



## Monsoonal variations in atmospheric surfactants at different coastal areas of the Malaysian Peninsula



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### ABSTRACT

This study determined the effect of monsoonal changes on the composition of atmospheric surfactants in coastal areas. The composition of anions ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{F}^-$ ) and the major elements (Ca, K, Mg, Na) in aerosols were used to determine the possible sources of surfactants. Surfactant compositions were determined using a colorimetric method as methylene blue active substances (MBAS) and disulphine blue active substances (DBAS). The anion and major element compositions of the aerosol samples were determined by ion chromatography (IC) and inductively coupled plasma mass spectrometry (ICP-MS), respectively. The results indicated that the concentrations of surfactant in aerosols were dominated by MBAS ( $34\text{--}326 \text{ pmol m}^{-3}$ ). Monsoonal changes were found to significantly affect the concentration of surfactants. Using principal component analysis-multiple linear regressions (PCA-MLR), major possible sources for surfactants in the aerosols were motor vehicle emissions, secondary aerosol and the combustion of biomass along with marine aerosol.

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### 1. Introduction

Surfactant is one of the organic pollutants which can affect the solubility and surface tension of atmospheric aerosols (Brimblecombe and Latif, 2004; Koopal et al., 2004). It has naturally been associated with marine sources, particularly through the contribution of surfactants from the sea-surface micro-layer (SML) (Becagli et al., 2011; Cincinelli et al., 2001; Elliott et al., 2014; Jaafar et al., 2014; Mustafa et al., 2014; Sellegri et al., 2006; Smoydzin and Von Glasow, 2007). Furthermore, surfactants in aerosols can be generated through various anthropogenic activities, such as: vehicular emissions and biomass burning (Roslan et al., 2010; Wahid et al., 2013). At high concentrations, atmospheric surfactants disrupt the stability of the human respiratory system and cause asthma, allergies and dry eyes (Vejrup and Wolkoff, 2002; Zimmer et al., 2002). They also have the potential to diminish vegetation, particularly any which is growing around coastal areas (Becagli et al., 2011; Nicolotti et al., 2005; Paoletti et al., 2005). In the upper part of the atmosphere, atmospheric surfactants have been found to affect the surface tension of water droplets, which in turn leads to more

droplets being generated. This fundamentally impacts climate change through the formation of cloud condensation nuclei (CCN) (Moore et al., 2011; Westervelt et al., 2012).

A seasonal effect is also found to contribute to the quantity of surfactants in the atmosphere, including those in coastal areas (Baduel et al., 2012; Mustafa et al., 2014; Wahid et al., 2013). The movement of wind over the ocean usually brings a sea breeze containing surfactants to the coastal area (Alsalahi et al., 2014; Nicolotti et al., 2005; Olkowska et al., 2014). Moreover, surfactants can also be contributed to by the destruction of humic-like substances which originate from soil dust, biomass burning and other combustion-related activities and are carried elsewhere by the wind. Additionally, regional monsoon winds potentially carry surfactants in fine mode particles (with diameter ( $d$ ) below than  $1.5 \mu\text{m}$ ) via a long range transport from their original sources. Surfactants in coarse mode particles ( $d > 1.5 \mu\text{m}$ ) are usually contributed to by local sources (Latif et al., 2005). The rainy season, as a result of the movement of monsoonal winds, reduces the concentration of surfactants through the dilution effect. Whereas the dry season generally encourages the movement of soil dust, and as a result, increases the likelihood of a burning episode which would in effect contribute to the quantity of surfactants in existence, particularly in tropical regions (Latif et al., 2011; Mustafa et al., 2014).

Seasonal monsoons play a significant role when it comes to changes occurring in atmospheric aerosols and air quality. This is especially true

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for tropical countries such as Malaysia. On the Malaysian Peninsula, the southwest monsoon is linked to biomass burning from agricultural peat areas in Sumatra. The northeast monsoon also has connections with the wind which comes from Indochina and brings with it more rain from the South China Sea (Anwar et al., 2010; Juneng et al., 2009). Taking all these points into consideration, the aim of this study is to determine the concentration of surfactants in both coarse (diameter size,  $d > 1.5$ ) and fine mode ( $d < 1.5$ ) aerosols collected from the western and eastern coastal areas of the Malaysian Peninsula within two different monsoon periods, namely the northeast and southwest monsoons. Source apportionment analysis was also conducted to analyze the composition of surfactants in atmospheric aerosols. Additionally, principal component analysis (PCA), in combination with multiple linear regressions (MLR), will be used to estimate the source apportionment of aerosols from the two study areas.

## 2. Materials and methods

### 2.1. Sampling site

Sampling was conducted at two different coastal sites on the Malaysian Peninsula, namely: Port Dickson (on the west coast of the Peninsular on the Malacca Straits) and Kuala Terengganu (on the east coast of the Peninsular on the southern part of South China Sea). Samples were collected to coincide with different monsoons in Southeast Asia. The sampling undertaken for the southwest monsoon was conducted between May and September 2012 during a period when relatively drier weather would be expected (Juneng et al., 2011). While for the northeast monsoon, sampling was conducted between November 2012 and March 2013, during which heavy rainfall, particularly in the east coastal states of the Malaysian Peninsula, is prevalent. Fig. 1 shows details of the sampling locations.

### 2.2. Sampling procedure

#### 2.2.1. Atmospheric aerosols

The aerosol samples were taken from two sampling stations between May 2012 and March 2013. These samples were collected using a high volume air sampler (HVAS) (Thermo Scientific Model GS2313-105, USA) in combination with a two-stage cascade impactor (Staplex, USA). Slotted filter papers measuring  $12 \times 10$  cm (Westech Instrument, UK) were used to collect coarse mode aerosols ( $d > 1.5 \mu\text{m}$ ) whilst  $20.3 \times 25.4$  cm backup filter papers (Whatman EPM 2000) were used to collect fine mode aerosols ( $d < 1.5 \mu\text{m}$ ). After collection, the filter papers were wrapped in aluminium foil and pre-heated in a muffle furnace (Carbolite, UK) ( $500^\circ\text{C}$ , 4 h) in order to remove any organic contaminants. The filters were then conditioned in a desiccator for a minimum of 24 h before being weighed with an electronic balance (Shimadzu AUX220, Japan) and placed in the HVAS. After installation, the HVAS was placed in an open field at close proximity to the sea so as to avoid any disturbance to the flow of aerosols entering the instrument. The sampling continued for 72 h over a three day campaign with a flow rate of  $1.13 \text{ m}^3 \text{ min}^{-1}$ , during which the samples were collected at every 24 h. After sampling, the filter papers were wrapped in aluminium foil and conditioned in desiccators (24 h) before being weighed. For quality control purposes, the blank filter papers were prepared for sampling in the same way as the filter papers had been but without the HVAS being turned on.

