COMPOSITION OF SURFACTANTS AND IONIC ELEMENTS FROM DIESEL AND PETROL EXHAUST PARTICULATE MATTER

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Abstract. Emission from vehicular exhaust is an important source of particulate matter (PM) especially in the urban area. This study was conducted to determine the compositions of surfactants and ionic elements from diesel and petrol exhaust particulate matter (exhaust soot). The exhaust PM sample was collected using soft paint brush from different types of petrol and diesel motor vehicles, during dry days. Methylene Blue Active Substances (MBAS) which are anionic surfactant and Disulphine Blue Active Substances (DBAS) which are cationic surfactant were determined using UV-Vis Spectrophotometer, based on colorimetric method. Whereas, ion chromatography (IC) was used to determine ionic elements from the exhaust PM samples. In addition, the Field Emission Scanning Electron Microscope (FESEM) was used to study the morphological structure of exhaust PM. Results indicated that the concentration of surfactants were dominated by MBAS compared to DBAS for all types of motor vehicles studied. In addition, heavy duty vehicles/ lorries showed the highest concentration of MBAS (1.50 ± 0.10 µmol g^{-1}) and DBAS (0.28 ± 0.04 µmol g^{-1}). The ionic elements compositions followed the trend of $SO_4^{2-} > Ca^{2+} > Na^{+} > K^{+} > NO_3^{-} > Cl^{-} > NH_4^{+} > Mg^{2+} > F^{-}$ for all types of soot samples. Exhaust PM from petrol and diesel automobiles appear as agglomerates of many fine spherical primary particles in the form of cluster soot particles, according to FESEM image. The emission from diesel and petrol vehicles should be a concern as they could pose negative effects to human health as well as towards environment.

Keywords: MBAS, DBAS, ionic composition, air pollution, vehicle emission

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Introduction

In a developing country, the air pollution situation is getting more critical especially in urban areas. The increase in population due to the development of urban areas has resulted in a high road traffic density. In Kuala Lumpur city centre of Malaysia, road traffic growth has increased with average annual growth of 5.03% [1-3]. Generally, motor vehicles may cause the air pollution problem due to exhaust emissions as well as brakes and tyres abrasion [1, 4-5]. One of the highest pollutants in the urban atmosphere is particulate matter (PM). The control of atmospheric PM is an important task, because of its high level may affect human health and influence the climate change system [6].

PM in the ambient atmosphere, which contains surface-active agents (surfactants), is believed to have more impact on climate change due to its physical and chemical characteristics [7-9]. In the atmosphere, surfactants can come from both anthropogenic (manmade) and natural sources. Surfactants in ambient air might be originated from the burning of biomass, the sea-surface microlayer, humic compounds, emission from motor vehicles, as well as combustion activities [10-11]. Surfactants might increase solubility in the aqueous phase and reducing surface tension, and resulted in the formation of cloud albedo [12]. On the other hand, surfactants might also affect human health, associated with eyes and lung infection [13].

Due to urbanization and rapid economic, which might increase the traffic volume, motor vehicles emission was projected to be the primary source of PM in the urban environment [14-15]. As a result, numerous research on the effects of transportation on atmospheric aerosol concentrations have been conducted [14, 9]. However, just a few research have looked into the impact of automotive emissions (exhaust particulate matter) on surfactant levels in atmospheric aerosol. This study seeks to establish the composition of surfactants in exhaust particulate matter from petrol and diesel motor vehicles, as well as to investigate the morphological structure of exhaust PM. This research is a continuation of a study by Wahid et al. [14] who discovered that motor vehicles constitute the primary source of surfactants in the atmosphere.

Materials and Methods

Sampling procedures. For diesel and petrol exhaust particulate matter, five samples were collected randomly from several types of vehicles which used diesel fuel (buses, medium lorries and heavy lorries/ construction machineries) and petrol fuel (motorcycles and cars) (total samples, n=25). The types of selected vehicles were chosen based on the road traffic volume data for Kuala Lumpur city centre [3]. Table 1 shows the traffic composition by vehicle type for the nearest census station in Kuala Lumpur. A soft paintbrush was used in sample collection during dry weather. The samples were kept in a glass vial with a Teflon screw cap until further analysis.

