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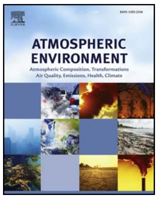

Review article

# Impact of regional haze towards air quality in Malaysia: A review

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## ABSTRACT

Haze is a common phenomenon afflicting Southeast Asia (SEA), including Malaysia, and has occurred almost every year within the last few decades. Haze is associated with high level of air pollutants; it reduces visibility and affects human health in the affected SEA countries. This manuscript aims to review the potential origin, chemical compositions, impacts and mitigation strategies of haze in Malaysia. “Slash and burn” agricultural activities, deforestation and oil palm plantations on peat areas, particularly in Sumatra and Kalimantan, Indonesia were identified as the contributing factors to high intensity combustions that results in transboundary haze in Malaysia. During the southwest monsoon (June to September), the equatorial SEA region experiences a dry season and thus an elevated number of fire events. The prevailing southerly and south-westerly winds allow the cross-boundary transportation of pollutants from the burning areas in Sumatra and Kalimantan in Indonesia, to Peninsular Malaysia and Malaysian Borneo, respectively. The dry periods caused by the El Niño - Southern Oscillation (ENSO) prolong the duration of poor air quality. The size range of particulate matter (PM) in haze samples indicates that haze is dominated by fine particles. Secondary inorganic aerosols (SIA, such as $SO_4^{2-}$ and $NH_4^+$) and organic substances (such as levoglucosan, LG) were the main composition of PM during haze episodes. Local vehicular emissions and industrial activities also contribute to the amount of pollutants and can introduce toxic material such as polyaromatic hydrocarbons (PAHs). Haze episodes have contributed to increasing hospital visits for treatments related to chronic obstructive pulmonary diseases, upper respiratory infections, asthma and rhinitis. Respiratory mortality increased 19% due to haze episodes. Children and senior citizens are more likely to suffer the health impacts of haze. The inpatient cost alone from haze episodes was estimated at around USD 91,000 per year in Malaysia. Almost all economic sectors also experienced losses, with the heaviest losses in the agriculture and tourism sectors. This review suggests several ways forward to reduce haze episodes in SEA and Malaysia. These include economic approaches, research collaborations and science-

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policy interface. Improving forecasting capabilities can help reduce response time to burning events and subsequently reduce its impacts. Lastly, commitment and involvement by individuals, government agencies, and the entrepreneurial private sectors are crucial to reduce biomass burning (BB) and haze episodes in SEA.

## 1. Introduction

Haze is a phenomenon that occurs when a sufficient concentration of aerosols in the atmosphere scatter visible light and this results in a measurable reduction in visual range (Seinfeld and Pandis, 2006). Haze formation is highly related to meteorological conditions, emissions of pollutants and gas-to-particle conversion (Sun et al., 2006; Watson, 2002). Haze occurs under stable weather conditions such as low wind speed and inverted thermodynamic structure and is formed due to high levels of particles and gas-to-particle conversion (Niu et al., 2016). Haze can also occur due to high amounts of secondary gas such as surface ozone which can interact with volatile organic compounds to form secondary organic aerosols (SOAs) (Liu et al., 2016; Seinfeld and Pandis, 2006; Sun et al., 2016; Wu et al., 2016). Haze can be defined as the existence of dry particles and smoke in the atmosphere when the relative humidity is considered lower than usual ($< 80\%$) and visibility is below 10 km (Kusumaningtyas and Aldrian, 2016; Sun et al., 2016).

Haze conditions have been correlated with industrial activities, traffic emissions and biomass burning (BB) (Kunii et al., 2002; Liu et al., 2016). Incomplete combustion, particularly from large areas of BB usually contributes high amounts of smoke and fine particles to the atmosphere (Abdullah et al., 2012; Fujii et al., 2014; Pongpiachan et al., 2017). These fine particles are able to be transported across borders from their main sources due to regional wind directions (Amil et al., 2016; Kusumaningtyas and Aldrian, 2016). Meteorological factors such as temperature, rainfall, relative humidity, and atmospheric stability as well as physical and chemical interactions will determine the fate of suspended particles and their composition during transport processes (Dotse et al., 2016; Khan et al., 2016a). Due to the persistence of fine particles in the atmosphere and the ability of particles to be transported over large areas, the effects of haze are usually regional in scale (Heil et al., 2007; Huang et al., 2016; Koe et al., 2001).

Haze from BB due to forest fires and peat burning is of major concern because of its adverse impact on regional air quality in Southeast Asia (SEA) (Chuang et al., 2016; Huang et al., 2016; Nichol, 1997; Pani et al., 2016a; See et al., 2007; Taylor, 2010). Almost every year, haze episodes resulting from BB affect the economy and daily lives of people in this region, especially in Indonesia, Malaysia and Singapore (Awang, 2000; Nichol, 1997; Quah and Johnston, 2001). Haze episodes contribute large amounts of air pollutants which have a negative effect on human health (Afroz et al., 2003; Behera et al., 2015a; Khamkaew et al., 2016; Quah, 2002). Findings from scanning electron microscope data showed that 94% of the particles in the haze were below 2.5 μm in diameter and therefore can easily bypass the normal body defence metabolism and penetrate deeply into the alveoli of the lungs (Ramadhan et al., 2017). Furthermore, fires from BB potentially contribute to global warming and climate change due to the emission of large amounts of greenhouse gases and other pyrogenic products (Huang et al., 2013). Therefore, understanding the haze phenomenon is very important for future planning and mitigation procedures. Hence, this paper specifically discusses the historical and recent haze episodes in Malaysia due to BB in SEA. Factors that contribute to the haze episodes are discussed using air pollutant data and meteorological parameters acquired from the Malaysian Department of Environment (DOE) and the Malaysian Meteorological Department (MET Malaysia). An analysis of particulate matter (PM) and its composition is presented to indicate the levels of air pollutant compositions and to understand the sources and impacts of air pollutants during haze episodes.

## 2. Chronology of haze episodes in Malaysia

Haze in Malaysia is not a new phenomenon as it was first recorded back in the year 1982 when regional haze from BB disrupted daily life in Malaysia (Mohd Shahwahid and Othman, 1999; Sham, 1984). Since then, several haze episodes have been recorded. These episodes included severe events when the concentrations of PM with an aerodynamic diameter of less than 10 μm ($PM_{10}$) far exceeded the Malaysian Air Quality Guideline for $PM_{10}$ concentration (150 $\mu g\,m^{-3}$ for a 24-h average) at one or more locations throughout Malaysia. These severe episodes occurred in 1997, 2005 and 2015. Prior to the setting up of the continuous ambient air quality monitoring network by the DOE in 1996, three haze episodes were recorded in the years 1990, 1991 and 1994. During these haze episodes, visibility was reduced to 500 m and the maximum concentration of total suspended particulates (TSP) for a 12-h average recorded at urban industrial area, Petaling Jaya was higher than 500 $\mu g\,m^{-3}$ (Soleiman et al., 2003). In 1997, a severe haze episode was reported by Keywood et al. (2003), Abas et al. (2004a), Mott et al. (2005) and Mahmud (2009). The 24-h average concentration of $PM_{10}$ recorded near to Kuala Lumpur city centre exceeded 300 $\mu g\,m^{-3}$ (Abas et al., 2004a). The Malaysia Air Pollutant Index (API) recorded this year exceeded the very unhealthy level of 350 in September (Mahmud, 2009), and at its most severe visibility was reduced to only 500 m (Afroz et al., 2003). The concentration of $PM_{2.5}$ (62.1 $\mu g\,m^{-3}$) was higher than the United States Environmental Protection Agency (USEPA) suggested standard value for a 24-h $PM_{2.5}$ average (35 $\mu g\,m^{-3}$) (Pinto et al., 1998). Another severe haze episode was recorded in 2005 (Norela et al., 2013; Sahani et al, 2014). This episode occurred mainly on the central west coast of the Peninsula. After this year, haze occurred occasionally almost every year during the dry season between June to September. The latest severe and long haze episode in SEA, including Malaysia, was recorded in September 2015 (Huijnen et al., 2016). The highest concentration of $PM_{2.5}$ for a 24-h average during the haze episode in 2015 was 136 $\mu g\,m^{-3}$, compared to between 14.3 and 24.5 $\mu g\,m^{-3}$ during non-haze episodes (Sulong et al., 2017).

Since 1997, the continuous air quality monitoring network has recorded the level of $PM_{10}$ and other major air pollutants during haze and non-haze episodes in Malaysia. An example of long-term air quality data for PM, CO, $NO_2$ and $SO_2$ are presented from four stations: at Shah Alam on the west coast of Peninsular Malaysia; Kuching and Kota Kinabalu (two stations in Malaysian Borneo) and Jerantut (a background-level station located in the middle of Peninsular Malaysia). The levels of $PM_{10}$ for a 24-h average recorded at these stations during haze episodes exceeded 100 $\mu g\,m^{-3}$, compared to around 40 $\mu g\,m^{-3}$ during non-haze episodes (Fig. 1). Other than $PM_{10}$, the concentrations of CO were compared to days without haze episodes. Other gases do not indicate any differences during haze and non-haze episodes which suggests the influence of local sources on these pollutants (Azmi et al., 2010). Further analysis of overall $PM_{10}$ and CO concentrations over Malaysia during haze and non-haze events is presented in Fig. 2. The total active fire counts are also compared for the period between June and September when haze episodes typically occur. The total active fire counts obtained through the spatial interpolation of the Kernel density estimation during haze episode showed a domination of fire hotspots in Sumatra and Southern Borneo. The wind directions indicate movement towards Peninsular Malaysia, contributing to the severe haze episodes in Malaysia.

## 3. Sources of haze

### 3.1. Peat combustion

The peatland area in SEA covers about 26 million hectares and is located mostly near the coast (Wosten et al., 2008). The last several decades have seen the exploitation of peatland areas where large-scale deforestation and drainage have been carried out (Page et al., 2009). This disturbance has led to fires in peatlands, which have been the main source of the haze episodes in SEA. According to Harrison et al. (2009), the main cause of peatland fires in SEA was illegal land clearing activity with the use of fire. Often, the fire will become out of control and then spread to large areas. Degraded peatlands are known to be susceptible to fire (Wosten et al., 2008). According to Usup et al. (2000), high organic matter in peatland, as either decomposed material or material continuing to decompose, is the factor that makes peatland prone to fire. The exploitation of peatlands also changes the water table in these areas, and lowering the water table increases the frequencies and extent of peat fires (Evers et al., 2017; Turetsky et al., 2015).

