention need to be focused on short-term pollution episode, particularly related to transboundary pollution during extreme weather condition such as El Niño event to ensure that human health on a wider population is protected.
Keywords: air quality, biomass burning, El Niño, PM_{10}, transboundary pollution.

1 INTRODUCTION

Particulate matter (PM) is composed by particles both solid and liquid (fluid) in suspension due to their small size [1]. These particles have a diverse composition, size, and sometimes are called aerosols or dust, but widely known as particulate matter (PM). Airborne particulate matter (PM) are classified as PM_{10} (particles with an aerodynamic diameter, Ø, less than 10 µm), $PM_{2.5}$ (Ø < 2.5 µm), and PM_1 (Ø < 1 µm). According to [2], [3] the coarse particles (PM2.5–10 µm) has predominantly natural sources including the geological and biological materials. Meanwhile, the fine fractions ($PM_{2.5}$ and PM_1) are dominated by combustion-derived particles consisting mainly of organic and inorganic elements adsorbed onto the surface of a carbonaceous core, and the secondary particles produced by photochemical reactions in the atmosphere [3], [4]. The transport and fate of aerosol particles relatively depends on their size and the meteorological conditions, and for this reason, the concentration, composition and size distribution of atmospheric particles are temporally and spatially variable [5]. On the other hand, the size of atmospheric particles is proportional to their emission sources' particles sizes, which typically those emitted from anthropogenic sources are smaller than from natural sources [6].

On the basis of available scientific studies about human health, the coarse fraction is mainly deposited in the extra thoracic region and has been found to be associated with pro-

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inflammatory and cytotoxic effects [7]. Meanwhile, exposure to fine and coarse aerosol particles may reach the alveolar region of the lungs and this has been associated mainly to a higher genotoxic potential [9]. Analytical study for both a single-pollutant model and two-pollutant model for natural mortality that was conducted in Malaysia reveals that relatively significant associations with respiratory mortality at different lags were found for all pollutants except SO_2 [10]. PM_{10} and daily mean of O_3 had the highest relative risk (RR) in the single pollutant model. These findings indicated that an increase in exposure to PM_{10} was associated with a maximum increase of 0.99% in natural mortality and a 3.63% increase in respiratory mortality. Exposure to elevated levels of particulates has been associated with health effects in many epidemiological studies in temperate countries, as well as in tropical countries [11], [12]. Several studies recently conducted in North America and Europe have analysed the effects of air pollution on mortality by seasons and have provided important evidence that the effects of air pollution depend on temperatures [13].

Particulates come from emissions of diverse pollutants from different stationary and mobile sources, and from the reaction between primary pollutants that formed secondary pollutants [14]. The major sources of fugitive dust emission are traffic on paved and unpaved roads, construction, agricultural operation, mineral industries, and wind erosion from both agricultural and non-agricultural lands [15]. The processes involved produce direct emission of loose particles by the wind, abrasion of immobile aggregates and crust by saltation impacts and creep-sized aggregates and particles into suspension size.

In the case of Malaysia, the concentration of air particulate matter is influenced by the southwest monsoon wind and the occurrence of biomass burning during dry season that transported over to Malaysia [16], [17]. Furthermore, Malaysia lies in the main pathway of the Southeast Asian pollution outflow, which also contributes to significant local aerosol and pollutant emissions [18]. Most of these studies were carried out in the Klang Valley area, which is the worst area affected by transboundary haze from Sumatra. The burning of peat soil and plant waste in Sumatra, Indonesia and Indochina release vast quantities of smoke, consisting of a high quantity of particulate matter, into the atmosphere [16], [19]. During normal periods, the level of particulate matter is mostly influenced by local biomass burning, traffic and industries [17]. Generally, the level of particulate matter tends to be lower during the rainy October–November period [20], which is partially due to the particulate washout effects and the absence of strong ground-based inversions [21]. In view of these serious implications of particulate pollution from a wide range of potential sources and subjected to the influence of various factors, this study aims to identify the spatial and temporal variations of PM_{10} over Malaysia for a long-term period (1997–2015). Assessment on the long-term pollution trend, influence of the seasonal and El Niño events and the transboundary pollution sources will be explored to enhance the regional understanding of the PM_{10} pollution variations over the region and factors that influence their trends and associated sources.

