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$PM_{2.5}$ and Associated Ionic Species in a Sub-urban Coastal Area of Kuala Terengganu, Southern South China Sea (Malaysia)

($PM_{2.5}$ dan Spesies Ion Berkaitan di Kawasan Bandar Pesisir Pantai, Kuala Terengganu, Perairan Selatan Laut China Selatan (Malaysia))

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ABSTRACT

$PM_{2.5}$ mass concentration and associated water-soluble ionic species in a sub-urban coastal area of Kuala Terengganu, Malaysia were investigated intermittently from year 2006 to 2009. A total of 78 weekly $PM_{2.5}$ samples were analyzed. The mass concentration of $PM_{2.5}$ exhibited annual, seasonal and diurnal variations. Temporal distributions of rainfall, sporadic haze episodes and local air flow (sea breeze circulation) were factors controlling $PM_{2.5}$ mass variations in the study area. Although the $PM_{2.5}$ concentrations were increased during haze episodes in 2006 (August and October) and 2007 (October), their concentrations however, were still within the international guidelines. The average concentration of individual ions was in decreasing trend; $SO_4^{2-} > NH_4^+ > K^+ > Na^+ > NO_3^- > Cl^- > Ca^{2+}$. The concentrations of SO_4^{2-} and NH_4^+ accounted for > 70% of the water-soluble aerosol mass. More than 80% of ionic species associated with $PM_{2.5}$ are from non-marine sources. Major processes affecting the ionic composition of $PM_{2.5}$ are biomass burning, crustal loading and sea spray. Air quality mitigation strategies should focus on anthropogenic activities emitting SO_2, which promotes aerosol SO_4^{2-} formation.

Keywords: Aerosols; fine particles; source apportionment, trans-boundary haze episode; water-soluble ionic species

ABSTRAK

Satu kajian mengenai kepekatan jisim $PM_{2.5}$ dan spesies ion larut air di kawasan bandar pesisir pantai Kuala Terengganu telah dijalankan secara berkala mulai tahun 2006 hingga 2009. Sejumlah 78 sampel mingguan telah dianalisis. Kepekatan jisim $PM_{2.5}$ mempamerkan perubahan harian, musim dan tahunan. Taburan hujan, jerebu yang berlaku sekali-sekala dan aliran udara tempatan (kitaran bayu laut) adalah faktor yang mempengaruhi perubahan kepekatan $PM_{2.5}$ di kawasan kajian. Walaupun kepekatan $PM_{2.5}$ didapati meningkat semasa berlakunya jerebu pada 2006 (Ogos dan Oktober) dan 2007 (Oktober), namun nilainya masih di bawah aras piawai kualiti udara antarabangsa. Kepekatan purata ion individu berkurang mengikut turutan $SO_4^{2-} > NH_4^+ > K^+ > Na^+ > NO_3^- > Cl^- > Ca^{2+}$. Kepekatan SO_4^{2-} dan NH_4^+ menyumbang lebih 70% daripada keseluruhan jisim spesies ion yang dianalisis. Di samping itu, lebih 80% spesies ion ini didapati berpunca daripada sumber bukan marin. Faktor utama yang mempengaruhi kandungan $PM_{2.5}$ adalah pembakaran biojisim, elemen semula jadi daripada kerak bumi dan semburan daripada laut. Justeru itu, strategi menangani kualiti udara seharusnya memberi penekanan kepada aktiviti antropogenik yang menyebabkan pelepasan SO_2 yang menggalakkan pembentukan aerosol SO_4^{2-}.

Kata kunci: Aerosol; jerebu merentasi sempadan; pengenalpastian sumber; spesies ion terlarut air; zarah halus

INTRODUCTION

Airborne particulate matter (APM) is known to influence the environmental processes and human health. Among APM of different sizes, particulate matter with aerodynamic diameter <2.50 μm ($PM_{2.5}$) is of considerable concern as it poses great risk to health and long-term exposure to $PM_{2.5}$ is found to be associated with non-accidental mortality (Dockery et al. 1993; Pope III et al. 2002; Samet et al. 2000). Furthermore, the measurement of $PM_{2.5}$ is thought to be a better approach to assess the impact of anthropogenic activities on air quality because coarse particles are highly affected by natural sources such as sea spray and wind-blown dust (Almeida et al. 2005). The awareness on the impact of APM to human has led to intensive studies on ambient aerosols, focusing on the chemical composition and source apportionment (Fang et al. 2002; Park & Kim 2004).