Peat is flammable in dry conditions and when it burns, smouldering combustion will take place (Budisulistiorini et al., 2017; Zaccone et al., 2014). According to Rein et al. (2008), smouldering combustion is a process when an organic soil like peat burns steadily without flames and the burning slowly permeates into the soil. Smouldering combustion can last for long periods, which can span from a week to a month under low temperature, high moisture content and low oxygen concentration conditions (Turetsky et al., 2015). Sometimes, peat fires cannot even be detected when they smoulder deep underground, and can burn again during the next dry period making them hard to extinguish (Blake et al., 2009; See et al., 2007). Grishin et al. (2007) found that the temperature of combustion in deep layers of peat was higher than on its surface, where the moisture content, botanical composition

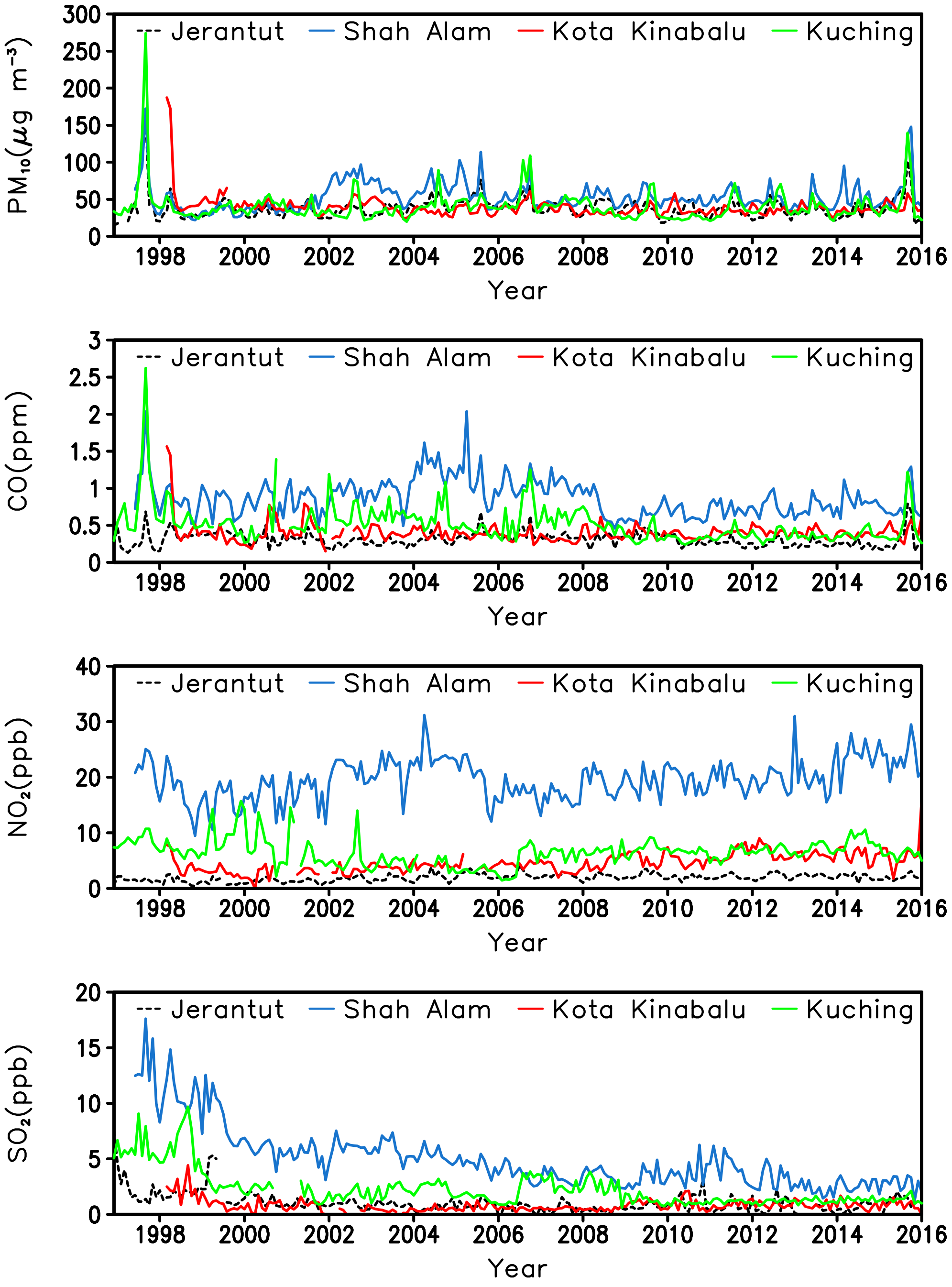

**Fig. 1.** Concentration trend of PM$_{10}$, CO, NO$_2$ and SO$_2$ in Shah Alam, Jerantut, Kota Kinabalu and Kuching from year 1997 to 2015.

and density are the main factors for smouldering combustion. The resulting smoke from peat combustion also depends on the heat intensity, aeration and duration of smouldering and flaming conditions (Abas et al., 2004a; Simoneit et al., 1999).

Several studies indicated that smouldering combustion of peat soils emits high concentrations of particles and gases to ambient air (Budisulistiorini et al., 2017; Fujii et al., 2014; Kuwata et al., 2017). A study by Othman and Latif (2013) on the concentration of major air pollutants from small-scale peat combustion showed that the burning of peat produces high amounts of PM and gases. The results showed that the dominant gas in the emissions from burning peat soil was CO (13,850–20,610 $\mu g\,m^{-3}$) followed by $NO_2$ (608–831 $\mu g\,m^{-3}$) and $SO_2$ (113–367 $\mu g\,m^{-3}$). High $PM_{10}$ concentration was released from 1-h peat soil combustion with a range of 778–3,444 $\mu g\,m^{-3}$. Organic substances such as levoglucosan (1,6-anhidro-β-D-glucopyranose, LG) were found to be good indicators for peat combustion at a range of 1.44–3.90 $\mu mol\,m^{-3}$. A detailed study by Stockwell et al. (2016) on emission factors (EF) from peat combustion collected in peat fires in tropical areas (Central Kalimantan, Indonesia) showed that $CO_2$ (1,564 ± 77 $g\,kg^{-1}$) was the major gas emitted from peat combustion. Other dominant major trace gas emissions included CO (291 ± 49 $g\,kg^{-1}$), methane (9.51 ± 4.74 $g\,kg^{-1}$), hydrogen cyanide (5.75 ± 1.60 $g\,kg^{-1}$), acetic acid (3.89 ± 1.65 $g\,kg^{-1}$), ammonia (2.86 ± 1.00 $g\,kg^{-1}$) and methanol (2.14 ± 1.22 $g\,kg^{-1}$). A study by Ramadhan et al. (2017) highlighted the potential for the combustion of peat and other biomass types to lead to the formation of chromophores such as oxygenated-conjugated compounds and nitroaromatics. These compounds could impact organic aerosol aging and growth through photochemical oxidation (Budisulistiorini et al., 2017). Even though there are many studies on peat emissions and the quantification of major air pollutants from peat combustion, most studies were conducted on a laboratory scale and further detailed confirmation is needed from field studies.

### 3.2. Agricultural activities

"Slash and burn" is a common agricultural practice in several countries in SEA and can be described as a cheap land clearing technique for agricultural development purposes that has been used traditionally (Nganje et al., 2001; Varma, 2003). This technique involves the process of cutting vegetation and setting it alight (Varma, 2003). Cotton (1999) and Jones (2006), have identified and described groups of people who practise "slash and burn" for agricultural purposes. These groups of people include traditional cultivators, small-scale cultivators

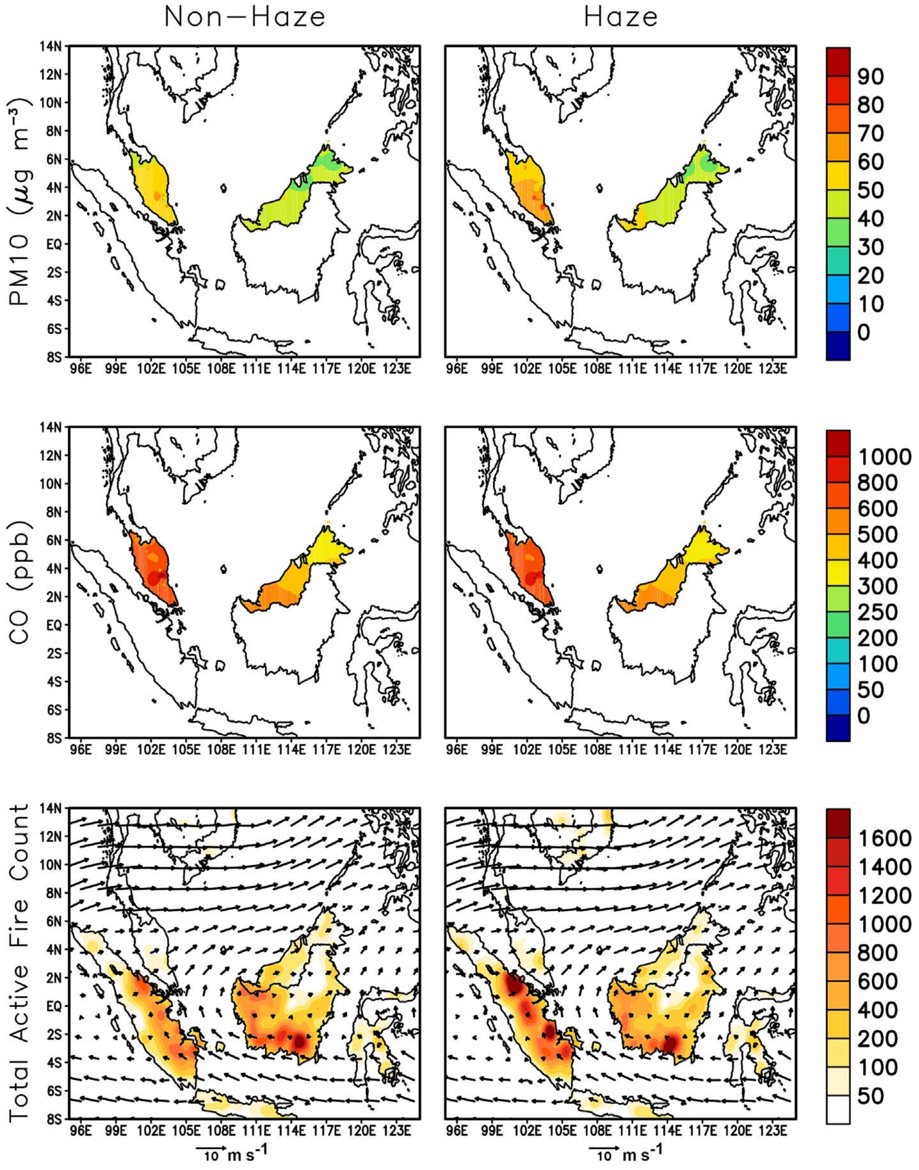

**Fig. 2.** Comparison between the annual mean of inverse distance weighting interpolated $PM_{10}$ and CO, kernel density of annual total active fire counts, and 950 hPa circulation pattern during June to September for non-haze and haze years.

and large-scale investors. Traditional cultivators usually clear small plots of land for planting every two or more years (Tacconi and Vayda, 2006), while the other groups use the technique to clear the land or forest for larger-scale agricultural purposes.