2 METHODOLOGY

2.1 Study area

The present study focuses on the analysis of PM_{10} at 20 monitoring stations of the 61 monitoring stations nationwide that are currently operated by the Malaysian Department of Environment. From all 20 air quality monitoring stations, six stations are located each in industrial areas, urban areas and sub-urban areas, respectively, and two in rural areas (Table 1 and Fig.1). The air quality monitoring stations were divided into five sub regions, namely

Table 1: Locations of the selected air quality monitoring stations in Malaysia.

Sub-Region	Monitoring Stations	Location	Coordinate (Lat, Long)	Area Status
Northern Peninsular Malaysia	S1	Kangar	6.38, 100.24	Sub-urban
	S2	Alor Setar	6.14, 100.38	Urban
	S3	Perai	5.38, 100.39	Industrial
	S4	Taiping	4.87, 100.68	Industrial
Central Peninsular Malaysia	S5	Cheras	3.21, 101.27	Urban
	S6	Shah Alam	3.11, 101.56	Urban
	S7	Port Klang	3.01, 101.41	Urban
Eastern Peninsular Malaysia	S8	Tanah Merah	5.81, 102.13	Industrial
	S9	Kuala Terengganu	5.31, 103.12	Urban
	S10	Jerantut	3.81, 103.30	Rural
	S11	Kuantan	3.94, 102.37	Sub-Urban
Southern Peninsular Malaysia	S12	Nilai	2.82, 101.81	Industrial
	S13	Seremban	2.72, 101.97	Urban
	S14	Melaka	2.22, 102.24	Urban
	S15	Kota Tinggi	1.56, 104.23	Urban
	S16	Pasir Gudang	1.47, 103.91	Industrial
Malaysian Borneo	S17	Bintulu	3.17, 113.04	Sub-urban
	S18	Miri	4.49, 114.04	Sub-Urban
	S19	Labuan	5.31, 115.22	Sub-urban
	S20	Kota Kinabalu	5.89, 116.04	Urban

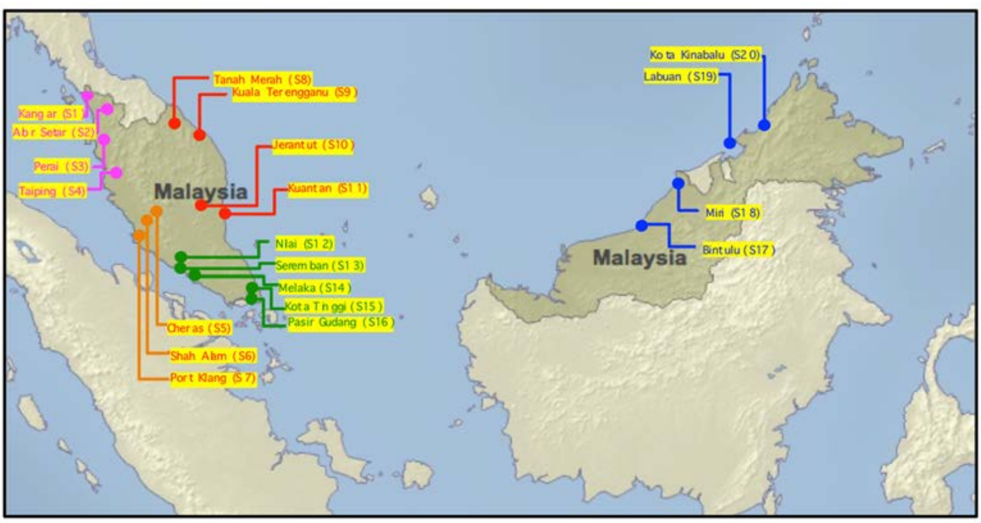

Figure 1: Selected air quality monitoring stations at five sub regions over Malaysia.

the central, eastern, northern, and southern regions of Peninsular Malaysia and Sabah and Sarawak region of the Malaysian Borneo.

2.2 Meteorology and PM_{10} datasets

The climate of Malaysia is typically a tropical climate, which is characterized by warmth and humidity throughout the year. Due to its location in the central part of Southeast Asia, Malaysia is governed by the regional wind systems, which result from the atmospheric pressure distribution over the region. The climatic variation is influenced by the monsoons resulting from the seasonal fluctuation of the inter-tropical convergence zone (ITCZ) in the South China Sea area. The annual movements of the ITCZ and the associated "trade" wind fields produced two monsoonal seasons, namely the Northeast Monsoon (NEM) (November to February) and the Southwest Monsoon (SWM) (June to August). The two monsoons are separated by two transitional periods. During the NEM, steady easterly or north-easterly winds of 10 to 20 knots prevail. The winds over the east coast states of Peninsular Malaysia may reach 30 knots or more during periods of strong surges of cold air from the north (cold surges). During the SWM, the prevailing wind flow is generally south-westerly and light, below 15 knots. During the inter-monsoon seasons, the winds are generally light and variable.