### 2.3. Sample preparation

#### 2.3.1. Aerosol sample extraction

With regard to the extraction processes used to determine the level of surfactants in the coarse mode aerosol samples, half of the filter papers were used whereas for the fine mode samples only a quarter of the filter papers were needed. The filter papers were initially cut into

one  $\text{cm}^2$  pieces and put into a centrifuge tube. About 40 mL of ultra-pure water was added to the samples before they were sonicated for 45 min, as undertaken by Razak et al. (2013); Wahid et al. (2013) and Jaafar et al. (2014). The ultra-pure water had a resistivity of  $< 2 \text{ M}\Omega\text{m}$  and was prepared using an Arium 611DI deionised water system (Sartorius, Germany). The samples were then filtered using cellulose acetate filter papers (Whatman, Germany –  $47 \text{ mm}/0.2 \mu\text{m}$  pore size) and a vacuum filter pump (Millipore, USA). After this they were diluted to 100 mL with ultra-pure water and stored in a refrigerator ( $< 4^\circ\text{C}$ ) until further analysis.

### 2.4. Surfactant analysis

#### 2.4.1. Anionic surfactants as methylene blue active substances (MBAS)

The sample solution (20 mL) was put into a 40 mL vial (vial A), equipped with a screw cap and Teflon liner. The alkaline buffer (2 mL) and neutral methylene blue solution (1 mL), followed by chloroform (5 mL), were added to vial A in that order. The vial was closed tightly and then vigorously shaken for two minutes using a vortex mixer. After being shaken, the vial was left to allow phase separation to occur after which the screw cap was loosened to release the pressure inside. Once the two phases were separated, a Pasteur pipette was used to transfer the chloroform layer into a new vial (vial B) that contained ultra-pure water (22 mL) and an acid methylene blue solution (1 mL). Vial B was shaken for two minutes in a vortex mixer before its cap was loosened for a few seconds to release pressure and then retightened. After the chloroform had completely separated from the water (after two minutes) the chloroform layer was collected using a Pasteur pipette and put into a 10 mm quartz cell. The absorbance of the chloroform phase was measured using an UV spectrophotometer (Labomed, Inc., USA) at a wavelength of 650 nm. The limit of detection was  $0.05 \mu\text{M}$  Sodium Dodecyl Sulphate (SDS) (Sigma-Aldrich, USA). The calibration curve was prepared using SDS for surfactants as MBAS and the average recovery value for MBAS was 87%.

#### 2.4.2. Cationic surfactants as disulphine blue active substances (DBAS)

A volume of 20 mL of the sample solution was put into a 40 mL vial equipped with a screw cap and Teflon liner. The acetate buffer (2 mL) and disulphine blue solution (1 mL), followed by chloroform (5 mL), were added in that order. The vial was closed tightly and then shaken vigorously for two minutes using a vortex mixer. The cap was loosened for a few seconds to release the pressure inside and then retightened. The vial was left for approximately two minutes until the two phases had completely separated. The chloroform layer was then removed from the vial using a pasteur pipette and placed into a 10 mm quartz cell. Light absorbance was measured at a wavelength of 628 nm. The limit of detection was  $0.04 \mu\text{M}$  for Zephiramine (benzyl-dimethyl-tetradecyl-ammonium chloride dehydrate) (Sigma-Aldrich, USA) standard solution. The calibration curve was prepared using Zephiramine for surfactants as DBAS. A linear calibration curve was formed from 0.05 to  $2.00 \mu\text{M}$ . The average recovery value for DBAS measured 89%.

### 2.5. Ionic and major elements composition

Ion Chromatography (Metrohm, 881 Compact IC Pro, USA) was used to determine the anions in the aerosol samples. Standard solutions were prepared using four individual standard anion solutions (Merck, USA). A Metrosep A-Supp 5–150/4.0 column, with a flow rate of  $0.7 \text{ mL min}^{-1}$ , was used.  $\text{F}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  were detected and used for data analysis, with method detection limits of  $0.005 \mu\text{g m}^{-3}$  for  $\text{F}^-$ ,  $0.005 \mu\text{g m}^{-3}$  for  $\text{Cl}^-$ ,  $0.005 \mu\text{g m}^{-3}$  for  $\text{NO}_3^-$  and  $0.001 \mu\text{g m}^{-3}$  for  $\text{SO}_4^{2-}$ . Inductively coupled plasma mass spectrometry (ICP-MS) (Perkin Elmer ELAN 9000, USA) was used to determine the major elements of the aerosol samples. The ICP-MS was calibrated using the PerkinElmer multi-element standard 3. In this study, four major elements were detected: Na, K, Mg and Ca. The detection limits for these elements are