Table 1. Traffic composition by vehicles type (Kuala Lumpur).

		Percentage of vehicles composition (%)						
16-hours traffic	**Peak hour**	**Cars**	**Small Van/ Utilities**	**Medium lorries**	**Heavy Lorries/ construction Machineries**	**Buses**	**Motorcycles**	**Total**
290,772	7.00 – 8.00 pm	58.0	9.8	6.0	0.8	0.5	24.8	100

Sample extraction. 0.05 g of exhaust from diesel and petrol automobiles was placed in a clean glass vial to extract the samples. The glass vial was then filled with 40 mL of deionized water. An ultrasonic agitator was used to sonicate the material for 45 minutes. After that, the sample solution was filtered using cellulose acetate filter paper with a pore size of 0.2 µm. Finally, the filtered solution was poured into a 100 ml volumetric flask and diluted with deionized water. The sample was kept at 4 ˚C until further analysis, as conducted by Razak et al. [16].

Surfactant analysis. The colorimetric method was used to analyse both anionic and cationic surfactants, known as Methylene Blue Active Substances (MBAS) and Disulphine Blue Active Substances (DBAS), respectively [17-18]. The calibration curves for Sodium Dodecyl Sulphate (SDS) as MBAS standard and Zephiramine as DBAS standard were prepared in the range of 0.05 – 2.00 µM. MBAS analysis required reagents such as methylene blue, alkaline buffer, chloroform, and acidic methylene blue solution, whereas DBAS analysis required disulphine blue, acetate buffer, and chloroform solution, as undertaken by previous researchers [19,4,14]. After analysis, the light absorbance of the extracted material was measured using a UV-Vis spectrophotometer (Labomed) at 650 nm for MBAS and 628 nm for DBAS. For this analysis, the method detection limit for MBAS and DBAS was 0.05 µM. MBAS had an average recovery of 87%, while DBAS had an average recovery of 89%, in this study.

Ionic elements analysis. The anions were determined using ion chromatography (Metrohm, model 881 Compact IC Pro) using 3.2 mmol L^{-1} Na_2CO_3 as the eluent and a Metrosep A-Supp 5-150/4.0 column. The flow rate was fixed to 0.7 mL min^{-1}, and standard elements (Merck) of seven distinct anions were employed. Only four elements (F^-, Cl^-, NO_3^-, and SO_4^{2-}) were found in this investigation, with a method detection limit of 0.002 to 0.005 g m^{-3}. The average recoveries for all anions measured in this study were in the range of 98-118%. Whereas, Ion Chromatography (Dionex, IC model ICS-1100) was used with 20 mM methanesulfonic acid as the eluent with the flow rate of 1.0 mL min^{-1} was used to determine cations. IonPac CS12A 4 mm column (Dionex) was used as the column, and the type of suppressor was CSRS 300 4 mm (Dionex). The instrument was calibrated in the range of 1 to 500 ppb from diluting the multi-element stock standard solution (Dionex, The Six Cation-II Standard). The method detection limits for five elements (Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}) in this investigation ranged from 0.002 to 0.004 g m^{-3}. All of the cations examined had the recoveries ranging from 85 to 114%.

FESEM analysis. The field-emission scanning electron microscopy (FESEM, Hitachi SU 8020) coupled with Energy Dispersion System (EDS, Horiba Xmax Model) was used to examine the surface morphology of petrol and diesel exhaust particulate matter. To adhere the sample, the exhaust PM was first affixed directly to the stubs using a double-sided adhesive film. Then, the sputter coater was used to coat the sample with platinum (Pt) coating. This sputter coater used an electric field and argon gas. The Pt atom was settled onto the surface of the sample producing a thin Pt coating. Finally, a 5 kV accelerating voltage was used on the FESEM. Simultaneously, the images were modified with a specific contrast to brightness ratio to provide the best image, as done by Razak et al. [16].

Statistical techniques. After all the data was in a normal distribution, several statistical analyses were conducted for descriptive statistical analysis. The data collected were statistically analyzed using XLSTAT version 2019 (AddinSoft, Inc, NY).