The PM_{10} air quality datasets were obtained from the Malaysian Department of Environment (DOE) for the selected monitoring stations across Malaysia over a period of nineteen years (1997–2015). The PM_{10} concentration was measured by continuous air quality monitoring (CAQM) program using the β-ray attenuation mass monitor (BAM-1020). The BAM-1020 is controlled by an advanced microprocessor system that makes it fully automatic. The diurnal, seasonal and yearly PM_{10} concentrations of the air quality time series were visualized using line graph while the variations used the errors bar from the standard deviation of the averaging diurnal, seasonal and yearly PM_{10} concentration data. For the computation of diurnal variations, hourly data was used while for the computation of seasonal and yearly variations, daily data was used.

2.3 Trend analysis and coefficient of variations

In exploring the interannual variation characteristics of PM_{10} over Malaysia, trend analysis was applied to analyse the temporal dynamic. In this study, the Mann-Kendall (MK) test was considered to assess the trends in the time series of annual values of PM_{10} concentrations. The Mann-Kendall test is applicable to the detection of a monotonic trend of a time series with no seasonal or other cycles. The coefficient of variation (CV), which is defined as the ratio of the standard deviation (σ) to the mean (μ), was also calculated to convey the extent of variability relative to the mean PM_{10} concentrations during the study period. This coefficient was used to examine the interannual variability of PM_{10} from 1997 to 2015.

2.4 Trajectories analysis

The emission sources and transport pathways of the high PM_{10} values were further investigated by backward trajectories using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) Model. The model is provided by the US National Oceanographic and Atmospheric Administration through the website http://www.arl.noaa.gov/ready.html at different monsoon seasons in Malaysia. The model calculation method is a hybrid between the Lagrangian approach, using a moving frame of reference for the advection and diffusion calculations as the trajectories or air parcels move from their initial location, and the Eulerian methodology, which uses a fixed three-dimensional grid as a frame of reference to compute

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pollutant air concentrations [22]. HYSPLIT trajectory error calculation was estimated 10–30% of the distance travelled after 24 hours to the direction of flow [23]. The trajectory is also not representing the path of an air parcel within the planetary boundary layer (PBL) as the air parcel quickly loses its identity through turbulent mixing processes [24]. However, the HYSPLIT model is adequate to classify regional-scale air mass motions in which local scale winds are embedded.

Back trajectories at the selected stations were developed for 48 hours on the three monsoonal seasons (NEM, SWM, and intermonsoon) to determine the long-range transport of PM_{10}. To investigate the potential sources of PM_{10}, regional fire maps were obtained from the Fire Information for Resource Management System (FIRMS) web fire mapper (https://firms.modaps.eosdis.nasa.gov/firemap/). FIRMS fire hotspot records are based on data acquired by the Moderate Resolution Imaging Spectroradiometer (MODIS) observation instrument on board the NASA's Terra and Aqua EOS satellites which passes over the Southeast Asia region at least four times daily. Fire detection is performed using a contextual algorithm that exploits the strong emission of mid-infrared radiation from fires. The MODIS algorithm examines each pixel of the MODIS swath, and ultimately assigns to each one of the following classes: missing data, cloud, water, non-fire, fire, or unknown [25].

3 RESULTS AND DISCUSSIONS

3.1 Spatial and temporal variation of PM_{10}

The spatial and temporal variations of monthly PM_{10} concentrations at the five selected sub regions over Malaysia from 1997 to 2015 are shown in Fig. 2. In northern Peninsular, the monthly PM_{10} concentrations showed distinct spatial patterns and large variations. A long-term measurement of PM_{10} concentration at Perai was comparatively higher than other stations of 55.3μg/m^3, while PM_{10} concentration at Alor Setar was relatively lower of 35.5μg/m^3. For the whole period of measurement, the highest daily mean ever recorded was at Kangar in October 2015 with a concentration of 338.08 μg/m^3. The annual PM_{10} averages over the sub regions were greatly varied with a large coefficient variation of between 26.0% and 32.1%. There were small decreasing trend of PM_{10} in Kangar, Perai and Taiping at 95% confidence interval trend were observed but no significance trend in Alor Setar (Table 2).