In Malaysia, there is a general lack of studies on APM chemistry, particularly in sub-urban coastal areas such as Kuala Terengganu. During dry season (June to October), southeasterly seasonal wind often facilitates the northward advection of smoke-haze emitted from biomass burning (occur naturally and/or set intentionally) in Sumatra, Indonesia to Kuala Terengganu, east coast of Peninsular Malaysia (Anwar et al. 2010; Juneng et al. 2009). Though affected by seasonal smoke-haze episodes, specific studies

on Kuala Terengganu air quality are still limited. Two recent studies has been reported but the focus has been on the total particulate matter (Mohd Tahir et al. 2009) and PM_{10} (Mohd Tahir et al. 2008). In view of the gaps, this study seeks to investigate the temporal distribution and ionic composition of $PM_{2.5}$ at Kuala Terengganu coast. The results will provide baseline information for future research as the environmental condition in Kuala Terengganu becomes more complex with progressive urbanization. The understanding on $PM_{2.5}$ will also help to improvise abatement strategies for improving air quality in Kuala Terengganu.

METHODS

$PM_{2.5}$ samplings were conducted intermittently for four sequential years, 2006 to 2009, at Kuala Terengganu Meteorology Station (KTMS) (Figure 1) using low-volume air sampler (Casella APM 950). KTMS is located on an open flat ground; approximately 1.0 km from the coast and 0.10 km from the airport. The study site has low traffic and relatively low population density within 2.0 km radius. There is no important industrial operation within KTMS vicinity.

Real time mass concentration of $PM_{2.5}$ was monitored and recorded at 30 min interval using integrated data logger. The air sampler was operated 24 h continually, except during sample collection, at a flow rate of 17 L min^{-1}. The $PM_{2.5}$ was collected on pre-weighed Teflon filter of 47 mm diameter and with 0.2 μm pore size (Whatman). Mass of retained $PM_{2.5}$ was determined using a gravimetric method. For water-soluble ionic species determination, the exposed filter was cut, added with 10 mL of de-ionized water in a centrifuge tube and subjected to ultrasonic extraction (15 min interval) for an hour at <27°C. All extracts were analyzed using ion chromatography technique (Dionex Model DX-120). The results reported in this study are corrected with blank filter papers. Secondary data such as monthly total rainfall and wind speed during the study period were obtained from the Malaysia Meteorological Department (MMD).

Prevailing environmental features at KTMS in each sampling year are summarized in Table 1.