Quality assurance/ quality control. Before being used, all instruments used in the sampling and laboratory analysis were calibrated. Because of the degree of surfactants that could affect the results, detergents and soap were not allowed in the cleaning operations. All glassware was hexane-washed, then acetone-rinsed before being rinsed again with deionized water. To eliminate organic contaminants, all of the glass vials were soaked in 20% HNO_3 overnight before being heated in a furnace (500 °C) for 5 h. Method detection limit (MDL) and recovery test were conducted for each laboratory analysis. In order to maintain the greatest precision, all laboratory analysis procedures were repeated in triplicate for each sample.

Results and Discussion

MBAS and DBAS in exhaust particulate matter. Table 2 indicates the average concentration of surfactants as MBAS and DBAS in diesel and petrol exhaust particulate matter. From the result obtained, diesel vehicles show a greater amount of surfactants concentration than petrol vehicles. However, statistical analysis showed no significant differences ($p > 0.05$) between surfactants in diesel and petrol vehicles. Whereas, MBAS always showed higher value than DBAS for each vehicle studied with significant difference ($p < 0.05$). Exhaust PM sample collected from diesel vehicles consists of buses, medium lorries, and heavy lorries have the average MBAS and DBAS value of 1.40 ± 0.07 µmol g^{-1} and 0.32 ± 0.04 µmol g^{-1} respectively. Among all the diesel vehicles, heavy lorries/construction machineries show the highest value of MBAS (1.50 ± 0.10 µmol g^{-1}) and DBAS (0.38 ± 0.04 µmol g^{-1}) compared to other types of vehicles. This may be due to construction machinery, and heavy trucks, which do not emphasize the engine maintenance compared to buses which are public transport vehicles. On 18 October 2019, Malaysia has launched the National Transport Policy (NTP) 2019-2030, which aims to develop a secure and sustainable transport sector, including maintenance for public transport [20]. Lack of maintenance will result in the engine operating less properly and cause increased emissions of pollutants from the exhaust. The average concentration of surfactants in exhaust PM from this study were 32% and 70% higher for diesel and petrol vehicle, respectively, compared to the study by Latif [21], who obtained samples of diesel and petrol vehicle soot in the UK. This might be resulted from different maintenance frequency or different engine oil used in the vehicles that were chosen as the research samples.

Several types of surfactants have been utilized in vehicle engines and gear oils to prevent and decrease the formation of deposits caused by the degradation process of lubricating oils and fuels in the automotive sector [22]. This combustion process may contribute to greater suspended particle emissions via exhaust particulate matter and gases, resulting in high levels of surfactants in urban aerosols, particularly near busy traffic regions [23].

For petrol vehicle exhaust, particle samples were collected from two types of vehicle groups namely motorcycles, and cars. The results for the concentrations of surfactants as MBAS and DBAS in petrol vehicle exhaust PM are shown in Table 2. From these results, MBAS has recorded higher readings than DBAS for all types of vehicle groups studied. Cars recorded higher MBAS and DBAS (1.27 ± 0.04 µmol g^{-1} for MBAS, 0.23 ± 0.09 µmol g^{-1} for DBAS) compared to motorcycles (1.15 ± 0.02 µmol g^{-1} for MBAS, 0.21 ± 0.04 µmol g^{-1} for DBAS).

The amount of MBAS and DBAS in vehicle soot is likely to be influenced by oxidation processes in engines and additional detergents [21]. According to Resitoglu [24], unpredictable operating circumstances and incomplete hydrocarbon combustion in fuel are the most likely causes of excessive surfactant levels. Furthermore, excessive air pollution has resulted from congested traffic areas, particularly in urban areas due to high number of motor vehicles during peak hours [16, 25]. Thus, regular usage of surfactants in engine oil may produce high PM emissions through exhaust, which coincidentally contribute to high surfactant levels in congested areas.