Central Peninsular sub region maybe consider as the most polluted in term of PM_{10} in Malaysia, with average PM_{10} concentration of 59.2 μg/m^3, which was relative the highest in all the sub regions. Relatively, Port Klang notably experienced the highest PM_{10} concentration in the sub region of 66.3 μg/m^3, while Shah Alam was experiencing the lowest concentration of 52.3 μg/m^3. The highest daily mean ever recorded was at Port Klang in October 2015 with a concentration of 346.43 μg/m^3. Similarly, with the northern Peninsular region, the annual PM_{10} averages over the central Peninsular that is greatly varied with a large coefficient variation of between 28.3% and 56.4%. In this subregion, there was a mixed trend, where a small increasing trend in Shah Alam, a decreasing trend in Cheras and no significant trend in Port Klang were observed (Table 2).

Meanwhile, the eastern Peninsular sub region was comparatively the least polluted in the Malaysian Peninsular region with a PM_{10} average concentration of 41.3μg/m^3. Relatively, Jerantut monitoring station showed the highest PM_{10} concentration in the sub region with 49.3 μg/m^3, while Kuala Terengganu experienced the lowest concentration of 34.8 μg/m^3. During the long-term measurement period, Kuantan recorded the highest daily mean of 275.25 μg/m^3 in September 2015. The annual PM_{10} averages over the sub region were also varied with a large coefficient variation of between 21.6% and 31.4%. Small but significance

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decreasing trends were observed in all monitoring stations except Kuala Terengganu where no significance trend was observed (Table 2).