The "slash and burn" technique becomes a serious problem when large areas of forest perish due to a lack of education among cultivators about cropping techniques and the economic aspects of such fire use. In Indonesia "slash and burn" is one of the causes of fires originating from agricultural and industrial plantations (Saharjo and Munoz, 2005). Referring to Vogl and Ryder (1969), the process of "slash and burn" can alter the physical structure of the soil, affecting the density and growth of trees due to high temperature burning and additional ash and charcoal. The damage could persist for 15 years or longer. A study by Saharjo and Munoz (2005) on the behaviour and characteristics of fire in land preparation by traditional cultivators had found high fire flame temperature and intensity due to high fuel load. The authors suggested an alternative method of controlled burning as the best technique for land preparation.

The process of land clearing, especially over large areas, can lead to deforestation. Deforestation of peatland areas is mainly due to the high demand for palm oil and pulpwood. This demand encourages the opening of forests for plantations of these two crops on a massive scale in Indonesia and Malaysia (Abood et al., 2015; Busch et al., 2015; Murdiyarso et al., 2010). According to Saharjo and Munoz (2005), the plantation process includes the logging of natural forests, especially peatland areas, which opens the canopy of the forest and dries out the ground cover. Peatlands that have experienced drainage as a result of conversion into plantations can very easily be affected by fire, especially during the dry season.

### *3.3. Non-agriculture sources*

Haze in certain areas is also related to anthropogenic activities including transportation, industry and local BB. According to Du et al. (2011), haze formation depends on both aerosol composition and trace gases influenced by atmospheric chemical processes. Other than particles, sources of trace gases such as CO, NOx and $SO_2$ were recorded as higher during haze episodes compared to days without hazy conditions, especially in the Klang Valley (Azmi et al., 2010). The formation of secondary aerosol via gas-to-particle conversion under suitable meteorological conditions will further deteriorate the air quality during a haze episode (Behera et al., 2015b; Ram et al., 2012). Secondary photochemical particle production also occurs when gases from vehicles, industrial activities and other emissions undergo gas-to-particle conversion producing low-vapour-pressure products (Keywood et al., 2003).

## 4. El Niño - Southern Oscillation (ENSO) and haze events

Malaysian climatology is largely dominated by the Asian-Australian monsoon with two opposite monsoon regimes i.e. the boreal winter monsoon (northeast monsoon, NEM) and the boreal summer monsoon (southwest monsoon, SWM) (Tangang et al., 2012). The NEM usually falls within December to February and is the rainy period in Malaysia, while the SWM usually falls within June to September and is the dry period (Tangang et al., 2017). In addition, year-to-year climate variations are largely influenced by the phases of the El Niño - Southern Oscillation (ENSO) phenomenon (Juneng and Tangang, 2005; Salimun et al., 2014; Tangang et al., 2017; Tangang and Juneng, 2004) with a periodicity of 2–7 years. During El Niño (the warm phase), Malaysia as well as other regions of SEA, tends to experience prolonged drought, while during La Niña (the cold phase) the region tends to be impacted by severe floods (Juneng and Tangang, 2005; Tangang and Juneng, 2004).

An El Niño event usually develops in spring, evolves and peaks in winter, and its signature does not remain static over the entire SEA during its evolution. The associated rainfall anomaly can show considerable spatial and temporal variations (Aldrian and Susanto, 2003; Chang et al., 2003; Juneng and Tangang, 2005; Tangang et al., 2017; Tangang and Juneng, 2004). However, the anomalous conditions associated with El Niño do not only depend on the strength and intensity of El Niño but also the anomalous regional sea surface temperature in the Southeast Asian seas (Tangang et al., 2017). In general, the rainfall anomaly patterns show a north-eastward propagation of the drought-prone area from the initial El Niño state to its termination. During the early stage of El Niño in the summer months, almost the entire Maritime Continental region experiences drier-than-normal conditions, particularly over Sumatra and Kalimantan. These dry conditions, coupled with the warm temperatures associated with El Niño (Tangang et al., 2007), create an extremely favourable and conducive environment for large-scale fire outbreaks in Sumatra and Kalimantan (Reid et al., 2012; Tangang et al., 2012). Anomalous winds during June to August are largely southerly, enhancing the climatological summer monsoon winds, and this facilitates the long-range transport of smoke from Sumatra and Kalimantan northward to Singapore, Peninsular Malaysia, Sarawak, Brunei and Sabah. During the September to November period, the southern parts of Sumatra and Kalimantan continue to experience a deficit in rainfall, with affected areas extending to the entire Borneo region. Fires can become very active during this period of time over southern Sumatra and Kalimantan. This condition continues until the mature phase of El Niño. The occurrence of haze in northern Sarawak, Brunei and Sabah in 1998 was related to local fires associated with dry conditions in this region (Radojevic, 2003). By the March to May period, after the event's peak, only the northern tip of Borneo was anomalously drier. During this period, the conditions in Peninsular Malaysia, Sumatra and Kalimantan returned to normal or slightly below normal, minimizing the risk of fire outbreaks in this region. During February to April, while Sabah and northern parts of Sarawak tend to experience much drier conditions during a strong El Niño, the areas in southwestern parts of Sarawak, including the western region of Kalimantan, experience anomalously wet conditions. In fact, during February 2016, floods occurred in the Kuching and Pontianak areas, while in Sabah and northern Sarawak drought was severe.

During the period of June to November in an El Niño year, the risk of uncontrolled forest fires in Sumatra and Kalimantan increases because of the drier conditions induced by the El Niño. This typical evolution of the ENSO signature in rainfall and atmospheric circulation provides useful meteorological information for long-range forecasts. Juneng and Tangang (2008) showed that precipitation anomalies in the region can be forecasted at least five months in advance using sea-surface temperatures in the tropical Pacific as predictors. Given the fact that an El Niño is a predictable event at least six months in advance (Latif, 1998; Tangang et al., 1998), regional climate forecast information is invaluable in mitigating the risk of forest fires. Fig. 3 shows the difference of the composite of June-September hotspot counts, rainfall and the 850 hPa winds between the El Niño years (2002, 2004, 2006, 2009, 2015) and non-El Niño years (2003, 2005, 2007, 2008, 2010, 2011, 2012, 2013, and 2014). The hotspot data was available after 2002. Hence, the analysis only takes into account the years after 2002. The difference in the hotspots numbers (Fig. 3a) suggested strong ENSO modulation of the fire activities over the maritime continent, particularly over Sumatra and the southern part of Borneo Island, Kalimantan. Generally, during the El Niño dry period, the number of detected hotspots could be 3 to 4 times more than the number detected during the non-El Niño years. This is due to the reason that El Niño typically induces negative rainfall anomalies over the region south of the equator during the summer monsoon (Fig. 3b), particularly over the southern regions of Sumatra and Kalimantan. The wind anomalies field suggested a much stronger northerly cross-equatorial flow during El Niño years. This enabled more effective northward transport of the air mass toward the Malaysian regions, including the Peninsula and the states of Sarawak and Sabah.

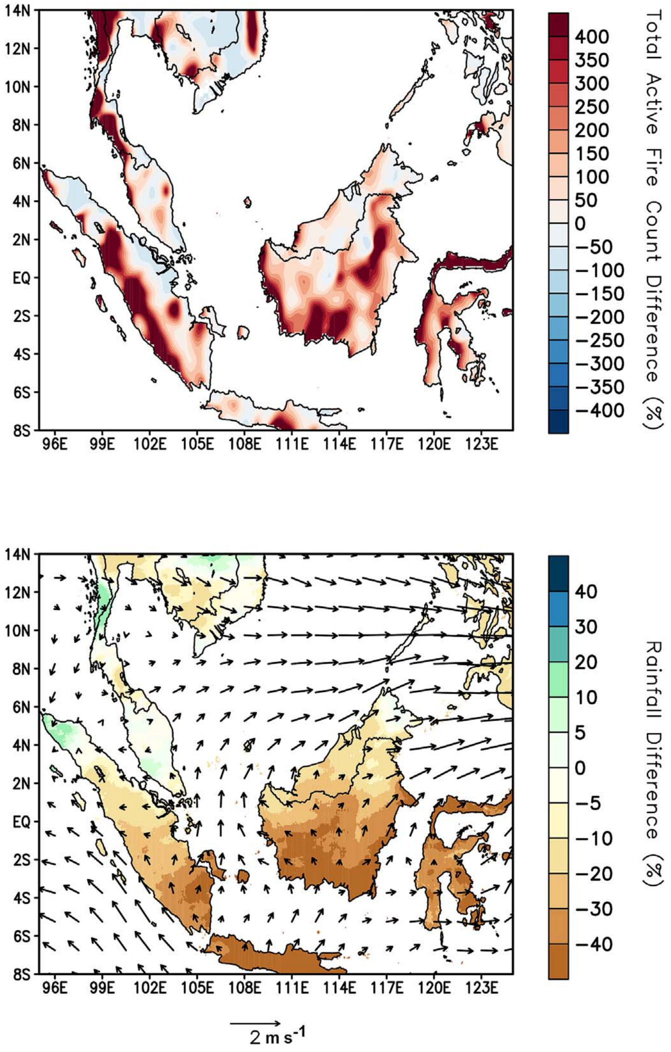

**Fig. 3.** Composite of June-September hotspot counts, rainfall and the 850 hPa winds between the El Niño (2002, 2004, 2006, 2009, 2015) and non-El Niño years (2003, 2005, 2007, 2008, 2010, 2011, 2012, 2013, and 2014).