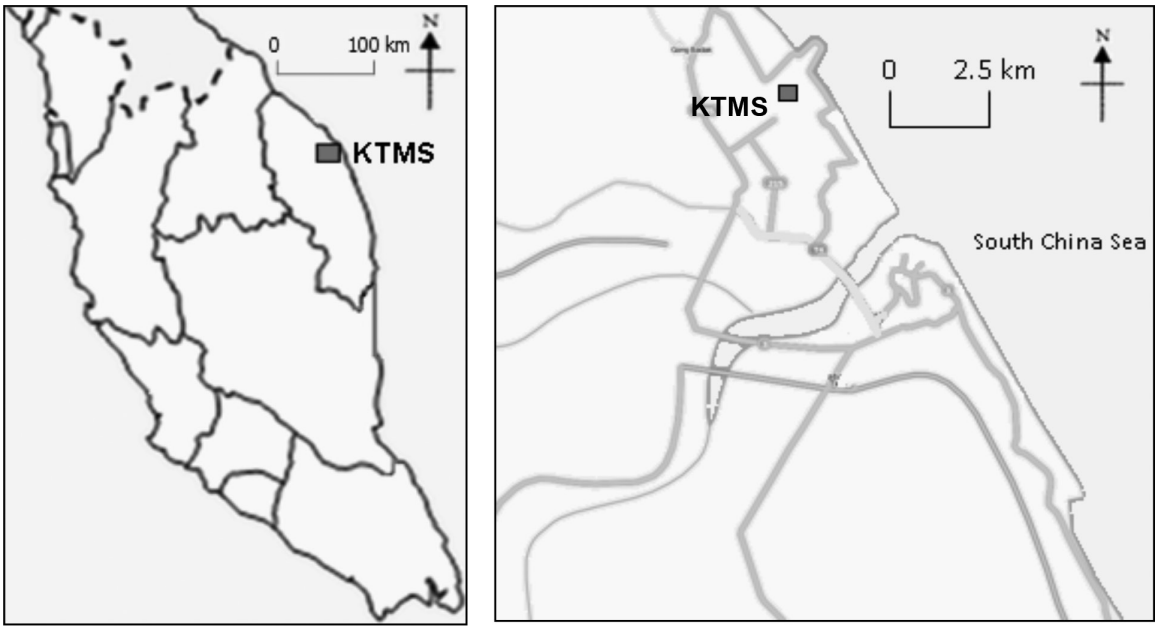

FIGURE 1. Location of Kuala Terengganu Meteorological Station (KTMS)

TABLE 1. Prevailing environmental features at KTMS in each sampling year

Year	Environmental features at KTMS
2006	KTMS area was affected by smoke-haze due to large-scale Indonesian forest fire. El Nino phenomenon caused reduction of rainfall in wet season
2007	No major haze episode was being reported. Manifest of El Nino phenomenon extended the dry season and this is shown in Figure 2
2008	No major smoke-haze episode was being reported. Rainfall was significantly increased in November and December
2009	No major smoke-haze episode was being reported. Rainfall was high throughout the sampling period

Results and Discussion

Annual variations of $PM_{2.5}$ mass concentration

In general, $PM_{2.5}$ mass concentrations were higher in the years 2006 and 2007 compared with 2008 and 2009 (Figure 2). The annual 24 h mean concentrations of $PM_{2.5}$ at KTMS were 9.50±4.00, 8.00±2.50, 5.30±1.00 and 5.40±1.50 μg m^{-3}, respectively, for 2006 to 2009. The annual mean value is calculated based on the available monthly 24 h average data. Generally, the $PM_{2.5}$ concentrations in KTMS showed amplification during haze episodes in 2006 and 2007, however, the values recorded were still within the 24 h exposure limits set by the World Health Organization (WHO 2008) and the United States Environmental Protection Agency (USEPA 2010) at 25 and 35 μg m^{-3}, respectively.

Higher $PM_{2.5}$ mass concentration in 2006 and 2007 could be attributed to the reduction of rainfall and sporadic haze episodes. El Nino phenomenon that hit Southeast Asia in 2006 reduced the amount of rainfall and conduced long dry season (Tangang et al. 2010). The dry season prolongs APM residence time and consequently resulted in higher $PM_{2.5}$ mass concentration. In August and October 2006, in conjunction with the strong manifest of El Nino, KTMS area was severely affected by smoke-haze transported from Indonesian forest fire. El Nino appeared to persist in 2007 as evidenced by relatively low rainfall, even in the wet season (November-December). Though no major haze episode was being reported in 2007, the dry season allowed accumulation of low intensity haze transported from Indonesia, causing seasonal $PM_{2.5}$ maximal to occur in between June and October. This seasonal amplification of $PM_{2.5}$ level is also observed in other sampling years. Local wind-blown dust could not be ruled out in contributing $PM_{2.5}$ as large-scale sea reclamation and construction activities commenced around KTMS since 2007. Open burning of solid wastes, particularly garden refuse, could be another crucial contributing factor since it is widely practiced by the local community.