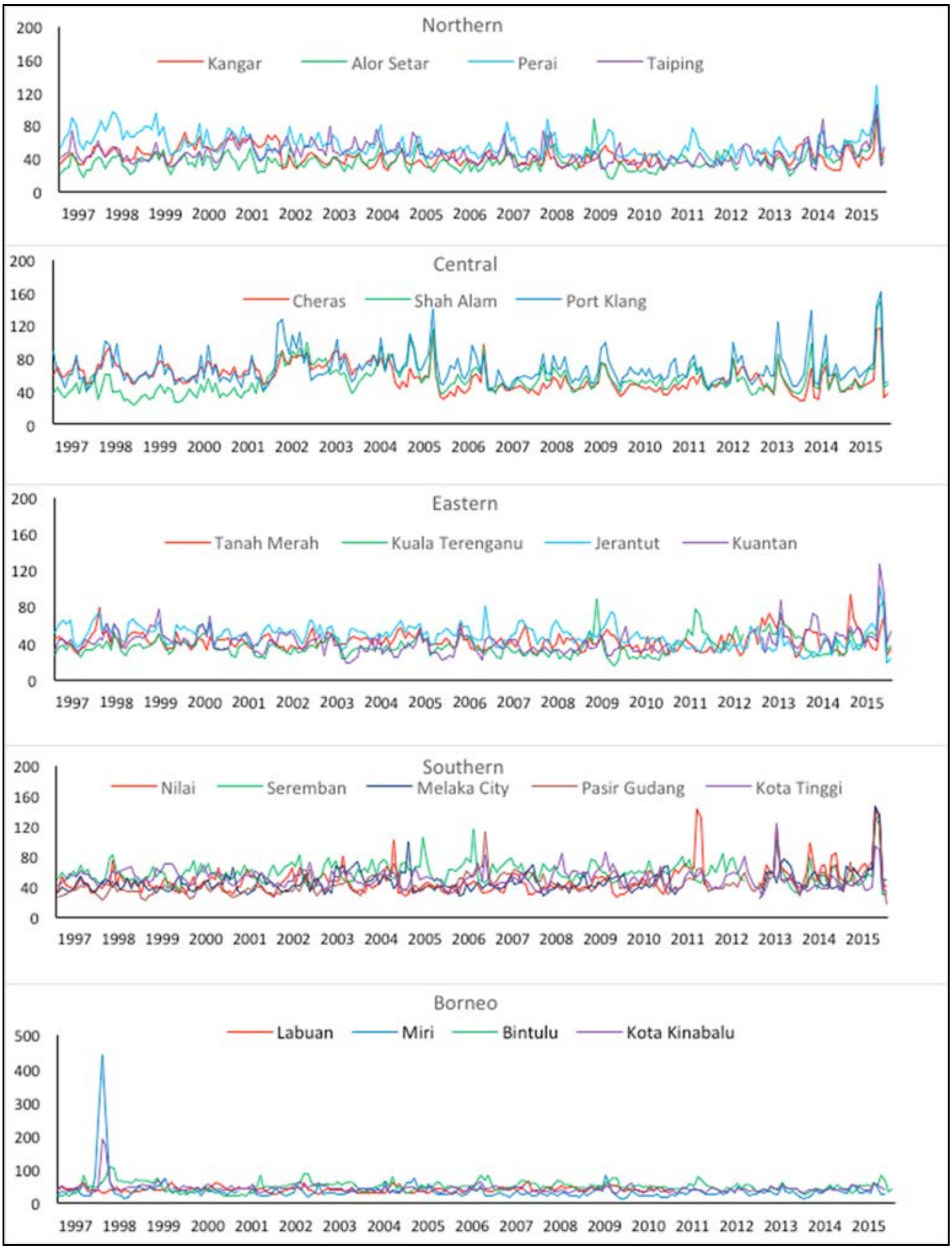

Figure 2: Time series of the monthly mean PM_{10} concentrations at twenty monitoring stations in five sub-regions over Malaysia.

Table 2: Values of minimum, mean, maximum, coefficient of variation (CV) and confidence interval trends of PM_{10} concentrations from 1997 to 2015.

Sub-Region	Min	Mean	Max	CV	95% Confidence Interval Trend
Northern Peninsular	15.1	44.3	125.8	32.0	
• Kangar	24.9	42.1	87.4	25.7	(-0.070, -0.041)
• Alor Setar	15.1	35.5	103.0	32.1	No Trend
• Perai	30.7	55.3	125.8	26.0	(-0.117, -0.087)
• Taiping	26.1	43.8	100.7	26.9	(-0.066, -0.040)
Central Peninsular	21.9	59.2	159.0	31.2	
• Port Klang	39.8	66.3	159.0	28.3	No Trend
• Shah Alam	21.9	52.3	147.8	35.2	(0.045, 0.079)
• Cheras	32.4	61.3	115.6	56.4	(-0.120, -0.088)
Eastern Peninsular	15.1	41.3	124.9	28.6	Decreasing
• Tanah Merah	24.9	41.3	91.2	21.6	(-0.032, -0.011)
• Kuala Terengganu	15.1	34.8	86.5	31.2	No Trend
• Jerantut	17.1	49.7	101.9	22.4	(-0.086, -0.061)
• Kuantan	18.6	39.0	124.9	31.4	(-0.062, -0.033)
Southern Peninsular	16.5	47.8	144.7	30.9	
• Nilai	26.6	45.1	140.2	31.9	No Trend
• Seremban	28.3	57.6	130.5	23.8	No Trend
• Melaka	25.0	43.5	144.7	34.1	(0.028, 0.055)
• Pasir Gudang	16.5	41.9	130.9	34.0	(0.072, 0.096)
• Kota Tinggi	30.4	50.2	90.8	21.7	(-0.040, -0.013)
Malaysian Borneo	15.1	41.0	436.8	56.1	
• Bintulu	16.7	46.9	436.8	33.2	No Trend
• Miri	15.1	39.8	58.9	95.1	(-0.049, -0.022)
• Labuan	22.0	36.7	187.6	21.4	No Trend
• Kota Kinabalu	24.7	39.8	499.3	40.7	(-0.054, -0.035)

The Southern Peninsular sub region, which is bordering the central Peninsular to the north and Singapore to the south, is the second most urbanised and industrialised after Klang Valley in central Peninsular. This sub region showed the least distinctive spatial and temporal patterns of PM_{10} concentrations over a long-term measurement. Relatively, Seremban monitoring stations showed the highest concentration, while Pasir Gudang showed the lowest concentration with record values of 57.53 $\mu g \cdot m^{-3}$ and 41.93 $\mu g \cdot m^{-3}$ respectively. Seremban was also observed to record the highest daily mean PM_{10} concentration of 353.83 $\mu g \cdot m^{-3}$ in September 2015. The annual PM_{10} averages over the sub region were also varied with a large coefficient variation of between 21.7% and 34.1%. This sub region was observed to have a mixing trend, where small but significant decreasing trends were observed in Pasir Gudang and Kota Tinggi, a small increasing trend was observed in Melaka and no trends were observed in Nilai and Seremban (Table 2).