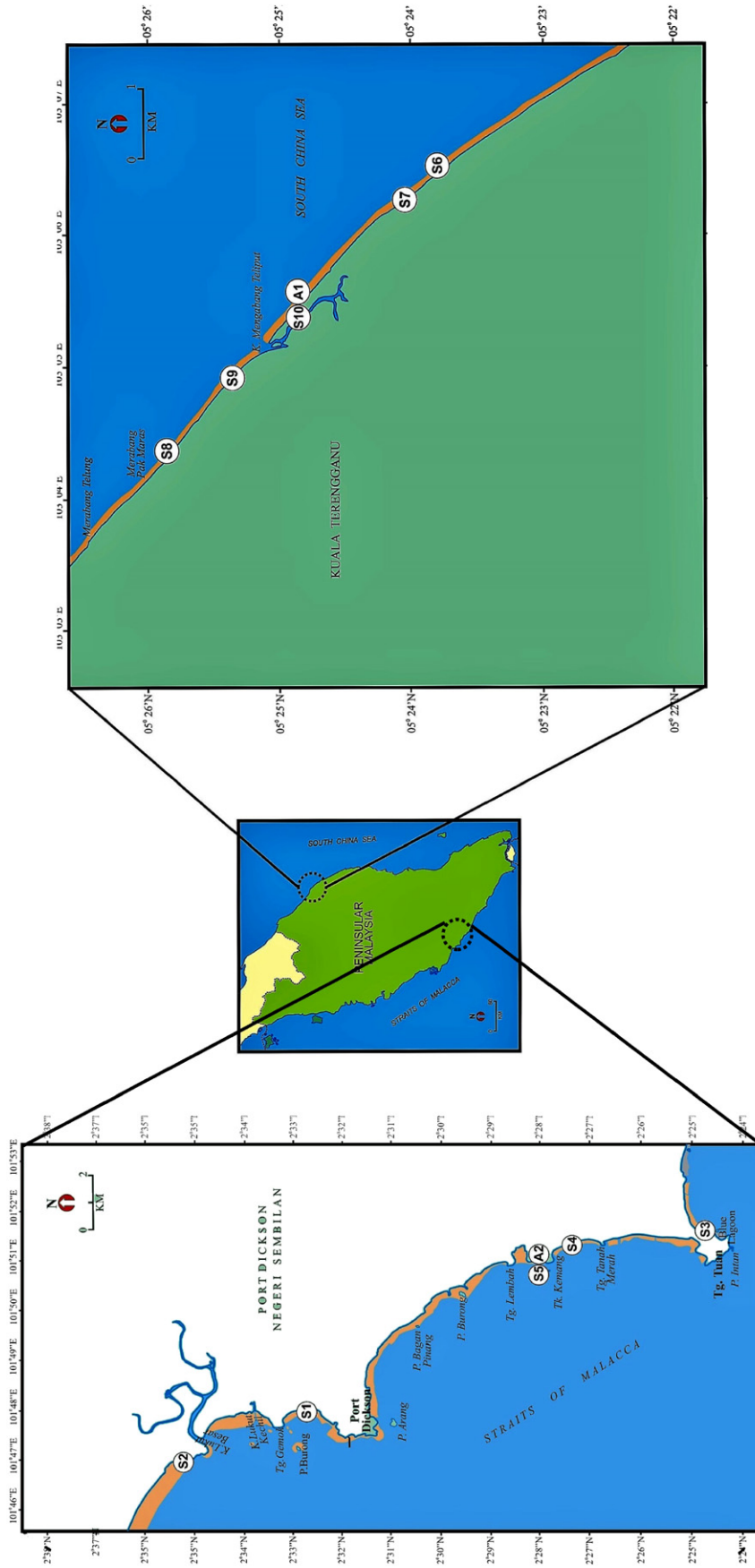


Fig. 1. Sampling stations at Port Dickson and Kuala Terengganu.

0.003  $\mu\text{g m}^{-3}$ , 0.003  $\mu\text{g m}^{-3}$ , 0.004  $\mu\text{g m}^{-3}$  and 0.002  $\mu\text{g m}^{-3}$ , respectively, with a flow rate of 0.01 to 0.3  $\text{mL min}^{-1}$ .

## 2.6. Quality assurance/quality control

As part of the quality assurance/quality control (QA/QC) to ensure the accuracy and validity of the results, all glassware was washed before use: first with hexane, followed by acetone and de-ionised water. Vials were submerged in a 20% nitric acid bath for 24 h before being heated in a furnace (500 °C, 3 h). Non-powdered gloves were worn and extra care was taken during both the experiment and when handling the filter papers so as to avoid any contamination from exposure to the hands. In addition, no detergents were used for any cleansing purposes. Prior to field sampling, blank filters were heated in a furnace at 500 °C for 4 h (as explained in the previous section) in order to remove any organic contaminants. Blank filter papers were also analyzed in the same manners for control analysis, and the results were corrected based on the blank average concentration. To ensure the precision and reliability of the results, recovery analysis was undertaken. The results were carefully compared and any data below the limit of detection were discounted.

For recovery analysis, a mixture of standard solution was used. A similar concentration of several samples was prepared from the standard solution and taken through the extraction procedures, as followed for the filter samples. An analysis of the recovery samples was conducted using IC, ICP-MS and UV Spectrophotometry as for the determination of anion, major elements and surfactants in the samples, respectively. Thus, the recovery of each variable from the observed and actual concentrations was estimated with the results of the recovery expressed in percentage (%).

The limit of detection (LOD) for anions, major elements and surfactants was estimated as the times of the *t*-value with standard deviation for each variable. Calculation of LOD, to determine the Method Detection Limit (MDL), was based on the United State Environmental Protection Agency (USEPA, 1997) suggested method, using the formula below:

$$\text{MDL} = s \times t_{(n-1, 1-\alpha=0.99)}$$

where:

*n* = Number of replicate spike determinations at 1 to 5 times the estimated MDL.

*s* = Standard deviation for measured concentrations of *n* spike determinations.

*t* = Student's *t* value at *n* – 1 degree of freedom and 1 –  $\alpha$  (99%) confident level.

$\alpha$  = Level of significance.

## 2.7. Statistical analyses

The Statistical Package for the Social Sciences (SPSS version 18) (IBM, USA) was used to analyse all the data collected. Statistical analyses such as the paired *t*-test were carried out after the data was found to be normally distributed. Paired *t*-tests were used to compare between different types of surfactants (MBAS and DBAS concentrations); surfactants (as MBAS, DBAS), anions and major elements in coarse and fine mode aerosols (between different sizes); surfactants (as MBAS and DBAS), anions and major elements in different seasons (between northeast and southwest monsoons). XLSTAT 2012 software (Addinsoft, USA) was used to obtain the source apportionment of surfactants in the atmosphere combining PCA and MLR. The variables were optimally correlated with one component but had the least correlation with other components. In order to maximize the variance of the loading in all variables for all factors, the varimax rotation method was applied (Shrestha and Kazama, 2007; Singh et al., 2004; Singh et al., 2005; Wahid et al., 2013).

In order to obtain the significant value for the principal component, an eigenvalue greater than one was chosen while the analyses ran. The factor loading after rotation can be classified as strong (>0.75), moderate (0.50–0.75) or weak (<0.50). A factor loading of >.75 was chosen for MLR for the analysis of source apportionment (Liu et al., 2003). Studies by Chatterjee et al. (1999), as well as Petrie and Sabin (2000), have suggested that in order to calculate the contribution of each parameter to the level of pollution, the variability between independent and dependent variables using MLR should be predicted. Two variables, the factor scores and the anion and elemental concentrations from each sampling site, were used in the MLR models. Both variables were then compared based on the modelling performance referring to the coefficient of determination,  $R^2$ . According to Norusis (1990), the largest  $R^2$  value indicates the best linear model. In this study, each variable of anion and elemental concentrations was independently introduced to a linear regression model with the surfactant concentrations as the dependent variable. After the sources of surfactants were obtained, the contribution of each source was calculated based on the  $R^2$  value (Dominick et al., 2012; Ilten and Selici, 2008).

## 2.8. Trajectory analysis

The mean cluster of 72 h backward trajectories was estimated using the Hybrid-Single Particle Lagrangian Integrated Trajectory (HYSPPLIT 4.9) model. The backward trajectories were calculated to determine the transport path of the air mass before its arrival at the sampling stations.

## 3. Results and discussion

### 3.1. Concentrations of atmospheric aerosols

The concentration of atmospheric aerosols in the vicinity of Port Dickson and Kuala Terengganu are shown in Table 1. For each sampling site, three sampling campaigns were undertaken during each monsoon. Based on the analysis conducted during the southwest monsoon, the range of concentrations of coarse mode atmospheric aerosols ( $d > 1.5 \mu\text{m}$ ) in the vicinity of Port Dickson was found to be between 7.7  $\mu\text{g m}^{-3}$  to 23  $\mu\text{g m}^{-3}$  while the concentrations of fine mode ( $d < 1.5 \mu\text{m}$ ) ranged from 5.6  $\mu\text{g m}^{-3}$  to 14  $\mu\text{g m}^{-3}$ . As for total concentration of aerosols for this monsoon, the range was identified as being between 18  $\mu\text{g m}^{-3}$  and 33  $\mu\text{g m}^{-3}$ . The concentrations of aerosols for coarse and fine mode during the northeast monsoon were detected as being between 5.7  $\mu\text{g m}^{-3}$  and 23  $\mu\text{g m}^{-3}$ , and from 6.5  $\mu\text{g m}^{-3}$  to 20  $\mu\text{g m}^{-3}$  respectively. At the same time, the total concentration of atmospheric aerosols was recorded at between 12  $\mu\text{g m}^{-3}$  and 37  $\mu\text{g m}^{-3}$ .