Compared with 2006 and 2007, the average $PM_{2.5}$ mass concentration at KTMS during 2008 and 2009 were lowered by >30%. The overall reduction of $PM_{2.5}$ mass concentration is ascribed to the significant increased of rainfall in November to December of 2008 and throughout 2009 sampling period. The rainfall events enhance air particulate removal from the atmosphere (Khare & Baruah 2010; Kocak et al. 2007). Other key factor could be the decreased of biomass burning in Indonesia, owing to stringent law enforcement after the smoke-haze havoc in October 2006.

Diurnal variations of $PM_{2.5}$ mass concentration

Diurnal variations of $PM_{2.5}$ mass concentration were recorded in the study period. Diurnal pattern and $PM_{2.5}$ mass concentration in non-haze and haze episodes were found to be distinct (Figure 3). In non-haze episode, the $PM_{2.5}$ mass concentration showed gradual increment at local time 07:00 to 09:00 (land to sea breeze) and 19:00 to 21:00 (sea to land breeze), respectively. The transition between land and sea breeze promoted aerosols accumulation in horizontal boundary of opposing breezes (convergence zone) by lowering wind speed (Liu & Chan 2002; Pillai et al. 2002). During land breeze (00:00 to 06:00), low wind speed (~1.40 -1.50 m s^{-1}) limited the dispersion of APM and hence $PM_{2.5}$ mass concentration fluctuated within a narrow range of 5.00 to 6.00 μg m^{-3}. As sea breeze sets in (10:00 to 17:00), mass concentration of $PM_{2.5}$ decreased by approximately 50% to a minimum value of 2.60 μg m^{-3}. Higher wind speed (~1.70-2.60 m s^{-1}) during sea breeze could have blown away land air to inner land and therefore enhanced the dispersion of $PM_{2.5}$. In addition,

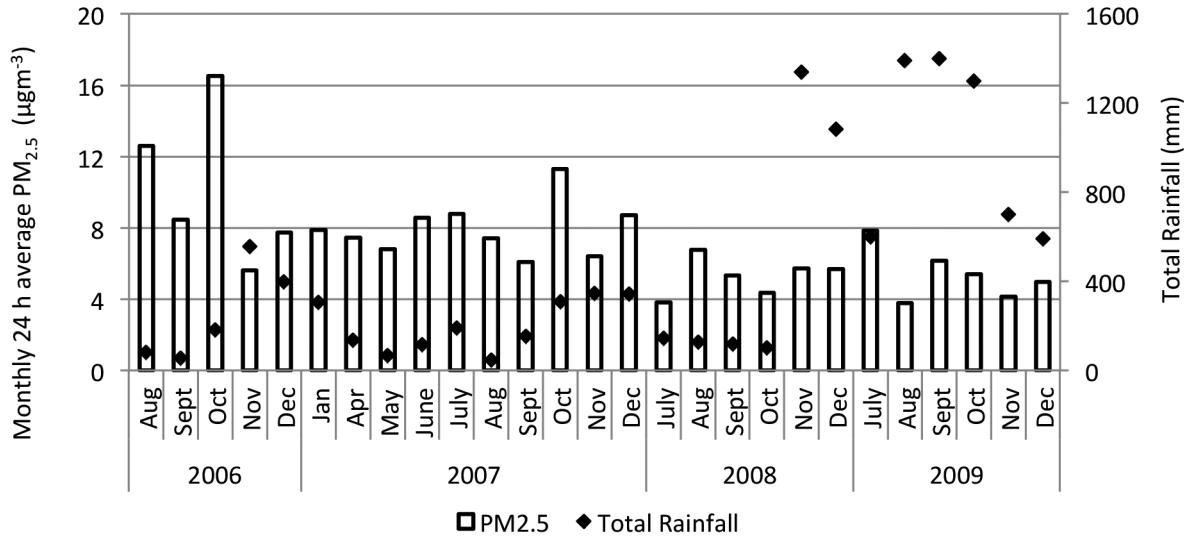

FIGURE 2. Monthly 24 h average $PM_{2.5}$ mass concentration and total rainfall at KTMS. The monthly total rainfall was provided by MMD

sea breeze may bring cleaner marine air landward and thus reduced the $PM_{2.5}$ mass concentration (Pillai et al. 2002).