Across the South China Sea, the Malaysian Borneo sub region has recorded relatively the lowest PM_{10} concentration over the period of measurement with 40.70 $\mu g \cdot m^{-3}$. This sub region also showed the least distinctive spatial and temporal patterns over the periods of measurement. The highest and lowest PM_{10} concentrations were at Bintulu and Labuan with concentration values of 46.9 µg·m-3 and 36.7 µg·m-3 respectively. Meanwhile, this region has recorded the highest levels of PM_{10} concentration in Malaysian air quality history by reaching unhealthy levels of 436.8 µg·m-3 in September 1997 during the large and intense biomass burning in Kalimantan, Indonesia. Similarly, with other sub regions in Malaysia, the annual PM_{10} averages in the sub region were greatly varied with a large coefficient variation of between 21.4% and 95.1%. Small but significant decreasing trends were also observed in Miri and Kota Kinabalu but no significant trends were observed in Bintulu and Labuan (Table 2).

3.2 Associated sources and meteorological factors of PM_{10}

Over the period of measurement, the highest average of PM_{10} concentration in each sub region was further evaluated to determine its associated sources and the roles of regional meteorological conditions, particularly the El Niño event. The highest PM_{10} concentrations at all monitoring stations were observed in the month of September and October 2015 except Miri, which was observed in September 1997. Between September and October 2015, and September 1997, large distributions of hot spots, which symbolised intense biomass burning were detected by satellite remote sensing in Southeast Asia particularly in Sumatra and Kalimantan, Indonesia. Large biomass burning in the region has attributed to the significant emission of dust particles into the atmosphere. The regional wind speed and direction during this period was strongly influenced by the southwest monsoon that is responsible for the transboundary pollution, thus enhancing the PM_{10} concentration for the whole Peninsular region between September and October 2015.

Based on the calculated backward trajectories at Kangar (northern), Port Klang (central), Kuantan (eastern) and Seremban (southern) using the HYSPLIT model, biomass burning in Sumatera, Indonesia has been identified as potential associated sources through transboundary pollution (Fig. 3). The air mass trajectories at the three different altitudes were observed to come from the same direction in some cases and at different origins arriving at the locations during the southwest monsoon. The long-range transboundary pollution was the main cause for the thick haze in Malaysian Peninsular during this period. The highest PM_{10} concentration records during this period were 338.08 $\mu g \cdot m^{-3}$ (Kangar – northern Peninsular), 346.43 $\mu g \cdot m^{-3}$ (Port Klang – central Peninsular), 275.25 $\mu g \cdot m^{-3}$ (Kuantan – eastern Peninsular) and 353.83 $\mu g \cdot m^{-3}$ (Seremban – southern Peninsular).

For the Malaysian Borneo sub region, the highest PM_{10} concentration (436.77 $\mu g \cdot m^{-3}$) was recorded at Miri monitoring station in September 1997 during the dry period of southwest monsoon. All these values have exceeded the Malaysian Ambient Air Quality Guideline Standard of 150 $\mu g \cdot m^{-3}$. During this period, large hotspots of biomass burning were detected from the satellite image in Kalimantan, Indonesia (Fig. 3). Backward trajectory analysis using HYSPLIT at this station has identified that the air mass was originated from the Kalimantan region (Fig. 3). Thick haze and high particulate pollution in this sub region in September 1997 was found to be associated with the large biomass burning in Kalimantan, Indonesia. These analyses and assessments affirmed the analyses of the earlier studies [1], [10], [11] that biomass burning and transboundary pollution from Indonesia were responsible for the high concentrations of PM_{10} in all sub regions in Malaysia.

This study also investigates the role of the extreme weather conditions such as El Niño phenomenon on the spatial and temporal variations of PM_{10} particularly during the period when each monitoring sites recorded the highest concentration levels of PM_{10}. El Niño events in the Southeast Asia region from the period between 1997 and 2015 were analysed.