During the southwest monsoon, the concentration of coarse mode ( $d > 1.5 \mu\text{m}$ ) atmospheric aerosols for Kuala Terengganu was recorded as being between 17  $\mu\text{g m}^{-3}$  and 20  $\mu\text{g m}^{-3}$ . While the range of fine mode particles was found to be from 15  $\mu\text{g m}^{-3}$  to 20  $\mu\text{g m}^{-3}$ . For total atmospheric aerosols concentration, the range was identified as being between 34  $\mu\text{g m}^{-3}$  and 39  $\mu\text{g m}^{-3}$ . During the northeast monsoon, the concentrations of atmospheric aerosols for coarse and fine mode were recorded as being between 14 and 33  $\mu\text{g m}^{-3}$ , and from 2.0 to 17  $\mu\text{g m}^{-3}$  respectively. Overall, the total concentrations of atmospheric aerosols were noted as being between 20 and 51  $\mu\text{g m}^{-3}$ .

The results show that the average concentrations of coarse mode particles were greater than those for fine mode particles. This is because the accumulation and formation of fine aerosols has no significant association with the marine environment, which is more likely to spread coarse aerosols from the sea. Coarse mode aerosols itself is formed from compounds containing various salts and organic chemicals (Roelofs, 2008) and is dispersed into the air through breaking waves, wind dispersion, vaporisation and the formation of complex compounds with ions in the air (Cini and Loggio, 1997). However, fine mode aerosols with an aerodynamic diameter ranging from 0.1 to

**Table 1**  
The average concentrations of coarse and fine mode aerosols as collected in Port Dickson and Kuala Terengganu during different monsoon seasons.

Station	Concentrations of total atmospheric aerosols ( $\mu\text{g m}^{-3}$ )					
	Southwest monsoon			Northeast monsoon		
	Coarse ( $d > 1.5 \mu\text{m}$ )	Fine ( $d < 1.5 \mu\text{m}$ )	Total	Coarse ( $d > 1.5 \mu\text{m}$ )	Fine ( $d < 1.5 \mu\text{m}$ )	Total
Port Dickson						
Campaign 1 ( $n = 3$ )	21 $\pm$ 2.0 (20–23)	9.2 $\pm$ 4.2 (5.6–14)	31 $\pm$ 6.3 (29–33)	13 $\pm$ 9.4 (5.7–23)	12 $\pm$ 4.5 (6.5–13)	24 $\pm$ 14 (12–37)
Campaign 2 ( $n = 3$ )	10 $\pm$ 3.0 (7.9–14)	11 $\pm$ 2.3 (8.4–13)	21 $\pm$ 5.3 (19–22)	8.9 $\pm$ 2.3 (6.3–10)	12 $\pm$ 3.7 (9.6–16)	21 $\pm$ 6.0 (16–27)
Campaign 3 ( $n = 3$ )	8.5 $\pm$ 1.2 (7.7–9.9)	10 $\pm$ 0.44 (10–11)	19 $\pm$ 1.7 (18–20)	13 $\pm$ 4.00 (10–18)	11 $\pm$ 7.3 (6.8–20)	24 $\pm$ 11 (19–30)
Average	14 $\pm$ 6.6 (7.7–23)	10 $\pm$ 2.6 (5.6–14)	24 $\pm$ 9.1 (18–33)	11 $\pm$ 5.6 (5.7–23)	12 $\pm$ 4.7 (6.5–20)	23 $\pm$ 10 (12–37)
Kuala Terengganu						
Campaign 1 ( $n = 3$ )	18 $\pm$ 0.59 (17–19)	18 $\pm$ 2.3 (16–20)	36 $\pm$ 2.9 (34–38)	30 $\pm$ 4.4 (25–33)	14 $\pm$ 3.6 (10–17)	44 $\pm$ 8.0 (35–50)
Campaign 2 ( $n = 3$ )	19 $\pm$ 1.1 (18–20)	18 $\pm$ 0.16 (18–19)	38 $\pm$ 1.2 (36–39)	23 $\pm$ 7.1 (18–31)	3.7 $\pm$ 2.7 (2.0–7.0)	27 $\pm$ 9.8 (20–33)
Campaign 3 ( $n = 3$ )	20 $\pm$ 0.46 (19–20)	16 $\pm$ 1.2 (15–17)	36 $\pm$ 1.7 (34–36)	16 $\pm$ 3.4 (14–20)	13 $\pm$ 0.30 (13–14)	30 $\pm$ 3.8 (27–34)
Average	19 $\pm$ 0.95 (17–20)	18 $\pm$ 1.7 (15–20)	37 $\pm$ 2.7 (34–39)	23 $\pm$ 7.3 (14–33)	10 $\pm$ 5.8 (2.0–17)	34 $\pm$ 13 (20–51)

Note:  $n$  = Number of samples per sampling campaign in one monsoon. Values in parentheses indicate the concentration range.

1.5  $\mu\text{m}$  can survive for prolonged period of time in the atmosphere and have the ability to disperse to distant places (Calvo et al., 2013; Cincinelli et al., 2003).

### 3.2. Surfactants in marine aerosols

The average concentrations of anionic and cationic surfactants (as MBAS and DBAS) in aerosol samples collected from Port Dickson and Kuala Terengganu are shown in Table 2. Comparisons were made between the average concentration of surfactants in fine mode atmospheric aerosols ( $d < 1.5 \mu\text{m}$ ) and coarse mode atmospheric aerosols ( $d > 1.5 \mu\text{m}$ ) for the sampling area. Aside from the average, the concentrations of surfactant were compared between monsoons, namely from the southwest monsoon to the northeast monsoon. For the southwest monsoon, the average concentration of anionic surfactants as MBAS for the area of Port Dickson were recorded as being between 36 and 326  $\text{pmol m}^{-3}$ ; with the average concentrations of cationic surfactants as DBAS determined as between 23 and 80  $\text{pmol m}^{-3}$ . While for the northeast monsoon, the average concentrations of MBAS and DBAS were recorded at between 18 and 182  $\text{pmol m}^{-3}$ , and between 26 and 89  $\text{pmol m}^{-3}$  respectively.

For the sampling area of Kuala Terengganu, the average concentrations of anionic surfactants as MBAS were recorded as being between 87 and 97  $\text{pmol m}^{-3}$  on southwest monsoon. On the other hand, the average concentrations of cationic surfactants as DBAS were between 48

and 73  $\text{pmol m}^{-3}$ . While for the northeast monsoon, the concentrations of MBAS and DBAS are between 72 and 174  $\text{pmol m}^{-3}$ ; and 50  $\text{pmol m}^{-3}$  to 89  $\text{pmol m}^{-3}$ , respectively.