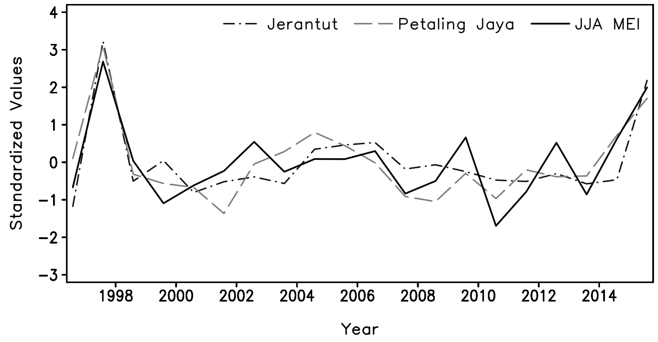

**Fig. 4.** The standardized time series of the defined annual haze index calculated for Jerantut and Petaling Jaya as well as the Multivariate ENSO Index during JJA period.

A strong El Niño modulates regional circulation and leads to prolonged dry conditions in SEA, especially in Sumatra and Kalimantan. Although the main source of the haze - forest burning - is anthropogenic, the transport and extension of the haze has to be modulated by the ENSO variations that control the regional surface circulation and climatic conditions (Reid et al., 2012). Although large-scale fires in Indonesia have occurred throughout paleo-history, their frequency before the 1960s was relatively rare (Field et al., 2009). Since the early 1960s, these events have occurred more frequently across the Maritime Continent, particularly in the southern region of Kalimantan and eastern Sumatra, due to increased land use activities in the region (Field et al., 2009; Spessa et al., 2015). These episodes are nearly always associated with El Niño events. As discussed earlier, El Niño tends to cause prolonged drought episodes over these regions during the developing phase (June to November). In addition to increasing the risk of uncontrolled forest fires, rainfall deficiency also prolongs the atmospheric residence time of fire products as they are less affected by precipitation (Heil and Goldammer, 2001). This situation often exacerbates the anthropogenic forest fires and causes widespread distribution of smoke over SEA. Although we expect the role of El Niño to be secondary since the fires are associated primarily with human-related activities in the agriculture, forestry and plantation sectors (Field et al., 2009), it plays an important role in altering the regional atmospheric compositions via the modification of the atmospheric meteorological field (Inness et al., 2015) as well as the emission and transport characteristics.

The influence of El Niño on the spatial distribution of haze episodes, especially the transport patterns, has been extensively studied since the disastrous haze event during the 1997/98 El Niño. A recent general overview of climate variability and its impact on the SEA haze episodes were reported in Tangang et al. (2010). Reid et al. (2012) provided an overview of fire hotspot activity over the Maritime Continent and how these fire activities are related to atmospheric oscillations of different time scales. They concluded that El Niño was the dominant factor that promoted fire activities over the region.

To further demonstrate the association between the local annual haze conditions in Malaysia and the ENSO, a local annual haze index was constructed. The annual index was calculated as the 95th percentile values of the daily $PM_{10}$ concentration for each of the years. The inter-annual variations of the constructed time series for two local stations (Jerantut and Petaling Jaya) are shown in Fig. 4. For comparison, the multivariate ENSO index (MEI) averaged over the June to August period is also depicted. It is noted that the year-to-year variability of local $PM_{10}$ was strongly associated with ENSO, with correlations values of 0.71–0.87. The correlation analysis of the annual time-series and MEI suggested that ENSO explains 50-75% of the local $PM_{10}$ inter-annual variance. To further illustrate the relationship between the $PM_{10}$ fluctuations and ENSO, the annual API recorded at Petaling Jaya's DOE station was correlated to the sea surface temperature to create the correlation coefficient map (Fig. 5). The sea surface temperature used in this comparison was the Optimum Interpolation (OI) Sea Surface Temperature (SST) version 2 obtained from the National Oceanic and Atmospheric Administration (NOAA), Earth System Research Laboratory (ESRL), Physical Sciences Division (PSD). The correlation maps show typical El Niño patterns with warm sea surface temperatures extending from the eastern Pacific and dominating the equatorial Pacific. The daily pattern of $PM_{10}$ at selected DOE stations in Malaysia is illustrated in Fig. 6 as one indicator of haze episodes during El Niño and non-El Niño years. This figure shows that in cases such as 1997 and 2015 El Niño influenced the high concentration of $PM_{10}$ but there are also several occasions, such as in 2005 and 2013, where high $PM_{10}$ that lead to a haze episode occurred in a non-El Niño year.

The results illustrate the importance of ENSO in modulating the year-to-year characteristics of haze in Malaysia. Note that 25-30% of the variation was due to other factors. It is clear that at shorter time scales (periods of a few days to weeks) the haze occurrence in Malaysia depended largely on anthropogenic burning in the region. While the large-scale fire activities over the region were strongly influenced by El Niño, the by-product transport pattern was also strongly tied to low-level circulation modulated by El Niño. The cross-equatorial flow generally transports the smoke north-westward south of the equator and north-eastward north of the equator. During the October-November transitional period, smoke transport paths are more zonally oriented compared to June-September (Xian et al., 2013). Using a numerical

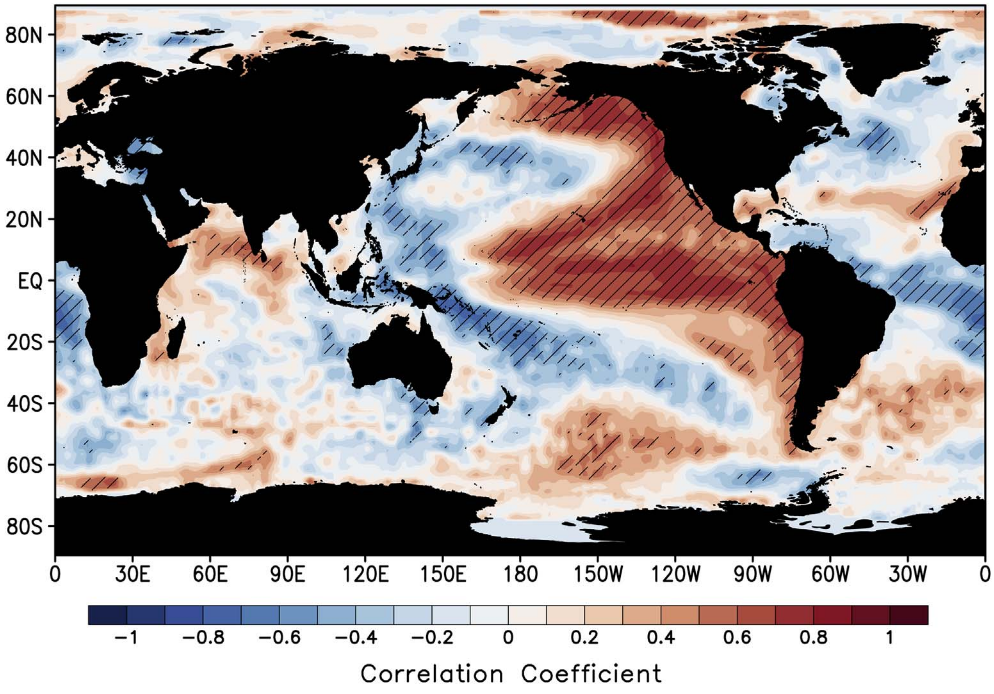

**Fig. 5.** The correlation coefficients between Petaling Jaya API time series and the global gridded mean SST for the period of June to August (1996-2015) at every grid point. Hatches signify the correlation between the API and the SST grid is significant at 0.05.

modelling experiment for the 2006 (El Niño) and 2007 (normal) SEA fire seasons, Xian et al. (2013) concluded that smoke typically lives longer and can be transported farther in El Niño years compared with non-El Niño years. During El Niño periods, due to stronger easterly winds in the region, there is a much stronger westward transport to the eastern tropical Indian Ocean.

## 5. Chemical properties of PM during haze episodes

PM, in the solid or liquid state, suspended in ambient air is a dominant pollutant. It acts as a precursor of numerous inorganic, organic, as well as microbial compounds and is usually measured during haze episodes. The composition of PM can be useful in determining the potential sources of air pollutants during haze episodes as well as the impact of haze on human beings and environments. The composition of PM during haze episodes can be separated into inorganic and organic compositions.

### *5.1. Inorganic composition of PM*

Concentrations of inorganic compositions in fine PM ($PM_{2.5}$) during haze and non-haze episodes in Malaysia are shown in Table 1. A study by Pinto et al. (1998) is recognised as one of the early studies reporting the key inorganic and organic compositions in $PM_{2.5}$ samples collected at Petaling Jaya, near to Kuala Lumpur city centre. The study recorded sulfur as the highest inorganic element with concentrations of 2400 ng $m^{-3}$ while other dominant elements were Si, K, and Fe. Currently there are only three specific studies in Malaysia concentrating on the composition of $PM_{2.5}$ during haze and non-haze days in Malaysia, by Amil et al. (2016), Fujii et al. (2015) (at Petaling Jaya) and Sulong et al. (2017) (at Kuala Lumpur city centre). Amil et al. (2016) found that the concentrations of secondary inorganic aerosols (SIA) such as $SO_4^{2-}$ and $NH_4^+$ were higher during haze episodes in 2011 and 2012 compared to non-haze days recorded in the same year. This finding was followed by findings from Sulong et al. (2017) for samples collected during severe haze episode in 2015. $K^+$ was found as a good indicator of BB due to its high concentration during haze episodes compared to non-haze days. Both studies emphasized the contribution of local anthropogenic sources during haze episodes recorded in the urban environment. Fujii et al. (2015), who studied elemental carbon (EC) during haze and non-haze days, indicated that there was no significant difference between the two conditions.