On the contrary, in haze episode, higher wind speed during sea breeze (11:00-16:00) appeared to transport more $PM_{2.5}$ to KTMS. Such observation may indicate the recirculation of $PM_{2.5}$ back to the land after it was being transported out to the sea during land breeze (Baumgardner et al. 2006; Eleftheriadis et al. 1998). The $PM_{2.5}$ level in haze episode was approximately ten times higher than in non-haze. High $PM_{2.5}$ mass concentration and prevailing southeasterly wind could signify long-range transport of smoke-haze from Indonesian forest fire to KTMS.

GENERAL AEROSOL CHEMISTRY

Major water-soluble ionic species identified in $PM_{2.5}$ were SO_4^{2-}, NH_4^+, K^+, Na^+, NO_3^-, Cl^- and Ca^{2+}. Table 2 presents the concentrations of ionic species associated with $PM_{2.5}$ collected throughout the sampling period and their relative weight percentage.

The concentrations of major ionic species were in decreasing trend of SO_4^{2-}> NH_4^+> K^+> Na^+> NO_3^-> Cl^-> Ca^{2+}. Among the ionic species, SO_4^{2-} and NH_4^+ accounted for > 70% of the water-soluble aerosol mass, suggesting these secondary aerosols are important components in the formation of $PM_{2.5}$. High SO_4^{2-} concentration (~60%) may enhance the acidity of $PM_{2.5}$ if there were no sufficient cations to neutralize SO_4^{2-}. Significant correlations of SO_4^{2-}-NH_4^+ (r=0.70) and SO_4^{2-}-Ca^{2+} (r=0.65) indicates that NH_4^+ and Ca^{2+} are important for SO_4^{2-} neutralization. Hence, SO_4^{2-} could exist as ammonium salt and gypsum. Letovicite [$(NH_4)_3H(SO_4)_2$] or solution with corresponding ions is the dominant species of SO_4^{2-} since NH_4^+/ SO_4^{2-} molar ratio (1.60) is above the theoretical value of 1.50 (Hernandez-Mena et al. 2010; Seinfield & Pandis 1998). The molar ratio suggests that substantial fraction of SO_4^{2-} is neutralized by NH_4^+. This result is reasonable since SO_4^{2-} and NH_4^+ are dominant species in $PM_{2.5}$.

The impact of marine sources on ionic composition of $PM_{2.5}$ is estimated by comparing mass ratio of ionic component (X) and Na^+ ([X] / [Na^+]) to the ratio in seawater (Table 3). Na^+ is used as tracer for estimating the contribution of marine sources, assuming all Na^+ to be of marine origin. The ionic ratio may overestimate NSS components as Na^+ could be originating from other sources such as soil dust. However, the ratio is still adequate for providing useful guidelines.

Ionic ratio larger than in seawater indicates incorporation of non sea-salt (NSS) constituents in $PM_{2.5}$. The concentrations of NSS-SO_4^{2-}, -K^+ and -Ca^{2+} are calculated as NSS-X = [X] - [Na^+] x (ionic ratio of X in seawater) (Table 3). Lower Cl^- / Na^+ ratio could be related to the fractionation of sea-salt (SS) and modification by non-marine constituents. The Cl^- is loss through reaction between NaCl with acidic species such as HNO_3, SO_2 and H_2SO_4 (Prodi et al. 2009; Ventakaraman et al. 2002). Higher correlation of Cl^--SO_4^{2-} (r=0.6) than Cl^--NO_3^- (r=0.20) indicates that SO_4^{2-} has more important role in Cl^- depletion. The NSS constituent calculation suggests that marine sources are main contributors of Cl^- while 87% of Ca^{2+}, 97% of both SO_4^{2-} and K^+ are from non-marine sources. On the whole, NSS ionic species (including NH_4^+ and NO_3^-) accounted for 88% of total ions associated with $PM_{2.5}$. Low marine contribution to $PM_{2.5}$ is expected because marine aerosol is typically associated with coarse particles (Almeida et al. 2005).