Table 2. Average compositions MBAS and DBAS in diesel and petrol exhaust particulate matter

Exhaust PM	**Types of Vehicles**	**MBAS µmol g^{-1}**	**DBAS µmol g^{-1}**
Diesel	Buses	1.34 ± 0.05	0.30 ± 0.04
	Medium Lorries	1.37 ± 0.05	0.27 ± 0.03
	Heavy lorries/ Construction machineries	1.50 ± 0.10	0.38 ± 0.04
Petrol	Motorcycles	1.15 ± 0.02	0.21 ± 0.04
	Cars/taxis	1.27 ± 0.04	0.23 ± 0.09

Ionic composition from aerosol and exhaust particulate matter. Table 3 and Table 4 show the average ionic concentrations of exhaust PM samples from diesel and petrol vehicles, respectively. The ionic elements followed the trend of SO_4^{2-} > Ca^{2+} > Na^+ > K^+ > NO_3^- > Cl^- > NH_4^+ > Mg^{2+} > F^- for all types of exhaust PM samples. SO_4^{2-} is the most dominant ion for the three types of vehicle groups compared to the others. This coincides with the hypothesis that SO_4^{2-} ions in atmospheric aerosols predominantly come from motor vehicle emissions through incomplete combustion [26-27, 21, 6]. Ca^{2+} and Na^+ concentration in this study was found quite high and similar to other research [14, 21, 28] which indicated that the concentration of these ions was usually related to emissions from motor vehicles.

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Table 3. Ionic composition for exhaust PM in diesel vehicles (mg g^{-1}).

Ionic element	Buses	Medium Lorries	Heavy Lorries/Construction Machineries
F^-	0.05	0.04	0.03
Cl^-	0.84	0.96	0.94
NO_3^-	0.96	0.90	0.94
SO_4^{2-}	8.22	8.19	8.31
Na^+	3.71	3.39	3.56
NH_4^+	0.69	0.71	0.76
K^+	2.36	2.55	2.76
Mg^{2+}	0.21	0.22	0.21
Ca^{2+}	4.54	4.03	4.49

Table 4. Ionic composition for exhaust PM in petrol vehicles (mg g^{-1}).

Ionic element	Motorcycles	Cars/ Taxis
F^-	0.04	0.04
Cl^-	0.70	0.87
NO_3^-	0.86	0.94
SO_4^{2-}	3.02	3.68
Na^+	1.12	1.57
NH_4^+	0.32	0.25
K^+	0.39	0.45
Mg^{2+}	0.24	0.27
Ca^{2+}	2.61	2.72

Similar to soot from diesel vehicle exhaust, SO_4^{2-} in petrol vehicles is the most dominant ion compared to other ion compositions, indicating that SO_4^{2-} in atmospheric aerosols mostly comes from motor vehicle emissions [21, 26], followed by Ca^{2+} ions and Na^+. Figure 1 shows the average composition of ionic elements from diesel and petrol exhaust PM.