Other studies during haze episodes, such as those by Khan et al. (2016b) and Jaafar et al. (2017), also showed that the concentrations of SIA such as $SO_4^{2-}$ dominated the concentration of $PM_{2.5}$. Other individual studies during non-haze days, such as by Tahir et al. (2013) and Ee-Ling et al. (2015), also indicated the same pattern. The concentration of $SO_4^{2-}$ can be contributed by the photochemical oxidation of $SO_2$ emitted from various different sources including motor vehicles, industrial activities, and power plant emissions (Amil et al., 2016; Keywood et al., 2003). A study by Keywood et al. (2003) during haze episodes at selected stations within the Klang Valley area also suggested that local sources may also contribute to the amount of $SO_4^{2-}$ during haze episodes in Malaysia. At the same time $NO_2$, particularly from motor vehicles in urban areas, can contribute to the amount of $NO_3^-$ which is also one of the main components of SIA in $PM_{2.5}$ (Du et al., 2011).

Trace metal concentrations in $PM_{2.5}$ during haze episodes were first reported by Pinto et al. (1998). The study revealed Fe, Pb and Zn as the major heavy metals recorded during haze episodes. A study on trace metals concentrations in $PM_{2.5}$ by Amil et al. (2016) during haze episodes showed that Fe, V and Zn were among the major heavy metals, recorded at higher concentrations compared to other trace metals. A recent study by Sulong et al. (2017) showed that Fe, Cr and Zn were the major heavy metals during haze episodes. According to Betha et al. (2014), trace metals during haze episodes originate from various sources during BB including peat smouldering, wood burning, incomplete combusted plant tissues, ash and resuspension of soil particles.

A comparison of inorganic concentrations during haze and non-haze episodes in SEA is listed in Table S1. Overall, SIA such as $SO_4^{2-}$ were recorded at higher concentrations in BB areas as shown by Pinto et al. (1998) at Palembang and Sriwijaya University, Indonesia; See et al. (2007) in Sumatra, Indonesia; Karthikeyan et al. (2007) in Singapore; Huang et al. (2013) in Myanmar, northern Thailand, Laos, parts of Vietnam and Cambodia; and Khamkaew et al. (2016) in Chiang Mai, Thailand. The concentrations of other pollutants including other SIA such as $NO_3^-$ had high concentrations, especially in the BB areas such

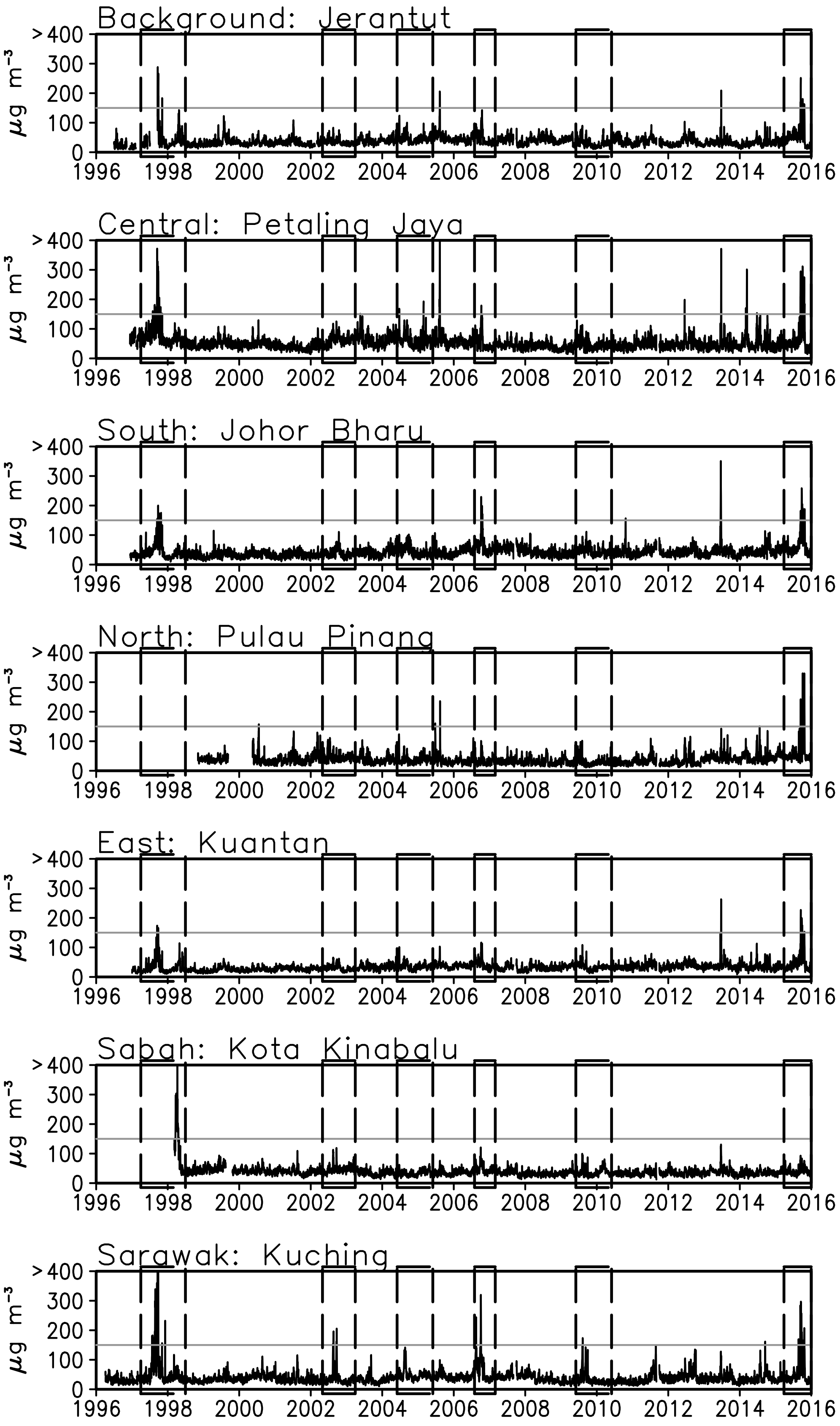

**Fig. 6.** $PM_{10}$ daily mean concentration from year 1996–2015 juxtaposed against El Niño periods (in boxes) for several stations according to the regions in Malaysia.

as Sumatra. Other trace metals, such as Fe, Al and Zn, were recorded at higher concentrations in $PM_{2.5}$ compared to other trace metals in BB areas (See et al., 2007). Different studies have shown that the concentrations of inorganic substances are influenced by different areas and land use, the type of BB, anthropogenic influences and other local factors (Lee et al., 2016). Further detailed sampling during haze and non-haze episodes in Singapore by See et al. (2006) showed that SIA ($SO_4^{2-}$, $NH_4^+$ and $NO_3^-$) and most trace metals, especially Al, Fe and

**Table 1**
Concentration of inorganic composition of $PM_{2.5}$ during haze and non-haze in Malaysia determined by different researchers.

| Reference | $SO_4^{2-}$ | $NO_3^-$ | $Cl^-$ | $NH_4^+$ | $Na^+$ | $K^+$ | $Ca^{2+}$ | Al | V | Cr |
|---|---|---|---|---|---|---|---|---|---|---|
| Unit | µg m$^{-3}$ | µg m$^{-3}$ | µg m$^{-3}$ | µg m$^{-3}$ | µg m$^{-3}$ | µg m$^{-3}$ | µg m$^{-3}$ | µg m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ |
| Haze | | | | | | | | | | |
| Pinto et al. (1998)<br>*(Petaling Jaya)* | 10.0 | | 0.07[a] | | | 0.20[a] | 0.098[a] | | 9.30 | 0.2 |
| Fujii et al. (2015)<br>*(Petaling Jaya)* | | | | | | | | | | |
| Amil et al. (2016)<br>*(Petaling Jaya)* | 2.40 | 0.21 | 0.030 | 2.21 | 0.23 | 0.51 | 0.28 | 2.78 | 1.02 | 2.87 |
| Khan et al. (2016b)<br>*(UKM Bangi)* | 1.98 | 0.28 | 0.023 | 0.54 | 0.14 | 0.21 | 0.20 | 0.011 | 1.31 | |
| Sulong et al. (2017)<br>*(Kuala Lumpur)* | 20.4 | 3.85 | 0.17 | 7.16 | 0.68 | 0.60 | 0.23 | 0.32 | 4.57 | |
| Jaafar et al. (2017)<br>*(UKM Bangi)* | 3.10 | 0.98 | 0.13 | | 0.27[a] | 0.61[a] | 0.12[a] | | | |
| Average<br>(range) | 7.58<br>(2.40–20.4) | 1.33<br>(0.21–3.85) | 0.08<br>(0.023–0.17) | 3.30<br>(0.54–7.16) | 0.33<br>(0.14–0.68) | 0.42<br>(0.20–0.61) | 0.18<br>(0.098–0.28) | 1.04<br>(0.011–2.78) | 4.05<br>(1.02–9.30) | 1.53<br>(0.20–2.87) |
| Non-haze | | | | | | | | | | |
| Tahir et al. (2013)<br>*(Kuala Terengganu)* | 3.80 | 0.31 | 0.16 | 1.11 | 0.45 | 0.62 | 0.13 | | | |
| Ee-Ling et al. (2015)<br>*(UKM Bangi)* | 3.52 | 0.26 | 0.05 | 0.76 | 0.46 | 0.27 | 0.10 | | | |
| Fujii et al. (2015)<br>*(Petaling Jaya)* | | | | | | | | | | |
| Amil et al. (2016)<br>*(Petaling Jaya)* | 1.33 | 0.21 | | 0.99 | 0.30 | 0.25 | 0.18 | 1.06 | | |
| Khan et al. (2016a)<br>*(UKM Bangi)* | 2.12 | 1.29 | 0.05 | 0.62 | 0.61 | 0.36 | 1.97 | 0.29 | 0.002 | 0.115 |
| Sulong et al. (2017)<br>*(Kuala Lumpur)* | 2.01 | 0.38 | | 0.28 | 0.27 | 0.11 | 0.18 | 0.24 | 0.002 | 0.010 |
| Average<br>(range) | 2.55<br>(1.33–3.80) | 0.49<br>(0.21–1.29) | 0.08<br>(0.05–0.16) | 0.75<br>(0.28–1.11) | 0.41<br>(0.27–0.61) | 0.32<br>(0.11–0.62) | 0.51<br>(0.10–1.97) | 0.53<br>(0.24–1.06) | 0.002<br>(0.002–0.002) | 0.06<br>(0.010–0.115) |

| Reference | Mn | Fe | Ni | Cu | Zn | As | Se | Pb | Cd |
|---|---|---|---|---|---|---|---|---|---|
| Unit | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ |
| Haze | | | | | | | | | |
| Pinto et al. (1998)<br>*(Petaling Jaya)* | 4.5 | 120 | 2.2 | 9.3 | 34.3 | 2.3 | 0.7 | 39 | |
| Fujii et al. (2015)<br>*(Petaling Jaya)* | | | | | | | | | |
| Amil et al. (2016)<br>*(Petaling Jaya)* | 1.39 | 1.90 | 0.59 | 3.49 | 175 | 0.337 | 0.207 | 94.0 | 0.13 |
| Khan et al. (2016b)<br>*(UKM Bangi)* | 1.19 | 5.67 | 0.73 | 2.46 | 15.8 | 3.29 | 0.37 | 4.54 | 0.21 |
| Sulong et al. (2017)<br>*(Kuala Lumpur)* | 11.6 | 129 | 5.54 | 8.34 | 19.0 | 1.11 | | 6.90 | 0.22 |
| Jaafar et al. (2017)<br>*(UKM Bangi)* | | | | | | | | | |