Correlations of Na^+-Cl^- (r=0.70) and Ca^{2+}-Cl^- (r=0.60) suggests that Na and Ca are mainly derived from marine sources. Strong to moderate correlations of Ca^{2+}-Na^+ (r=0.70), Ca^{2+}-K^+ (r=0.45) and Na^+-K^+ (r=0.40) imply the possible of crustal loading to $PM_{2.5}$ (Kumar & Sarin

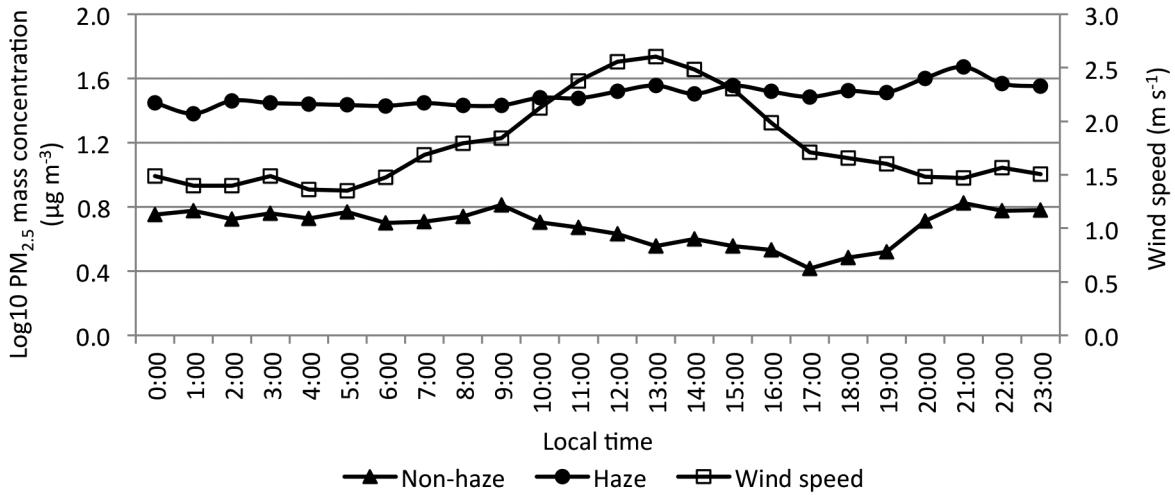

FIGURE 3. Diurnal variations of $PM_{2.5}$ mass concentrations (in non-haze and haze episodes) and a general wind speed pattern at KTMS

TABLE 2. Concentrations of ionic species associated with $PM_{2.5}$ and their relative weight percentage

Ionic species	Concentrations (ng m^{-3})			Weight percentage (%)
	Mean $_{n=78}$	Min	Max	
SO_4^{2-}	3804	93.13	1.512×10^4	57.57
NH_4^+	1119	263.0	5592	16.94
K^+	624.4	106.0	3746	9.450
NO_3^-	311.6	4.900	2353	4.717
Ca_2^+	132.6	2.530	1532	2.007
Cl^-	158.9	12.40	1874	2.405
Na^+	456.3	25.60	3648	6.906
Total average	6607			

TABLE 3. Ionic ratio and mean concentrations of NSS constituents

Ionic species	Ionic ratio		Non sea-salt (NSS)	Mean concentration (ng m^{-3})	Weight percentage (%)
	This study	Seawater			
SO_4^{2-}	8.337	0.2516[a]	SO_4^{2-}	3689	96.98
K^+	1.368	0.0400[a]	K^+	606.1	97.08
Ca^{2+}	0.2906	0.0385[b]	Ca^{2+}	115.0	86.75
Cl^-	0.3482	1.800[b]			

[a] Karthikeyan & Balasubramaniam 2006
[b] Balasubramaniam et al. 2003

2010; Pey et al. 2009; Wang et al. 2005). Meanwhile, high correlations of NH_4^+-SO_4^{2-} (r=0.70) and NH_4^+-K^+ (r=0.60) indicates their common source mainly from biomass burning (regional and/or local emission). The potential of biomass burning in contributing atmospheric SO_4^{2-}, NH_4^+ and K^+ have been widely acknowledged in literatures (Chan et al. 1997; Kang et al. 2004; Sun et al. 2006).