Overall results using  $t$ -test analyses showed that the concentrations of surfactants as MBAS in aerosols recorded at both areas were higher than those of DBAS ( $p < 0.05$ ). MBAS and DBAS concentrations in fine mode ( $d < 1.5 \mu\text{m}$ ) aerosols were greater ( $p < 0.05$ ) than the concentration of MBAS and DBAS in coarse mode ( $d > 1.5 \mu\text{m}$ ) aerosols. These findings correlate with the results from previous studies (Latif and Brimblecombe, 2004; Sukhapan and Brimblecombe, 2002; Wahid et al., 2013) which also recorded concentrations of surfactants as MBAS as being greater than surfactants as DBAS. According to Scott and Jones (2000) and Ying (2006), surfactants in the atmosphere are dominated by anionic surfactants due to the quantity of humic substances in atmospheric aerosols. The domination of anionic lipid, carboxylic acid and detergents, such as linear alkylbenzene sulfonate chain (LAS), from the ocean surface water as concurred by (Decesari et al., 2011), are also expected to contribute to the amount of anionic surfactants in atmospheric aerosols.

Further detailed analysis using a paired  $t$ -test demonstrated that there were significant differences ( $p < 0.05$ ) between surfactants concentrations (as MBAS and DBAS in the fine and coarse mode aerosols) recorded at Port Dickson and Kuala Terengganu. Similar statistical analyses also showed that there were significant differences between surfactants recorded during the southwest and northeast monsoons at

**Table 2**  
The average concentrations of MBAS and DBAS in atmospheric aerosols from different monsoon seasons.

Season	Particle type	Sampling Station			
		Port Dickson		Kuala Terengganu	
		MBAS ( $\text{pmol m}^{-3}$ )	DBAS ( $\text{pmol m}^{-3}$ )	MBAS ( $\text{pmol m}^{-3}$ )	DBAS ( $\text{pmol m}^{-3}$ )
Southwest monsoon (June–September)	Fine mode	160 $\pm$ 74 (84–326)	65 $\pm$ 9.4 (47–80)	91 $\pm$ 4.3 (88–97)	67 $\pm$ 7.5 (53–73)
	Coarse mode	73 $\pm$ 26 (36–129)	36 $\pm$ 6.5 (23–49)	91 $\pm$ 3.8 (87–96)	60 $\pm$ 7.1 (48–66)
Northeast monsoon (November–March)	Fine mode	93 $\pm$ 54 (40–182)	64 $\pm$ 10 (52–89)	112 $\pm$ 24 (87–141)	70 $\pm$ 10 (57–89)
	Coarse mode	30 $\pm$ 13 (18–52)	30 $\pm$ 2.8 (26–34)	109 $\pm$ 39 (72–174)	56 $\pm$ 3.6 (50–62)

Note: Values in parentheses indicate the concentration range.

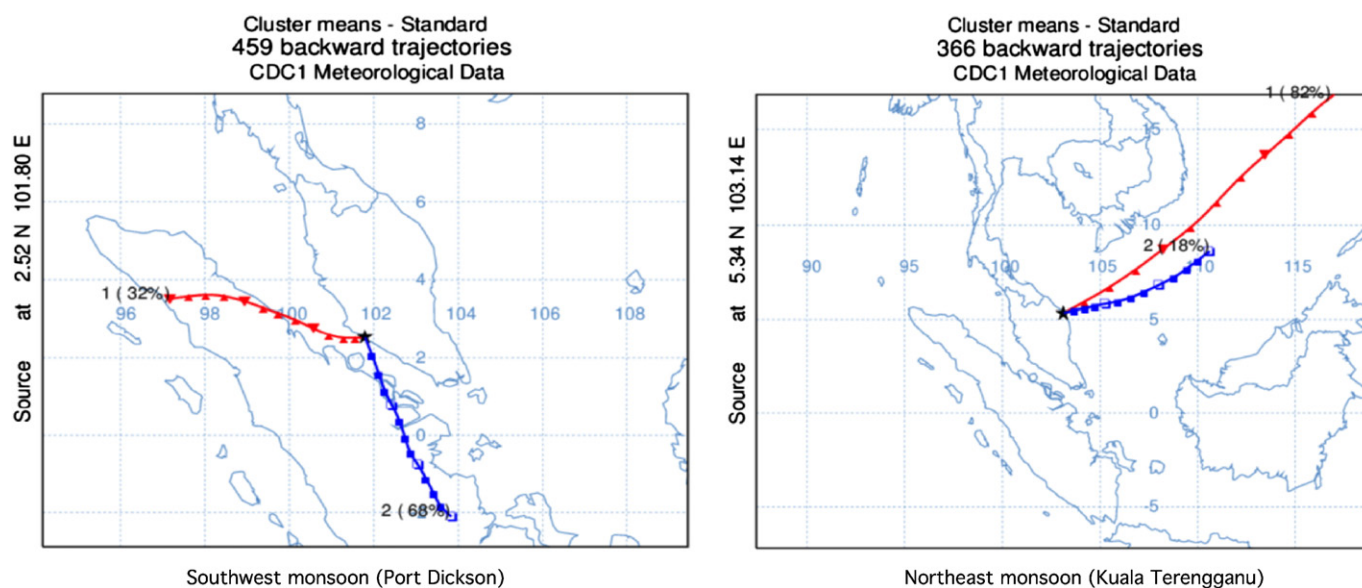


Fig. 2. Percentage of cluster backward trajectories to sampling stations during the southwest monsoon (Port Dickson) and the northeast monsoon (Kuala Terengganu).

both stations. The differences in surfactant concentrations between these two stations may be due to variations in their local surroundings and the possible sources of surfactants in atmospheric aerosols. The concentration of MBAS as an anionic surfactant was found to be higher during the southwest monsoon at Port Dickson when compared to Kuala Terengganu which is due to the influence of the wind effect. During the southwest monsoon, the prevailing wind flow was predominantly southwesterly from Sumatra. This usually contributes to the amount of biomass burning residue, where the residue usually contains greater quantity of humic-like substances consisting of surfactants (Latif et al., 2005). The 48 h backward trajectories developed for the sampling stations, according to the monsoons, and using HYSPLIT 4.9 software are shown in Fig. 2. As for northeast monsoon, the concentration of MBAS as an anionic surfactant was found to be higher at Kuala Terengganu. This is as a result of the wind predominantly originating from the South China Sea and heading towards the Malaysian Peninsula.

### 3.3. Source apportionment of surfactants using major elemental and anionic composition of aerosols

Table 3 shows the average concentration of anion and major element compositions in aerosols. The overall order of the average

concentration for the major elements is as follows: Na > Ca > K > Mg. Na recorded the highest value, compared to other major elements, with a concentration ranging from  $0.4 \pm 0.1 \mu\text{g m}^{-3}$  in coarse mode aerosols during the northeast monsoon in Port Dickson to  $3 \pm 0.5 \mu\text{g m}^{-3}$  in fine mode aerosols during the southwest monsoon at the same sampling area. The concentration of Na was also recorded as being at a higher level in fine mode aerosols from the sampling conducted at Kuala Terengganu ( $2 \pm 1 \mu\text{g m}^{-3}$ ). Whereas the other elements (Ca, K, Mg), were found to have significantly higher concentrations (paired *t*-test,  $p < 0.05$ ) in coarse mode aerosols collected in same sampling station during the northeast monsoon.