(continued on next page)

**Table 1** (*continued*)

| Reference | Mn | Fe | Ni | Cu | Zn | As | Se | Pb | Cd |
|---|---|---|---|---|---|---|---|---|---|
| Unit | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ | ng m$^{-3}$ |
| Average<br>(range) | 4.67<br>(1.19–11.6) | 64.1<br>(1.90–129) | 2.26<br>(0.59–5.54) | 5.89<br>(2.46–9.30) | 61.0<br>(15.8–175) | 1.75<br>(0.337–3.29) | 0.42<br>(0.207–0.7) | 36.1<br>(4.54–94.0) | 0.18<br>(0.13–0.22) |
| Non-haze | | | | | | | | | |
| Tahir et al. (2013)<br>(*Kuala Terengganu*) | | | | | | | | | |
| Ee-Ling et al. (2015)<br>(*UKM Bangi*) | 1.39 | 35.0 | 2.22 | 1.92 | 51.2 | | | | 0.06 |
| Fujii et al. (2015)<br>(*Petaling Jaya*) | | | | | | | | | |
| Amil et al. (2016)<br>(*Petaling Jaya*) | | 1.00 | | | 0.19 | | | | |
| Khan et al. (2016a)<br>(*UKM Bangi*) | 0.004 | 3.32 | 0.014 | 0.026 | 0.032 | 0.006 | 0.0007 | 0.018 | 0.0005 |
| Sulong et al. (2017)<br>(*Kuala Lumpur*) | 0.003 | 0.11 | 0.001 | 0.009 | 0.018 | 0.0004 | | 0.005 | 0.0001 |
| Average<br>(range) | 0.46<br>(0.003–1.39) | 9.85<br>(0.11–35.0) | 0.74<br>(0.001–2.22) | 0.65<br>(0.009–1.92) | 12.8<br>(0.018–51.2) | 0.0032<br>(0.0004–0.006) | 0.0007 | 0.01<br>(0.005–0.018) | 0.02<br>(0.0001–0.06) |

[a] Concentration represents major element concentration.

Zn, were recorded at higher concentrations during haze days compared to non-haze days. $K^+$ is a good marker of BB based on its concentrations in $PM_{2.5}$ recorded during BB days in SEA (Lin et al., 2013). Similar results were also recorded by Engling et al. (2014) and Huang et al. (2016).

### 5.2. Organic composition of PM

Numerous studies, predominantly laboratory-based, have contributed to the current knowledge of the organic products of non-fossil-biomass combustion (Balasubramanian et al., 2003; Jones et al., 2005; Simoneit et al., 2004a, 2004b). These studies found that organic material constitutes more than 50% of the compositions resulting from BB and the compounds with higher molecular weights, ranging from $C_{25}$ to $C_{36}$, dominate (Vasconcellos et al., 2010). The highest organic carbon (OC) concentration during haze episodes was recorded by Pinto et al. (1998) with a concentration of 26.1 μg m$^{-3}$ followed by Fujii et al. (2015) with a concentration of 10.0 μg m$^{-3}$. The two studies recorded the ratios of organic to elemental carbon (OC/EC) values of 13.7 and 3.33, both exceeding the value of 2.00 which indicates the occurrence of SOA (Chow et al., 1996).

Vegetation is the major fuel consumed in BB and is composed predominantly of cellulose, hemicellulose and lignin. Together, these three polymeric materials account for over 90% of the dry weight of most vascular plants, with the remaining mass composed of various lipids, proteins, and other metabolites, as well as minerals and water. The combustion of the organic components of biomass involves a complex series of physical transformations and chemical reactions including pyrolysis, depolymerization, water elimination, fragmentation, oxidation, char formation and volatilization (Graham et al., 2002; Shafizadeh, 1984).

The concentrations of PAHs and selected BB markers of PM (TSP, $PM_{10}$ and $PM_{2.5}$) during haze and non-haze episodes in Malaysia are shown in Table 2. A study of PAH concentrations in $PM_{2.5}$ during haze episodes in Petaling Jaya, Malaysia for 1997 was carried out by Pinto et al. (1998), where the total PAH concentration was 135.0 ng m$^{-3}$. A detailed study by Okuda et al. (2002) on PAHs in TSP collected in Peninsular Malaysia during a haze episode in 1997 showed that the concentration of total PAHs reached 12.5 ng m$^{-3}$ compared to 4.1 ng m$^{-3}$ during non-haze days. Based on isotopic compositions of PAHs, this study suggested that the molecular and isotopic signatures of PAHs recorded during haze episodes was similar to PAHs emitted from automotive exhausts. Quantitative estimation showed that the wood burning contribution to PAHs ranged from 25% to 35% with no relation to haze intensity, while automotive contribution ranged from 65% to 75%. Omar et al. (2006) compared concentrations of PAHs during haze and non-haze episodes in Kuala Lumpur, where significantly higher concentrations of total PAHs were recorded during haze episodes compared to non-haze episodes. This study found that the total benzo[a]pyrene (B[a]P) equivalent value during the haze episode in 2001 was four times higher than in street level particles. A recent study by Khan et al. (2015) showed a total PAH concentration of 3.85 ng m$^{-3}$ during the southeast monsoon and haze episodes. This result is higher compared to the previous studies by Bahry et al. (2009) and Jamhari et al. (2014) during non-haze episodes, with total PAHs of 2.73 ng m$^{-3}$ and 2.27 ng m$^{-3}$, respectively. The high molecular weight (HMW) PAHs (5 ring) were significantly prominent ($>$ 70%) compared to the low molecular weight (LMW) PAHs (4 ring) in $PM_{2.5}$ during haze episodes (Khan et al., 2015). The production of HMW PAHs is facilitated in smouldering processes due to a lack of oxygen and this group was found to be an indicator group for peat combustion (Tsibart et al., 2014). According to Urbančok et al. (2017), another reason for the domination of HMW PAHs is that the lighter PAHs are more prone to degradation, especially in tropical environments. Degradation processes are expected to influence the amount of PM-bound low molecular weight PAHs due to the long-ranged transport from biomass burning areas during haze episodes.

**Table 2**
Concentration of elemental (EC) and organic carbon (OC) (in µg m$^{-3}$), selected polyaromatic hydrocarbons (PAHs) and burning biomarkers (in ng m$^{-3}$) of particulate matter ($PM_{2.5}$, $PM_{10}$ and TSP) during haze and non-haze in Malaysia determined by different researchers.

| Reference | EC | OC | NAP | ACY | ACP | FLR | PHE | ANT | FLT | PYR | B[a]A | CYR | B[k]F | B[a]P | B[b]F | I[c]P | D[h]A | B[g]P | Total PAHs | LG | MN | GLC | CHL | DA |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Haze | | | | | | | | | | | | | | | | | | | | | | | | |
| Pinto et al. (1998)<br>(Petaling Jaya***) | 1.9 | 26.1 | < 4.9 | 6.0 | 3.9 | 12.5 | 50.9 | 12.1 | 10.9 | 11.9 | < 4.1 | < 4.1 | < 4.0 | < 3.8 | < 6.5 | < 3.6 | < 4.4 | < 4.1 | 135 | | | | | |
| Okuda et al. (2002)<br>(Peninsular Malaysia*) | | | | | | | | | | | | | | | | | | | 12.5 | | | | | |
| Abas et al. (2004b)<br>(Kuala Lumpur*) | | | | | | | | | | | < 4.0 | | 5–10 | | < 10 | | | | | 40240 | | | 232 | 117 |
| Omar et al. (2006)<br>(Kuala Lumpur **) | | | | | | | | | | | 1.23 | | 6.13 | 0.05 | 8.66 | | | 14.1 | 47.8 | | | | | |
| Khan et al. (2015)<br>(Bangi ***) | | | 0.10 | 0.04 | 0.18 | 0.09 | 0.06 | 0.06 | 0.16 | 0.12 | 0.06 | 0.13 | 0.33 | 0.37 | | 0.51 | 0.22 | 0.81 | 3.85 | | | | | |
| Fujii et al. (2015)<br>(Petaling Jaya***) | 3.0 | 10.0 | | | | | | | | | | | | | | | | | | 160 | 8.4 | 2.3 | 1.8 | 1.7 |
| Jaafar et al. (2017)<br>(Bangi***) | | | | | | | | | | | | | | | | | | | | 834 | | | | |
| Non-haze | | | | | | | | | | | | | | | | | | | | | | | | |
| Abas and Simoneit (1996)<br>(Petaling Jaya*) | | | | | | | | | | | | | | | | | | | | 766 | | | 32 | 110 |
| Okuda et al. (2002)<br>(Peninsular Malaysia*) | | | | | | | | | | | | | | | | | | | 4.1 | | | | | |
| Omar et al. (2006)<br>(Kuala Lumpur**) | | | | | | | | | | 0.04 | | | 0.38 | 0.16 | 0.25 | | | 0.76 | 3.10 | | | | | |
| Bahry et al. (2009)<br>(Peninsular Malaysia*) | | | | | | | 0.09 | 0.007 | | 0.10 | | 0.09 | 0.83 | 0.27 | | | | | 2.73 | | | | | |
| Jamhari et al. (2014)<br>(Kuala Lumpur**) | | | n.d | n.d | n.d | n.d | 0.05 | 0.02 | 0.07 | 0.09 | 0.20 | 0.11 | 0.20 | 0.20 | 0.29 | 0.33 | 0.08 | 0.75 | 2.27 | | | | | |
| Fujii et al. (2015)<br>(Petaling Jaya***) | 3.4 | 5.2 | | | | | | | | | | | | | | | | | | 40 | 2.83 | 0.69 | 1.16 | 0.95 |

NAP = Napthalene, ACY = Acenapthelyne, ACP = Acenapthene, FLR = FLuorene, PHE = Phenanthrene, ANT = Anthracene, FLT = Fluoranthene, PYR = Pyrene, B[a]A = Benzo[a]anthracene, CYR = Chrysene, B[k]F = Benzo[k]fluoranthene, B[a]P = Benzo[a]pyrene, B[b]F = Benzo[b]fluoranthene, I[c]P = Indenol[1,2,3-cd]pyrene, D[h]A = Dibenzo[a,h]anthracene, B[g]P = Benzo[g,h,i]perylene, LG = Levoglucosan, MN = Mannosan, GLC = Galactosan, CHL = Cholesterol, DA = Dehydroabetic Acid.

n.d = not detected.