The NO_3^-- SO_4^{2-} correlation (r=0.19) is unexpectedly low in this study possibly because NO_3^- exists mainly in coarse particles while SO_4^{2-} has bimodal (fine and coarse) distribution in aerosol (Kumar & Sarin 2010; Venkataraman et al. 2002). The SO_4^{2-}/NO_3^- mass ratio is regularly used as indicator to evaluate the relative importance of mobile (vehicular emission) versus stationary sources (biomass burning, open burning, industrial emission) of SO_4^{2-} and NO_3^- in atmosphere (Arimoto et al. 1996; Hu et al. 2002; Tan et al. 2009). The NSS-SO_4^{2-}/NO_3^- mass ratio in this study exhibits considerable variability, with value ranging from 8.00 to 53.0. Nevertheless, the ratio is overwhelmingly higher than in Beijing (1.70) and Shanghai (2.50) where stationary sources of SO_4^{2-} and NO_3^- were found to be dominant over mobile sources (Yao et al. 2002). High NSS-SO_4^{2-}/NO_3^- mass ratio at KTMS could indicate that SO_4^{2-} and NO_3^- are predominantly contributed by stationary sources rather than mobile sources. Determination of NSS-SO_4^{2-}/NO_3^- mass ratio in coarse APM is essential to give better insight on SO_4^{2-} and NO_3^- source apportionment.

As discussed earlier, $PM_{2.5}$ mass concentration shows annual variations due to temporal distributions of rainfall and sporadic haze episodes. These events could also affect the relative weight percentage of NSS ionic species in $PM_{2.5}$ (Figure 4).

Each pie chart represents ionic composition of $PM_{2.5}$ collected under different environmental conditions (Table 1). Compared with 2006, K^+ was more important for the formation of $PM_{2.5}$ collected in 2007, 2008 and 2009. In addition to Indonesian biomass burning, this notable variation could be ascribed to crustal input from sea reclamation and construction activities operating around KTMS since 2007. The Ca^{2+} shared similar variation trend as K^+, probably because of the aforementioned sea reclamation and construction activities. Similar results were obtained in the studies conducted in Hong Kong and Singapore (Balasubramaniam et al. 2003; Ka & Tanner 1999). Local air flows, such as land and sea breezes in this study, may facilitate the re-suspension and transportation of dust particles to KTMS.

Long dry season coupled with Indonesian forest fire in 2006 and 2007 has resulted in high SO_4^{2-} composition. This is expected as the peat bog in Indonesia is known for its high sulfur content due to wet and dry deposition of volcanic sulfur (Balasubramaniam et al. 2003; Tangang et al. 2010). In 2008, the weight percentage of SO_4^{2-} appeared to maintain high, probably because rainfall significantly increased only in November and