Overall, the order of anion concentrations for the two sampling stations are as follows;  $\text{SO}_4^{2-} > \text{NO}_3^- > \text{Cl}^- > \text{F}^-$ . The concentration of  $\text{SO}_4^{2-}$  ranged from  $1 \pm 0.7 \mu\text{g m}^{-3}$  as recorded in coarse mode aerosols during the southwest monsoon in Port Dickson to  $9 \pm 3 \mu\text{g m}^{-3}$  in fine mode aerosols during the southwest monsoon in Kuala Terengganu. The concentration of  $\text{NO}_3^-$  ranged from  $0.5 \pm 0.04 \mu\text{g m}^{-3}$  (coarse mode, Kuala Terengganu) during the southwest monsoon to  $2 \pm 1 \mu\text{g m}^{-3}$  (coarse mode, Port Dickson) during the northeast monsoon. The concentration of  $\text{Cl}^-$  was clearly higher in coarse mode aerosols with a concentration of between  $0.4 \pm 0.06 \mu\text{g m}^{-3}$  (fine mode, Port Dickson) during the southwest monsoon and  $4 \pm 1 \mu\text{g m}^{-3}$  (coarse mode, Kuala

Table 3

Average concentration ( $n = 9$ ) of major element and anion compositions in coarse and fine mode aerosol ( $\mu\text{g m}^{-3}$ ).

Elements	Sampling station		Kuala Terengganu						
	Port Dickson		Southwest monsoon				Northeast monsoon		
			Fine mode		Coarse mode				
	Fine mode	Coarse mode	Fine mode	Coarse mode	Fine mode	Coarse mode	Fine mode	Coarse mode	
Major elements									
Ca	$0.2 \pm 0.1$	$0.2 \pm 0.02$	$0.3 \pm 0.2$	$0.2 \pm 0.08$	$0.2 \pm 0.1$	$0.3 \pm 0.03$	$0.2 \pm 0.1$	$0.6 \pm 0.4$	
K	$0.1 \pm 0.02$	$0.3 \pm 0.04$	$0.2 \pm 0.04$	$0.3 \pm 0.1$	$0.2 \pm 0.1$	$1 \pm 0.07$	$0.1 \pm 0.02$	$1 \pm 0.6$	
Mg	$0.05 \pm 0.01$	$0.1 \pm 0.05$	$0.06 \pm 0.02$	$0.2 \pm 0.2$	$0.05 \pm 0.02$	$0.2 \pm 0.08$	$0.05 \pm 0.02$	$2 \pm 1$	
Na	$3 \pm 0.5$	$0.4 \pm 0.2$	$1 \pm 0.04$	$0.4 \pm 0.1$	$2 \pm 0.05$	$0.4 \pm 0.1$	$2 \pm 1$	$2 \pm 1$	
Anions									
$\text{F}^-$	$0.08 \pm 0.06$	$0.07 \pm 0.02$	$0.09 \pm 0.04$	$0.08 \pm 0.01$	$0.10 \pm 0.05$	$0.07 \pm 0.02$	$0.1 \pm 0.03$	$0.06 \pm 0.03$	
$\text{Cl}^-$	$0.4 \pm 0.06$	$3 \pm 2$	$0.4 \pm 0.2$	$2 \pm 1$	$0.5 \pm 0.04$	$3 \pm 0.9$	$0.8 \pm 0.3$	$4 \pm 1$	
$\text{NO}_3^-$	$0.8 \pm 0.2$	$0.8 \pm 0.2$	$0.9 \pm 0.5$	$2 \pm 1$	$0.7 \pm 0.02$	$0.5 \pm 0.04$	$0.3 \pm 0.2$	$2 \pm 0.7$	
$\text{SO}_4^{2-}$	$5 \pm 2$	$1 \pm 0.7$	$7 \pm 3$	$2 \pm 0.7$	$9 \pm 3$	$4 \pm 2$	$3 \pm 2$	$9 \pm 6$	

**Table 4**  
The factor loading for fine and coarse mode aerosol after varimax rotation using PCA – Port Dickson.

Elements	Fine mode ( $d < 1.5 \mu\text{m}$ )			Coarse mode ( $d > 1.5 \mu\text{m}$ )		
	F1	F2	F3	F1	F2	F3
	Motor vehicle/combustion of biomass	Earth crust/motor vehicle	Marine aerosol	Soil/earth crust	Sea spray/marine aerosol	Motor vehicle
F <sup>-</sup>	-0.137	<b>0.816<sup>a</sup></b>	0.170	0.459	-0.713	0.078
Cl <sup>-</sup>	0.060	-0.319	0.363	0.152	<b>0.876<sup>a</sup></b>	-0.221
NO <sub>3</sub> <sup>-</sup>	<b>0.929<sup>a</sup></b>	0.046	0.229	-0.104	-0.253	<b>0.923<sup>a</sup></b>
SO <sub>4</sub> <sup>2-</sup>	0.262	-0.129	<b>0.843<sup>a</sup></b>	0.325	0.517	0.679
Ca	<b>0.788<sup>a</sup></b>	-0.209	0.124	0.426	0.616	0.308
K	<b>0.838<sup>a</sup></b>	0.315	0.153	<b>0.9239<sup>a</sup></b>	0.003	0.165
Mg	0.267	<b>0.854<sup>a</sup></b>	-0.054	<b>0.955<sup>a</sup></b>	0.150	-0.135
Na	-0.195	-0.289	<b>-0.810<sup>a</sup></b>	<b>0.964<sup>a</sup></b>	0.019	0.040
Eigen value	2.965	1.672	1.116	3.413	1.884	1.450
Variability (%)	37.058	20.905	13.945	42.662	23.549	18.119
Cumulative (%)	37.058	57.963	<b>71.907</b>	42.662	66.210	<b>84.329</b>

<sup>a</sup> Factors in bold indicated as strong factor loading.

Terengganu) during the northeast monsoon. The concentration of F<sup>-</sup> was recorded at the lowest concentration with little difference (paired *t*-test,  $p > 0.05$ ) between modes or monsoons.