* = TSP samples, ** = $PM_{10}$ samples, *** = $PM_{2.5}$ samples.

Levoglucosan (LG) is a stable organic molecule and has been widely referred to as a biomarker for BB (Abas et al., 1995, 2004b). LG is formed through the thermal breakdown and alteration of the cellulose present in vegetation (Dos Santos et al., 2002). Due to its high stability, it shows no decay over an 8-h exposure to ambient conditions and sunlight (Larsen III et al., 2006; Puxbaum et al., 2007). Taking its stability and high abundance into account in ambient air, this molecule has been well recognised as an ideal marker for BB (Simoneit et al., 1999). A recent study by Fujii et al. (2015) in Petaling Jaya showed that in addition to LG, pyrolyzed organic carbon, mannosan (MN), galactosan (GLT), syringaldehyde, vanillic acid and cholesterol were also higher during the SWM. The composition of n-alkanes was dominated by high molecular weight alkanes with the maximum number of carbon atom values ($C_{max}$) ranging from 25 to 33, characteristic of biogenic sources (higher plant wax), whereas lower $C_{max}$ values may indicate major petrogenic input. Based on source apportionment analysis from this study, 60% of organic substances in $PM_{2.5}$ originated from BB from peat combustion and 8% from softwood and hardwood BB. A high concentration of LG was recorded by Abas et al. (2004b) with a concentration of 40,240 $ng\,m^{-3}$ during the severe haze episode in 1997, while other studies had $< 900\,ng\,m^{-3}$ concentrations during other haze episodes in Malaysia (Abas and Simoneit, 1996; Fujii et al., 2015; Jaafar et al., 2017) (Table 2).

Comparisons with other studies in SEA showed that PAHs were recorded at the highest concentrations in the BB areas (Table S2). This has been shown by Kunii et al. (2002) based on the measurement of PAHs in $PM_{10}$ collected in Jambi, Sumatra. Phenanthrene (PHE) was found to have the highest concentration of the PAHs during haze episodes as recorded by Pinto et al. (1998) and See et al. (2007) in Sumatra. Studies by Engling et al. (2014) in Singapore and Pongpiachan et al. (2017) in Thailand showed that the concentrations of PAHs were quite low and BB cannot be considered as the sole contributor of particulate PAHs. As reported by other studies, LG was found as the dominant organic marker during BB episodes (Khamkaew et al., 2016).

### 5.3. Chemical properties of rainwater

The concentration of PM usually influences the composition of rainwater. During the haze episode in 2005, the inorganic composition of rainwater measured in Petaling Jaya and Malacca, Malaysia by Norela et al. (2013) was dominated by $NO_3^-$, $SO_4^{2-}$ and $NH_4^+$ at both sites (Fig. 7). The study also revealed that rainwater pH values for both Petaling Jaya and Malacca were recorded below the pH of normal rainwater, indicating the contribution of haze to the acidity of the rainwater. High concentrations of $NO_3^-$ and $SO_4^{2-}$ influence the increase of acidity in rain water. In contrast, $NH_4^+$ neutralizes the acidity of the rainwater. A study by Balasubramanian et al. (1999) in Singapore recorded high concentrations of ions, *e.g.* $SO_4^{2-}$, $NO_3^-$, and $NH_4^+$, during the BB event and determined that the pH value of rain water ranged from 3.79 to 6.20. The author suggested that the chemical composition of precipitation trapped the BB gases and aerosols. However, the decrease in rainwater pH was marginal due to the neutralization of acid gases by $NH_3$ and $CaCO_3$. The emission of $SO_2$ and $NO_2$ from the combustion of BB can play a part in atmospheric chemistry during haze episodes. However, Radojevic and Tan (2000) and Radojevic (2003) observed that the emission of alkaline substances (*e.g.*, $Ca^{2+}$, $Mg^{2+}$, $K^+$) by forest fires can neutralize the acidic gases ($SO_2$ and $NO_2$) during haze episodes. Thus, there was no significant acidifying effect on rainwater during the forest fires in the SEA region. A subsequent study on rainwater chemistry conducted by Zhong et al. (2001) in Singapore reported that BB was an important source of HCOOH and $CH_3COOH$ to the troposphere over SEA. This study also observed that HCOOH was likely to be produced in the atmosphere in addition to its direct emission from biomass fire events. Water-soluble nutrients were also consistently higher during the haze period compared to non-haze in Singapore (Sundarambal et al., 2010). The chemical ionic compositions reported from the rainwater samples in Indonesia showed that the forest fires were a more dominant influence than anthropogenic activities on the increase in concentrations of the chemical compositions in rainwater in Kototabang, Palembang and Lampung of Indonesia (Budiwati et al., 2016).

Studies by Tay et al. (2014) and EANET (2013) in different areas of Peninsular Malaysia and Malaysian Borneo reported that the pH of rainwater ranged between 4.16 and 6.65. The lowest pH value was reported at a level of 4.16 in Petaling Jaya, in the greater Kuala Lumpur region. The pH of rainwater collected on the east coast of Peninsular Malaysia ranged between 5.35 and 6.65. In the remote rural location, the pH level was 5.01, and 5.43 in the Kuching in Borneo region. The spatial distribution of pH clearly indicated that land use is a considerable factor contributing to the change in pH of rainwater in Malaysia. A recent study by Khan et al. (2018) showed that the variation in the physicochemical nature of rainwater might be linked to the differences in land use at the two sites. The pH at the background rural site was compared to that at the urban site. The results showed that a higher level of pH was observed at the background site as compared to the urban environment. This study suggested that the anthropogenic input of acid precursors was high at the urban site but absent at the rural background site. Thus, the acidic gases less affected the rural site. This study also highlighted the change of acidity during haze and non-haze episodes at the two sites. During haze episodes, the pH was significantly decreased to 4.8 compared to non-haze value of 5.04 at the rural site. However, the average pH during SWM haze period was unchanged at the urban site (Khan et al., 2018).

## 6. Impact of haze

### 6.1. Health impacts

Smoke haze occurring in Malaysia and SEA contains high concentrations of air pollutants. Referring to Heil and Goldammer (2001), vegetation fires and BB produces high amounts of trace gases and PM. PM has been identified as one of the highest air pollutants during haze episodes. PM when in respirable range can contribute to mortality and serious illness (Kampa and Castanas, 2008), as fine particulates can easily enter into the respiratory system through inhalation.

Human health effects during haze episodes have been recorded based on increased hospital admissions, usually related to respiratory health problems during this time (Afroz et al., 2003; Betha et al., 2014; Sahani et al, 2014). According to Mott et al. (2005), hospital admissions during the 1997 haze episode in Kuching increased 8% compared to the average number of hospital admissions during the same month in the years 1995, 1996 and 1998. Moreover, the number of patients for chronic obstructive pulmonary diseases (COPD) also increased by about 42% for people aged 65 years and older compared to the number of patients for the same month in the years 1995, 1996 and 1998. The number of respiratory disease outpatient visits at Kuala Lumpur General Hospital also increased from 250 to 800 people per day during the 1997 haze episode (Afroz et al., 2003; WHO, 1998). Diseases that were directly affected by haze include upper respiratory tract infections, asthma and conjunctivitis (Awang, 2000; Emmanuel, 2000). Another study by Othman et al. (2014) determined that there was an increase of hospital admissions of 2.4 per 10,000 population each year during haze episodes in Malaysia, where children were the most affected group.

Another study on the health impacts of haze in Malaysia analysed the respiratory impact, mortality and impact of haze on specific ages. Wan Yaacob et al. (2016) performed a study on the impact of haze on the peak expiratory flow rate (PEFR) measurement of school children and the result showed that there was a 15% reduction in PEFR measurements, and 22 children with headache, cough, mucus, and sore throat symptoms had higher rates of five respiratory illnesses. Moreover, a study by Sahani et al. (2014) examined the risk of haze in the Klang Valley region between 2000 and 2007 where the results showed

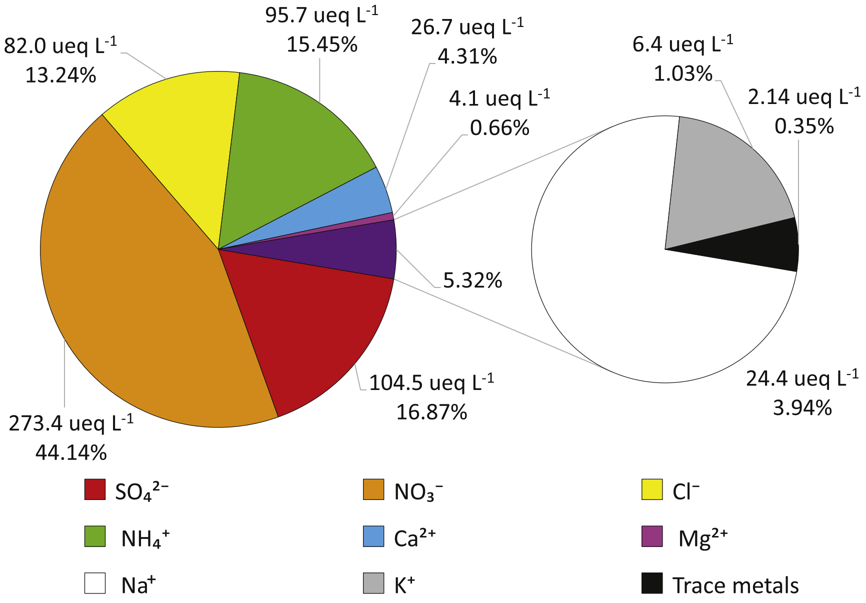

**Fig. 7.** Composition of major inorganic composition in rain water recorded in Klang Valley during the haze episode in 2005.

that the haze episode contributed to a 19% increase in respiratory mortality. The findings also showed that there was a 41.4% increase in the delayed effects of haze on the natural mortality of children and a 66% increase in the respiratory mortality of adult females. A study by Koplitz et al. (2016) for the 2015 haze episode joined the GEOS-Chem global chemistry model together with health response functions and estimated that excess deaths with 95% confidence intervals was 6,500 (1,700–11,300) in Malaysia. Another recent study by Sulong et al. (2017) showed that for non-carcinogenic health risk assessment, the infant group faced more significant health risks than the other age groups during haze episodes (Heath Index, HI = 1.06). On the other hand, for the carcinogenic health risk assessment, adult group was the most affected group for haze exposure (Excess Lifetime Cancer Risk, ECLR = $2.27 \times 10^{-5}$).