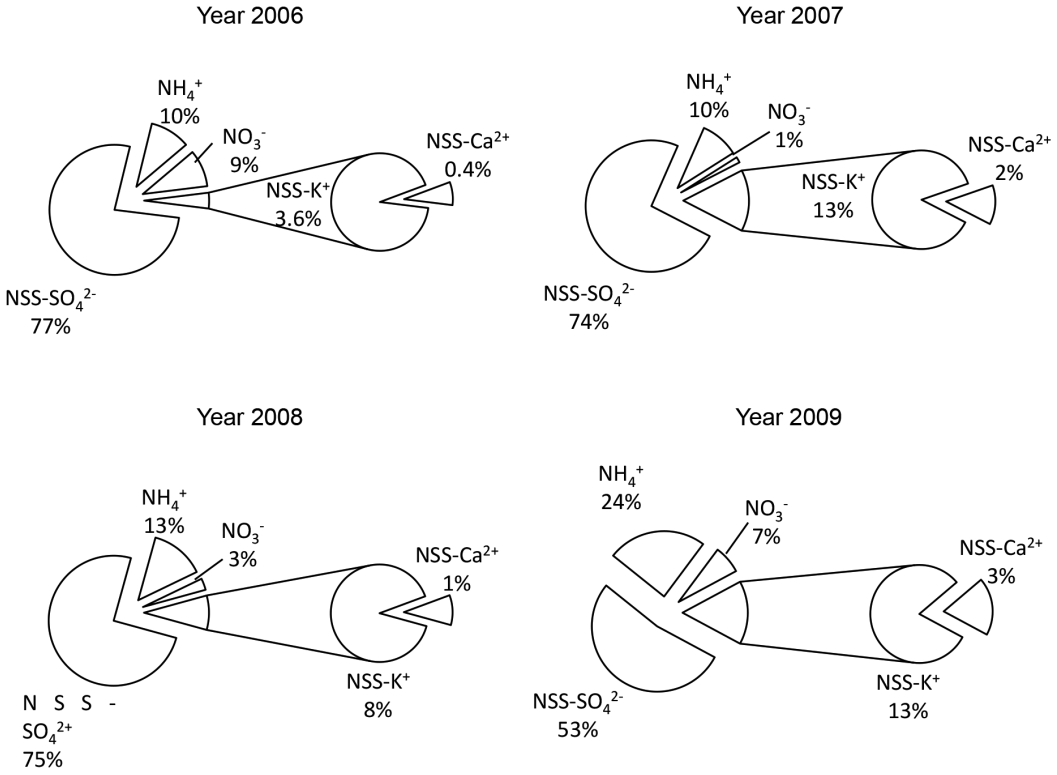

FIGURE 4. Relative weight percentage of NSS ionic species in $PM_{2.5}$ collected under different environmental conditions

December, after the peak season of Indonesian forest fire. Therefore, we could expect low intensity haze transported from Indonesian forest fire to KTMS in earlier months and increased the overall weight percentage of SO_4^{2-}. High rainfall throughout 2009 sampling subsequently decreased the weight percentage of SO_4^{2-} by 20-25%, suggesting that high rainfall could reduce the amount of smoke-haze transported from Indonesia to KTMS. The contribution of SO_4^{2-} from local sources could be high in view of the fact that $PM_{2.5}$ collected under improved environmental conditions in 2009 (no major smoke-haze emission from Indonesia and high rainfall) still contain 53% of SO_4^{2-}. Further verification is needed to confirm this as without measuring the ambient SO_2 level at KTMS, we are not able to determine the relative importance of local emission versus long-range transport in contributing SO_4^{2-}. Nevertheless, the SO_4^{2-} in $PM_{2.5}$ has to be reduced as it is proven to have direct link with lung cancer and cardiopulmonary mortality (Brook et al. 2004; Pope III et al. 2002). Hence, abatement strategies to improve air quality should focus on activities emitting SO_2, a precursor gas which is oxidized to SO_4^{2-} aerosol.

CONCLUSION

The results from this study showed that the variations of $PM_{2.5}$ mass concentration are related to temporal rainfall distributions and sporadic haze episodes. Prolong dry season caused by El Nino phenomenon exacerbates the $PM_{2.5}$ mass concentration in atmosphere. Diurnal variations of $PM_{2.5}$ mass concentration are related to the transition of land and sea breezes. Southeasterly wind facilitates the long-range transport of smoke-haze from Indonesia to KTMS. Annual average concentration of ionic species associated with $PM_{2.5}$ are in decreasing trend of $SO_4^{2-} > NH_4^+ > K^+ > Na^+ > NO_3^- > Cl^- > Ca^{2+}$. During this study period, contribution of NSS sources to the formation of $PM_{2.5}$ was found to be more important than marine sources. Correlation analysis of combined data set (2006-2009) indicates three major sources of ionic species associated with $PM_{2.5}$ viz. biomass burning, crustal loading and sea spray. It is suggested that strategies to improve air quality in this area should focus on anthropogenic activities emitting SO_2, which is conducive to the formation of SO_4^{2-} aerosol.

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