The results of the varimax rotated factor analysis for total aerosols using major element and anions analyses at Port Dickson and Kuala Terengganu are shown in Tables 4 and 5 respectively. Three key factors were identified in the overall data set for fine mode aerosols (71.91% of total variance) and coarse mode aerosols (84.33% of total variance) at Port Dickson. The first factor (F1) for fine particles ( $d < 1.5 \mu\text{m}$ ), with 37.06% of the total variance, showed high factor loading for NO<sub>3</sub><sup>-</sup>, Ca and K and can be categorized as originating from motor vehicles and biomass burning (Amil et al., 2016; Ee-Ling et al., 2015; Khan et al., 2016). The second factor was found to show a high factor loading for F<sup>-</sup> and Mg with 20.91% of the total variance. Studies by Zhang et al. (2007) demonstrated that Mg is an element which is present in soil and the earth's crust. While the third factor (F3), with a total variance of 13.95%, was categorized as resulting from marine aerosols for which SO<sub>4</sub><sup>2-</sup> and Na elements showed a high factor loading. According to a study by Querol et al. (2002), marine aerosols was composed of the elements Na and Cl<sup>-</sup> ion and can be traced on a particulate size of 1.5  $\mu\text{m}$ –11  $\mu\text{m}$ . Moreover, it was found that the fumes released by sea-faring ships also contributed to the presence of SO<sub>4</sub><sup>2-</sup> concentrations in aerosols (Calvo et al., 2013).

For coarse particles ( $d > 1.5 \mu\text{m}$ ), a total of three main components were obtained from this analysis with 84.33% of total variance. The first factor (F1) recorded 42.66% of the total variance and is categorized as having soil and the earth's crust as sources. It was also shown to have a high factor loading with the elements K, Mg and Na which are among the indicators for aerosols originating from the soil and earth's crust

(Karanasiou et al., 2009; Khan et al., 2010). The second factor (F2) showed the Cl<sup>-</sup> element as originating from sea spray and marine aerosols spray (Foltescu et al., 1996), recording a variance of 23.55%. While the high factor loading of NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> places the third factor (F3) as originating from motor vehicles and industrial activities with 18.12% of the total variance.

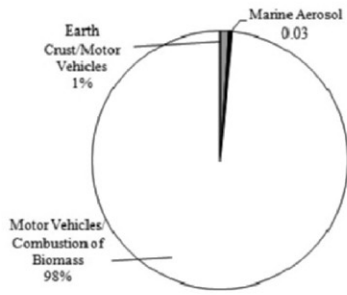
As for the sampling area Kuala Terengganu, three important factors were identified in the data set for fine atmospheric aerosols sized (81.56% of total variance) and coarse-sized atmospheric aerosols (91.70% of total variance). The first factors (F1) for fine particles ( $d < 1.5 \mu\text{m}$ ) had 41.43% of the total variance, showed high factor loading for NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and Na and can be categorized as originating from secondary and marine aerosol emissions (Amil et al., 2016; Mustaffa et al., 2014; Wahid et al., 2013). Emissions from small boats and ships can be included in this factor Calvo et al. (2013). The second factor (F2) was categorized as originating from soil and the earth's crust and showed a high factor loading for the elements F, Ca and Mg with 26.98% of the total variance. While the third factor (F3) was noted to originate from biomass combustion with a total variance of 13.14%, for which the element K showed a high factor loading (Amil et al., 2016; Dai et al., 2013; Khan et al., 2016; Mustaffa et al., 2014; Rahman et al., 2011).

For coarse particles ( $d > 1.5 \mu\text{m}$ ) in Kuala Terengganu, three main components were also obtained from this analysis with 91.70% of the total variance. The first factor (F1) recorded 72.88% of the total variance and its sources were categorized as being soil and the earth's crust. It also showed high factor loadings of SO<sub>4</sub><sup>2-</sup>, Mg and Ca. The second factor (F2) showed the Cl<sup>-</sup> element, categorized as originating from sea spray and marine aerosols, recording the variance of 11.12%. The third factor

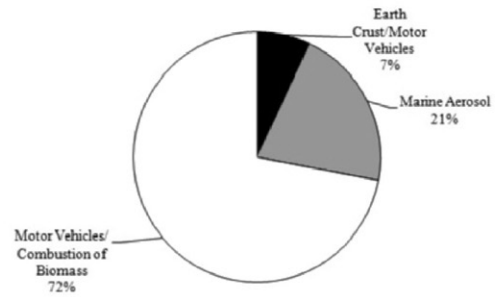
**Table 5**  
The factor loading for fine and coarse mode aerosol after varimax rotation using PCA – Kuala Terengganu.

Elements	Fine mode ( $d < 1.5 \mu\text{m}$ )			Coarse mode ( $d > 1.5 \mu\text{m}$ )		
	F1	F2	F3	F1	F2	F3
	Secondary aerosol/marine aerosol	Soil/Earth crust	Combustion of biomass	Soil/earth crust	Sea spray/marine aerosol	Combustion of biomass
F <sup>-</sup>	0.309	<b>0.795<sup>a</sup></b>	-0.320	-0.724	-0.125	-0.426
Cl <sup>-</sup>	-0.448	0.398	-0.428	0.161	<b>0.968<sup>a</sup></b>	0.099
NO <sub>3</sub> <sup>-</sup>	<b>0.884<sup>a</sup></b>	-0.054	0.291	0.676	0.561	0.378
SO <sub>4</sub> <sup>2-</sup>	<b>0.916<sup>a</sup></b>	0.103	0.137	<b>0.933<sup>a</sup></b>	0.179	0.150
Ca	-0.359	<b>0.826<sup>a</sup></b>	0.179	<b>0.885<sup>a</sup></b>	0.158	0.380
K	0.165	-0.060	<b>0.915<sup>a</sup></b>	0.282	0.096	<b>0.933<sup>a</sup></b>
Mg	-0.036	<b>0.887<sup>a</sup></b>	-0.095	<b>0.798<sup>a</sup></b>	0.325	0.471
Na	<b>-0.939<sup>a</sup></b>	0.149	0.061	0.593	0.415	0.633
Eigen value	3.315	2.159	1.052	5.831	0.889	0.616
Variability (%)	41.431	26.982	13.144	72.881	11.115	7.700
Cumulative (%)	41.431	68.413	<b>81.556</b>	72.881	83.996	<b>91.696</b>

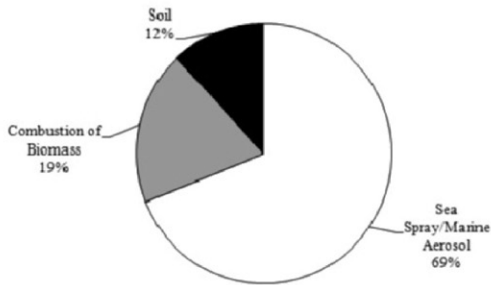
<sup>a</sup> Factors in bold indicated as strong factors.