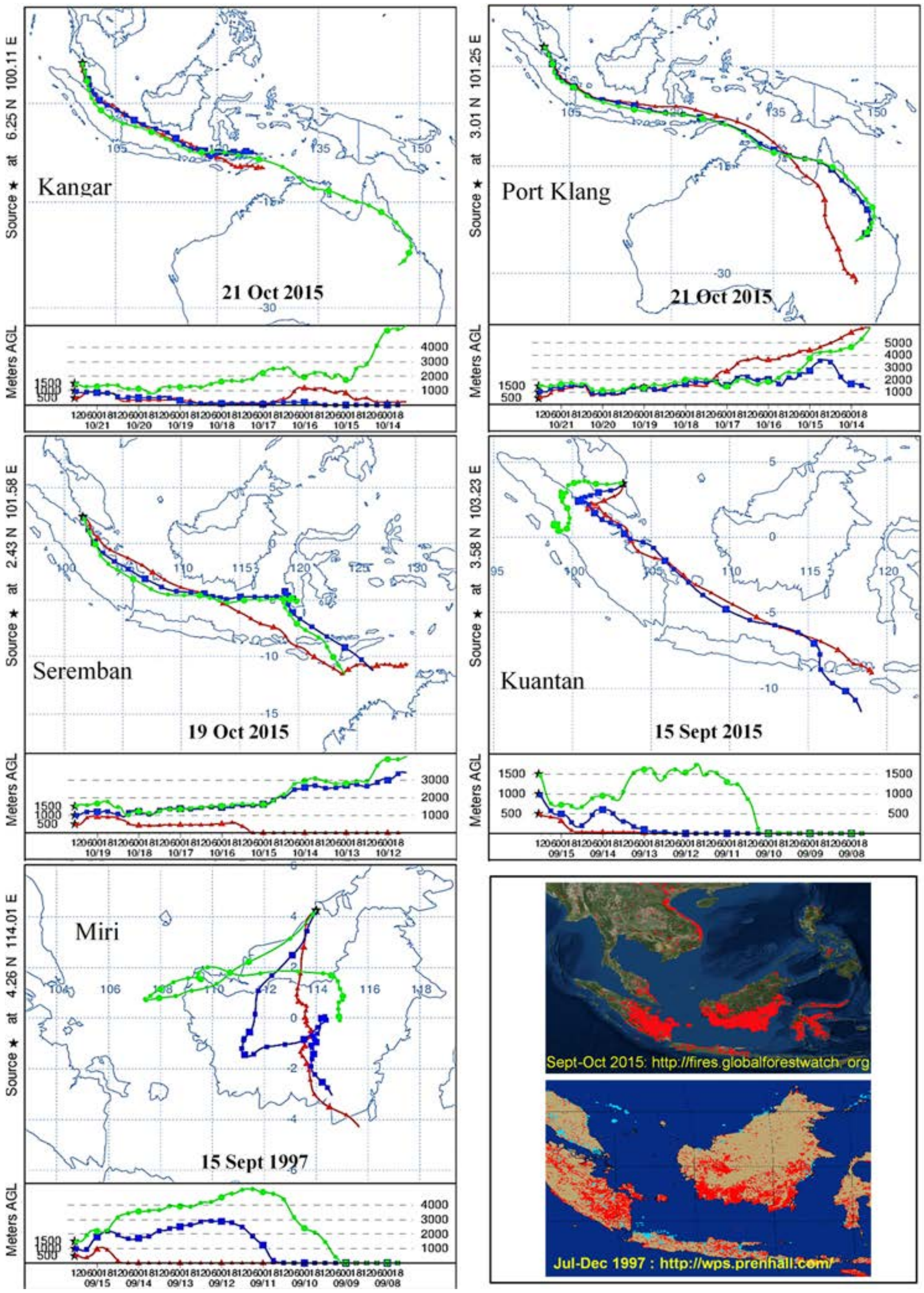

Figure 3: Backward air mass trajectory analysis (first, second and bottom-left panels) and MODIS fire records between 8–15 October 2015 and 16–23 September 1997 (bottom-right panel).