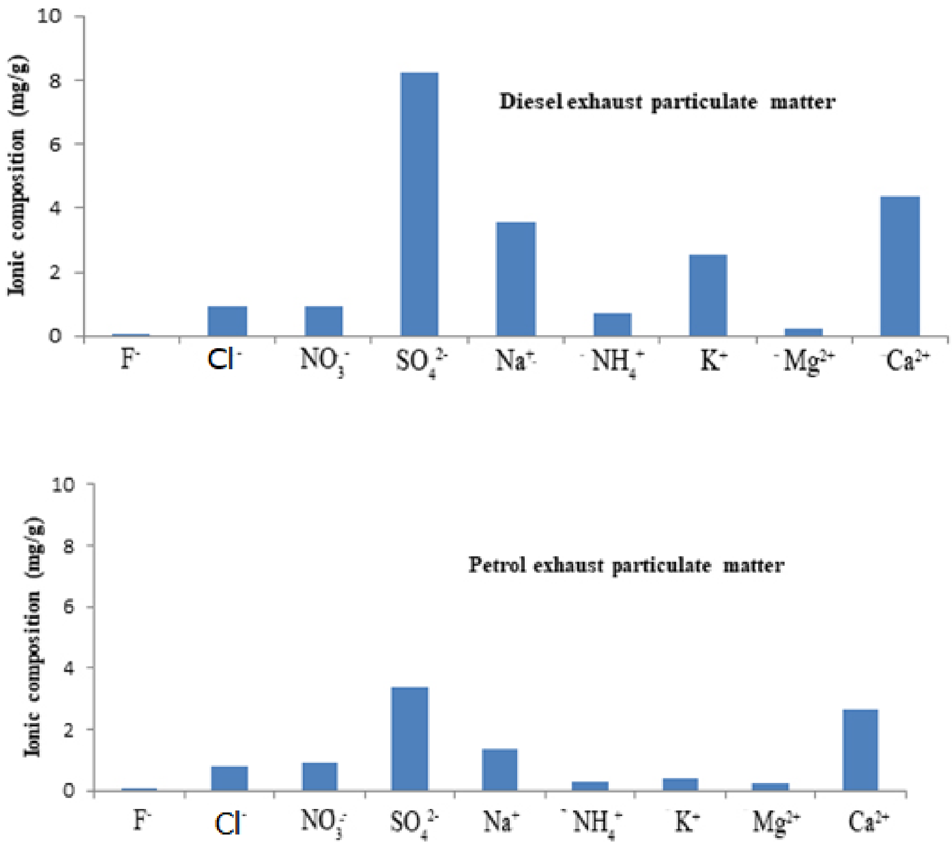

Figure 1. Average composition of ionic elements from diesel and petrol exhaust particulate matter (mg/g)

Morphological study of exhaust particulate matter. Figures 2 and 3 show the surface morphological characteristics of exhaust PM from diesel and petrol automobiles, respectively. The distinctive shape of exhaust soot particles distinguishes them from other particle (such as ambient PM, road dust), as revealed by FESEM images. Exhaust PM from both diesel and petrol vehicles was found as cluster soot particles, which are agglomerates of many fine spherical primary particles. Simple and chainlike soot particles were also seen. The findings are consistent with study by Razak et al. [16, 29]. The size of a soot sample from a diesel vehicle can be up to 0.1 µm (Figure 2), but the size range for a soot sample from a petrol vehicle is between 25 to 300 nm (Figure 3). According to Anake et al. [30], the agglomeration of carbon particles was due to incomplete combustion of fossil fuel, which can be found in the vehicles exhaust. In addition, heating system may also contribute to the amount of motor vehicles' exhaust PM. This situation may result to the large amount of fine particulate matter in the ambient air especially in urban area [14].

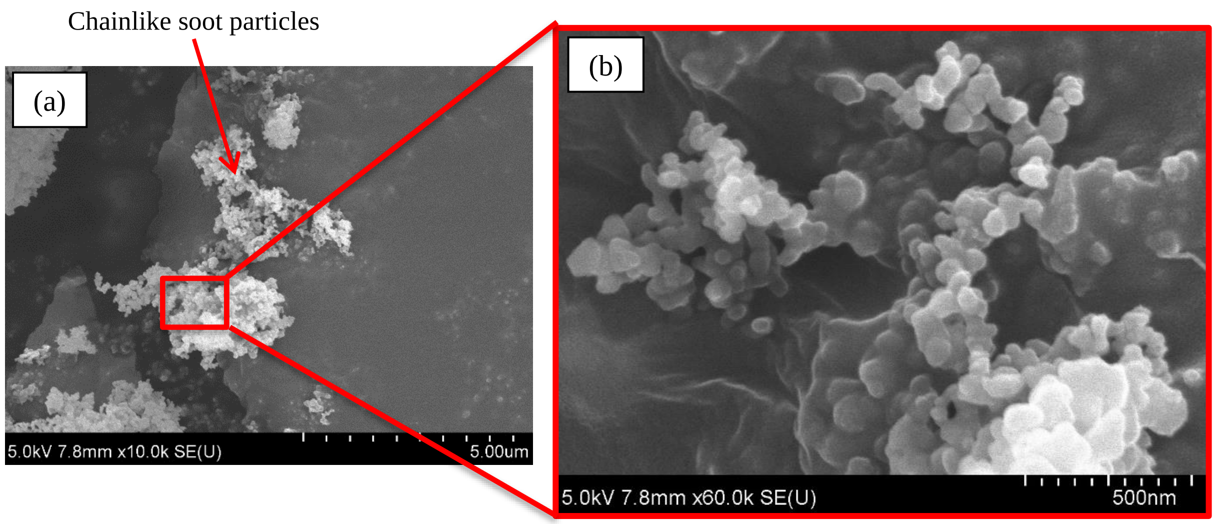

Figure 2. Morphological characteristic of exhaust PM from diesel vehicles (a) low magnification and (b) high magnification.