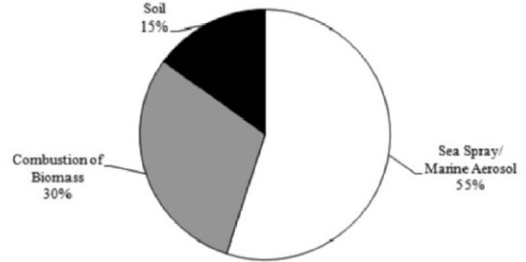
a) Sources of MBAS in fine mode aerosol at Port Dickson



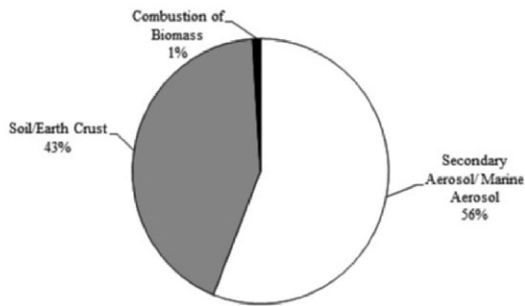
b) Sources of DBAS in fine mode aerosol at Port Dickson



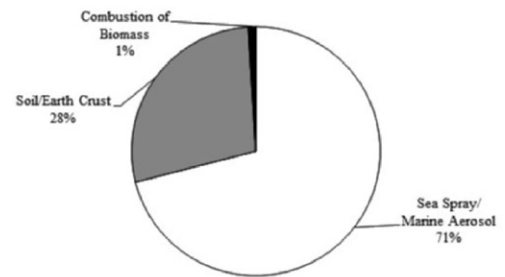
c) Sources of MBAS in coarse mode aerosol at Port Dickson



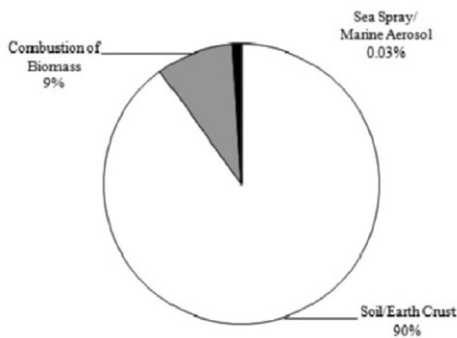
d) Sources of DBAS in coarse mode aerosol at Port Dickson



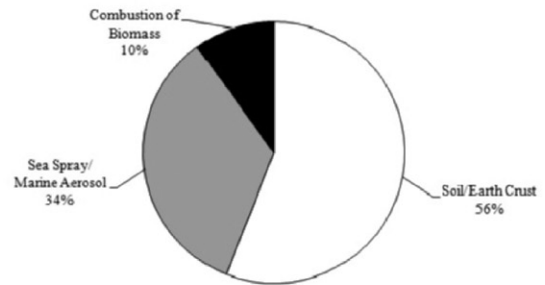
e) Sources of MBAS in fine mode aerosol at K. Terengganu



f) Sources of DBAS in fine mode aerosol at K. Terengganu



g) Sources of MBAS in coarse mode aerosol at K. Terengganu



h) Sources of DBAS in coarse mode aerosol at K. Terengganu

Fig. 3. Source contributions of surfactants in atmospheric aerosols.



(F3) had a significant factor loading of K, derived from the combustion of biomass, with a total variance of 7.70%.

Analysis using PCA-MLR showed that the concentration of surfactants for the Port Dickson sampling area was affected by six main factors, namely: biomass combustion, motor vehicle emissions, the earth's crust, soil, marine aerosols and sea spray. Referring to Fig. 3(a), the main sources contributing to the presence of the surfactant MBAS in fine particulates were motor vehicles and biomass burning, which dominated with a percentage of 98%. This was followed by the earth's crust and marine aerosols, each with a share of 1% and 0.03% respectively. DBAS surfactants are shown in Fig. 3(b). The main factors contributing to surfactants in fine particulates were motor vehicles and the combustion of biomass (72%), followed by marine aerosols (21%) and the earth's crust (7%). Motor vehicle emissions and the burning of biomass were shown to be major sources of pollution and as Port Dickson is a well-known tourist area, a greater number of vehicles are consequently used in this area. This site is also located close to the city centre and results clearly indicate the density of vehicles as used by locals. In addition, Port Dickson is situated in close proximity to Sumatra, Indonesia which as a result, leads to the study area being exposed to biomass burning emissions which are carried by the wind, especially during the southwest monsoon. According to previous studies, the sources of air pollution in developing countries, such as Malaysia, are smoke emissions from vehicles (Atash, 2007), industrial activity (Azmi et al., 2010) and also biomass combustion (Chantara, 2012).

For coarse mode aerosols, as shown in Fig. 3(c), the main factor contributing to surfactants as MBAS was from marine aerosols which recorded the percentage of 69% followed by 19% from biomass burning and 12% from soil. The same trend applied to the source apportionment of surfactants as DBAS (Fig. 3(d)) which showed that sources such as marine sea spray and aerosols recorded a high value of 55% followed by biomass combustion (30%) and soil at 15%. Factors such as sea spray and marine aerosols are notably more dominant than other factors because of the sampling station's close proximity to the sea.

Fig. 3(e) and Fig. 3(f) show the distribution of polluting factors contributing to Kuala Terengganu's sampling area for both surfactants as MBAS and DBAS in fine particulates. Referring to Fig. 3(e), the source of surfactants for fine mode particles was dominated by secondary aerosol and marine aerosol (56%) while for coarse mode particles the source was dominated by sea spray and marine aerosol (71%). Kuala Terengganu is the focal point of the state of Terengganu, which is near to a coastal area where fishing is a major activity. Emissions from vehicles and fishing boats are one source of air pollutants within the sampling area. Anthropogenic sources are usually present at the size of the fine particulates. According to Jonathan et al. (1997), in general it can be said that fine particulate matter (<2 µm) is produced as a result of combustion processes, including vehicle exhaust and gas exchange processes. As for coarse particulate matter, the source apportionment for surfactants MBAS and DBAS for the Kuala Terengganu sampling site is shown in Fig. 3(g) and Fig. 3(h). Sources of soil and the earth's crust recorded the largest percentage of surfactants MBAS and DBAS, respectively 90% and 56%. This is due to the presence of elements such as Ca, K, and Mg which were more dominant in coarse particulates.

In summary this study has shown that the concentration of surfactants at the two sampling sites was dominated by anionic surfactants as MBAS, compared with cationic surfactants as DBAS, in both seasons, the southwest and northeast monsoons. The average concentration of surfactants in aerosols was found to be at the highest level at Port Dickson during the southwest monsoon, with the value of 160.22 pmol m<sup>-3</sup> in fine mode aerosols. Meanwhile, the highest average concentration of surfactants for the northeast monsoon, at 112.40 pmol m<sup>-3</sup>, was recorded at Kuala Terengganu.

PCA-MLR shows that the source apportionment of surfactants at Port Dickson in fine mode aerosols was dominated by motor vehicle emissions and combustion processes while sea spray and marine aerosols were the major contributors to surfactants in coarse mode aerosols.

This result is due to Port Dickson being located close to industrial areas with a high volume of motor vehicles. As for Kuala Terengganu, the source apportionment of surfactants in fine mode aerosols was dominated by secondary and marine aerosol emissions. At the same time, soil and the earth's crust were the major contributor to surfactants in coarse mode aerosols. For future research, a comprehensive study of the effects of meteorological parameters on the concentrations of surfactants should be undertaken. In addition, the specification species of surfactants needs to be analyzed in more detail so as to gain a better understanding of surfactants.

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