### 6.2. Economic impact and other impacts

The occurrence of haze and its related air quality degradation also impacted the economy of Malaysia, not to mention the country's agriculture and biodiversity. As reported by Ho et al. (2014), haze episodes caused reductions of output in both manufacturing and construction, and a reduction of tourism earnings, particularly due to flight cancellations. An estimation of the economic loss from the 1997 haze episode in SEA was about USD 4 billion (Tangang et al., 2010). A study by Varma (2003) revealed that the estimated economic loss of the “slash and burn” practices that caused the SEA forest fires in 1997/1998 was USD 20.1 billion. The estimates of the economic impact of haze in the year 1997 by Mohd Shahwahid and Othman (1999) included several aspects such as cost of illness, productivity loss, declines in tourist arrivals, flight cancellations, decline in fish landings, cost of fire-fighting, cloud seeding, and expenditure on masks (Fig. 8). The total damage costs were USD 321 million. A study by Othman et al. (2014) on economic loss due to inpatient costs during the haze episodes in the years 2005, 2006, 2008 and 2009 in Malaysia found losses of USD 91,000 annually with an average of USD 4,789 loss for each haze day.

Other impacts of haze include the reduction in plant yields due to the limitation of light levels. The aerosol radiative effect can be modulated by the vertical distribution and optical properties of aerosols (Pani et al., 2016a, 2016b; Wang et al., 2015). According to Yanhong et al. (1996), the reduction of light reaching the Earth's surface during haze can affect various ecosystems, especially forest ecosystems and plant crops. Illuminance reduction during haze can lower photosynthesis rates which reduces plant production (Yoneda et al., 2000). During the 1994 haze episode, two varieties of hybrid rice in Malaysia, MR151 and MR123, displayed a 50% reduction in growth rate and abnormal ripening, while in Indonesia, there was a 2-3% reduction of paddy rice yield. Chameides et al. (1999) found that during haze episodes in China, there was a 5-30% reduction of solar irradiance which contributed to a reduction of crop optimal yield of at least 5-30%. Wildlife is also affected during haze episodes due to the high amount of toxic smoke, with a high risk of mortality among animals that are very sensitive to changes in their surroundings. Schweithelm and Glover (1999) stated that, during the 1997/98 fires and haze episode, there were declining numbers of orangutans in East and Central Kalimantan due to habitat loss and forest fires with high amounts of smoke.

## 7. The way forward

This paper has attempted to identify the roles of both anthropogenic activities and climatological variability in influencing the regional atmospheric composition. Several severe haze episodes with $PM_{10}$ concentrations exceeding $100\,\mu g\,m^{-3}$ were recorded in 1997, 2005 and 2015 while less severe haze episodes were documented in between. Anthropogenic causes, primarily related to agricultural practices, and meteorological factors, particularly during El Niño events, have been attributed to uncontrollable fires from peatland areas in Sumatra and Kalimantan, Indonesia. Variability of local $PM_{10}$ as an indicator for haze episodes was strongly associated with ENSO, with correlations values of 0.71–0.87. Dry seasons, especially during the summer monsoon in SEA, and wind directions also influence the intensity of haze episodes. The compositions of inorganic and organic substances in PM have been identified as markers for emissions from BB that contribute to the haze episodes in SEA. Overall, concentrations of several inorganic ($K^+$, $SO_4^{2-}$, $NH_4^+$, Zn, Al, Fe) and organic (LG) compounds shows substantial differences between haze and non-haze episodes. The composition of alkaline and acid base substances in different areas were found to influence the pH of rainwater during haze episodes. Haze was found to severely affect human health and it has been associated with an increased number of respiratory illnesses, and with increased cancer risk and mortality. Children and senior citizens were the most affected groups. Haze also affected the Malaysian economy due to the costs associated with illness, productivity loss, declines in tourist arrivals,

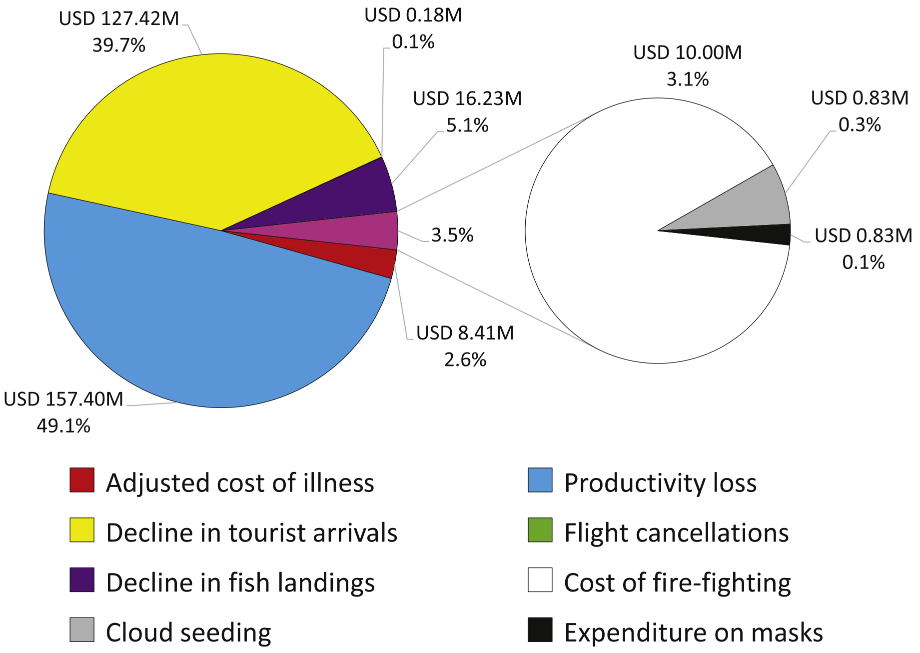

**Fig. 8.** Aggregate value of haze damage in 1997 as suggested by Mohd Shahwahid and Othman (1999).

flight cancellations, decline in fish landings, cost of fire-fighting, cloud seeding, and expenditure on masks. The inpatient costs during haze episodes were estimated at around USD 4,789 per day. Prevention and mitigation measures should be the priority in future research endeavours to reduce haze episodes. Among the steps taken on a regional level is the Association of Southeast Asian Nations (ASEAN) Agreement on Transboundary Haze Pollution that has been ratified by all ASEAN member countries. The agreement includes the requirement that all member countries cooperate in the development and implementation of mitigation measures such as assessment and early warning systems. In this section, several areas for better haze mitigation both within and outside of the legally binding agreement is discussed.

Reducing response time is crucial in any mitigation effort taken to reduce the costs highlighted in this paper. An increase in forecasting ability should be a priority as it can increase the preparedness of governments, which will in turn enable them to react quicker during haze events. One element of forecasting which could be improved is identifying dispersion patterns of smoke haze from its source so that potential hazard areas can be earmarked for early warnings to local governments and the general population. Developing a comprehensive database on emission factors for major air pollutants from BB areas would be valuable to modellers working on atmospheric dispersion and dilution of pollutants released by fires in this region. The improvement of seasonal forecasts and El Niño events would in turn improve BB forecasts that couple meteorological parameters with information such as class ignition potential of grass fuels provided by the ASEAN Specialised Meteorological Centre (ASMC) Fire Danger Rating System. Mobilizing resources from different countries within a region is crucial in decreasing response time. Resource sharing is also crucial for consensus building. For example, moving towards the concept of ASEAN as one ecosystem, the regional scientific community can be encouraged to come to an agreement on the specific major air pollutants breakpoints for the ASEAN Air Quality Index to be used for haze measurement and monitoring as additional provision in ASEAN Agreement on Transboundary Haze Pollution.

ASEAN can take the lead in encouraging government-private partnerships in the region to commit resources to identify and implement definitive economic approaches that promote the utilization of the ‘unwanted’ biomass as material, fuel, or as a source of electrical power as an alternative solution to “slash and burn.” Knowledge dissemination is also very important in haze prevention and mitigation. When peatlands are developed, the sustainable management of these lands must be properly understood and the information widely disseminated, especially among the communities on the ground. In this way, local-scale burning activities can be reduced due to increased awareness of the risks associated with burning. Awareness of increased burning risks during severe weather will also reduce accidental large-scale fires caused by traditionally followed habits that used to pose little risk. Reduction of local emissions during dry seasons and BB episodes will reduce the intensity of haze. The unscrupulous behaviour of some conglomerates involved in large-scale slash-and-burn for quick profits should also be communicated so that the population can exercise its buying power in order to prevent some segments of the population ending up as victims of these conglomerates.

The fires and haze are shared problems and indicate the importance of interdependence and the need to work together. The science-policy interface is crucial in prevention and mitigation measures. There is a need to strengthen the capacity to use science in decision-making creatively and effectively within ASEAN countries. Strengthening the science-policy interface, with an emphasis on developing forecasting ability and prevention methods, would generate resource sharing beyond just the scientific community within the region or between the policy makers and the scientific community in individual countries. The efforts of the government, academia, private sector and non-governmental organizations (NGOs) can individually contribute to address the fires and haze. However, greater coordination is a necessity to tie in emerging technologies with policy and an economy driven approach. Haze causes enormous local and regional harm to Southeast Asian economies and environments, but the difficulties faced also offer opportunities to promote research, innovation and strengthen the spirit of collaboration.

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## Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2018.01.002.

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