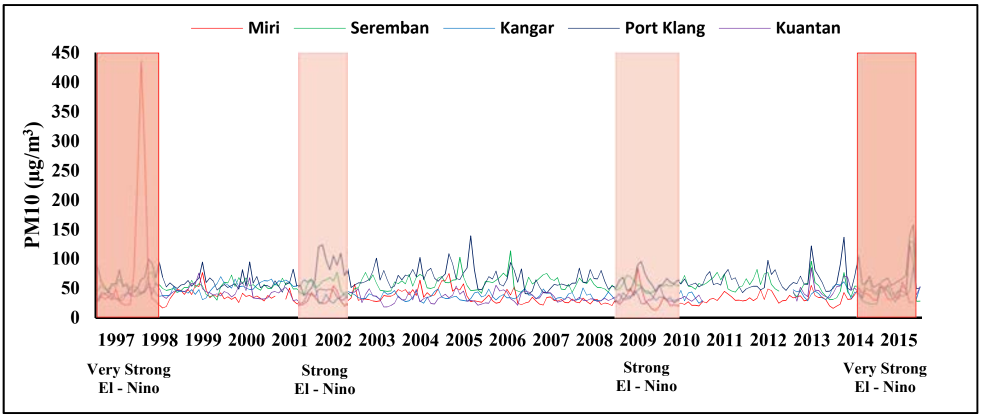

Figure 4: Time series analysis for selected monitoring stations (1997–2015) with El-Niño events.

During this period, seven El Niño events were recorded with the strongest intensities (super El Niño) were recorded in the year 1997 and 2015 (Fig. 4). El Niño effects have also played an important role on high PM_{10} concentration levels in Malaysia. The persistently high PM_{10} concentrations in all sub regions over Malaysia in 1997, 2002, 2009 and 2015 were primarily due to long-range transboundary pollution from the large biomass burning in Indonesia and coincidence with the occurrence of the El Niño events in this region. During the super El Niño events in the region, PM_{10} concentrations were significantly higher than usual for all the monitoring stations, being notably higher in Kangar, Port Klang, Kuantan and Seremban during the 2015 El Niño and Miri during the 1997 El Niño.

Nevertheless, the El Niño event alone does not directly cause the rise in PM_{10} concentration but the sub effects from the El Niño event are amplifying the cause of the PM_{10} increment. One of the most significant effect of the El Niño event is extremely dry seasons, coupled with high air ambient temperature. These conditions have led to severe drought and caused large-scale forest fires in Sumatera and Kalimantan in Indonesia, which were reflected during the super El Niño events. During the non-El Niño events or weak El Niño events, persistent high PM_{10} concentrations over Malaysia have also been identified due to long-range transboundary pollution from the neighbouring countries, particularly over the Peninsular region. From the satellite images (http://fires.globalforestwatch.org), large hot spot areas in Sumatera were observed in September and October between 2011 and 2014. The strong wind during the southwest monsoon, which generally occurred from June to October, accelerates the particulate transport towards the Peninsular Malaysia, thus enhancing the PM_{10} concentration over the region.

4 CONCLUSION

This study examined the spatial and temporal distribution of daily and monthly mean PM_{10} concentration based on a long-term monitoring data to render the air quality trends in Malaysia. The datasets spanned from 1997 to 2015, consisting of 20 selected monitoring stations across Malaysia, which were divided into five sub regions, namely the northern Peninsular, central Peninsular, Eastern Peninsular, Southern Peninsular and Malaysian Borneo. This study also assessed the potential associated sources and meteorological factors linked to the high PM_{10} concentrations. Long-term PM_{10} measurement in Malaysia has

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indicated a small increasing trend over the years in the northern and central peninsular regions. The highest concentration of PM_{10} at five monitoring stations representing the five sub regions were analysed to determine the potential associated sources and the roles of meteorological condition such as the El Niño events. Highest concentrations of PM_{10} were observed during the southwest monsoon during September and October in 1997 and 2015. Based on backward trajectories analysis using HYSPLIT, large and intense biomass burning in Sumatera, Indonesia was found to be the associated sources of PM_{10} in the Peninsular Malaysia region, while large and intense biomass burning in Kalimantan, Indonesia was found to be the associated sources of PM_{10} in Malaysian Borneo. As a conclusion, seasonal PM_{10} pollution in Malaysia is strongly related to long-range transboundary pollution due to biomass burning in the neighbouring countries. Extreme weather conditions such as El Niño events occurred at the strongest intensities in the region historical records that occurred in September and October of 1997 and 2015. The extremely dry season and warm ambient air temperature during this period promoted forest fires and delayed the wash off of aerosol by rainfall. Assessment on the daily highest PM_{10} concentrations at the selected five monitoring stations in all the five sub regions shows that concentrations were strongly linked with the El Niño events.

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