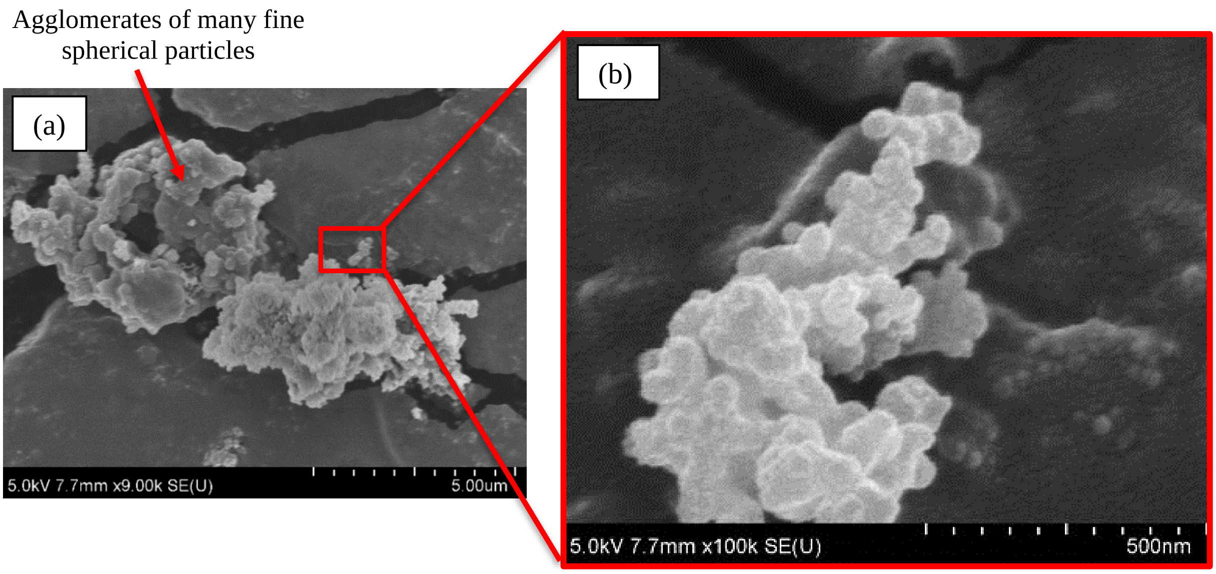

Figure 3. Morphological characteristics of exhaust PM from petrol vehicles (a) low magnification and (b) high magnification.

Conclusion

The compositions of surfactants were determined from exhaust particulate matter in diesel and petrol vehicles. Anionic surfactants (MBAS) show a high surfactant concentration compared to cationic surfactants (DBAS). Heavy lorries and cars demonstrated high surfactant emissions for diesel and petrol vehicles. For all types of exhaust PM samples, the ionic elements followed the pattern of $SO_4^{2-} > Ca^{2+} > Na^+ > K^+ > NO_3^- > Cl^- > NH_4^+ > Mg^{2+} > F^-$. From FESEM analysis, exhaust PM is found as cluster soot particles, which are agglomerates of many fine spherical primary particles, as well as simple and chainlike soot particles. In conclusion, a high amount of surfactants in atmospheric aerosol might come from exhaust PM. Frequent engine service and choosing high-quality oil can help reduce the emission of air pollutants in the atmosphere. For future studies, it is recommended to include the manufacturing year of each vehicle (samples) and the frequency of vehicles' maintenance activity to correlate these issues with the amount of surfactants in exhaust PM.

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Authors Contributions

All authors contributed toward data analysis, drafting and critically revising the paper and agree to be accountable for all aspects of the work.

Disclosure of Conflict of Interest

The authors have no disclosures to declare.

Compliance with Ethical Standards

The work is compliant with ethical standards.

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