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Characterization of Particulate Matter Collected at Mysore City Roadways in Association with Urban Traffic Condition

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Authors' contributions

This work was carried out in collaboration between both authors. Author AKF designed and performed the field-work experiments and data collection, analyzed the data, performed the statistical and computational analyses, managed the literature searches, and wrote the manuscript. Author GVV conceived the framework of the study, provided the sampling instruments, managed the laboratory analyses and literature searches, wrote and edited the manuscript. Both authors read and approved the final manuscript.

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ABSTRACT

Aim: To identify the source of particulate matter (PM) emissions in Mysore urban city roadways by characterizing PM of different aerodynamic diameters ($PM_{2.5}$ and PM_{10}) using various advanced techniques and finding their correlation with site traffic condition.
Place and Duration of Study: The study was conducted in urban area of Mysore city, Karnataka, India, from 2014 to 2017.
Methodology: Emissions of $PM_{2.5}$ and PM_{10} were estimated using mathematical model incorporating number of vehicles and their emission factors. The elemental composition, image interpretation, and size distribution of particles were analyzed comprehensively using energy

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dispersive X-ray (EDX) spectroscopy, scanning electron microscopy (SEM), and dynamic light scattering (DLS) methods, respectively.

Results: PM concentrations were found 2-4 times higher in commercial areas compared to industrial and residential areas, and are considerably correlated (p<0.05) with vehicle traffic and atmospheric temperature. Emissions of $PM_{2.5}$ and PM_{10} estimated numerically from road traffic data are significantly correlated (p<0.005 and p<0.0005, respectively) with PM concentrations measured experimentally. DLS and SEM image interpretation showed that about 90% of near-roadway particles were in the size of fine particles ($PM_{2.5}$) and 74% of them have circularity values above 0.75. EDX analysis found that roadside PM are carbon-rich particles containing 56% black carbon and trace amount of soil-derived particles, sea salts and metal-containing particles.

Conclusion: Experimental particle characterization by advanced laboratory analyses and numerical estimation of PM emission using model from road traffic survey both confirmed that fossil-fueled vehicles are the main source of PM emissions in urban area.

Keywords: Air pollution; particle characterization; particulate matter; urban roadways; vehicular emission.

1. INTRODUCTION

Air pollution in urban areas is one of serious concern in recent times, particularly for developing countries such as China and India, and it may further extend from local to global scale [1-3]. Awareness of health problems related to air pollution arising from industrialization and urbanization has increased, especially during the last two centuries. Numerous health effects attributable to particulate matter (PM) have been documented [4,5]. The health studies suggest that fine particles ($PM_{2.5}$) are more harmful than coarse particles (PM_{10}) [6]. Traffic is potential sources of both coarse and fine particles. Summary of reports explained that $PM_{2.5}/PM_{10}$ ratio in urban sites is nearly 0.5 and less than ratio reported in rural sites [7]. Several efforts have also been specifically aimed at studying concentrations and potential health effects of the so-called ultrafine particles [8,9].

The major emissions of total PM in the atmosphere are mainly from natural sources. However, it is rather difficult to differentiate between natural and man-made sources of emitted PM. The anthropogenic sources of PM in urban environments are mainly from vehicles, industries, fuel combustion, and biomass burning [10]. It has been clearly demonstrated that by using specific nanoparticle monitoring equipment the presence of nano- and ultrafine particles in outdoor urban environment are confirmed due to automobile traffic [11,12]. It is also well documented that vehicular PM emissions commonly consist of black carbon particles that are generated mostly by diesel- and gasoline-fueled vehicles [13,14]. Other trace elements of heavy metals are also commonly associated with non-exhaust vehicular emissions [15,16].

A number of comprehensive works has been taken to examine particulate matter such as particles of wear debris [17,18] and particles at construction jobsites [19]. Variation in particle size has been investigated by light [20] and electron microscopes [21]. The analytical methods used in the examination of atmospheric aerosols have been described comprehensively [22] including the transmission electron microscopy (TEM) and scanning electron microscopy (SEM) to investigate the microstructural characteristics of ambient particles, and the energy dispersive X-ray spectroscopy (EDX) to acknowledge the chemical composition of particulate matter. In addition to photomicrographic analysis, particle sizing can be examined by dynamic light scattering (DLS) method. Examples have been applied to nanoparticle study [23-25] and food system analysis [26,27].

Size of particles can be measured in terms of radius, diameter, area, and perimeter by combining the use of laboratory instruments and computational graphic applications. Studies in size, morphology, shape, and texture of different objects have been published [28]. The schematic diagram has also been proposed to classify the particulate matter according to the estimation of their element content [29]. But there is still a need to link between the physical and chemical characteristics of ambient particles and their possible sources of pollution, because unlike PM produced from point source emissions, the origin of particles dispersed in ambient air is not clearly identified. Therefore, the present investigation is

aimed to analyze the ambient particulate matter collected specifically at near-roadway urban environment at different habitation types. The analysis was conducted in full aspects of particle-related parameters including PM size distribution, elemental composition, and image shape interpretation as well as their concentration in relation with site traffic and weather condition variables.

2. MATERIALS AND METHODS

2.1 The Study Area

Mysore is one of the largest districts in the state of Karnataka, India. It lies at 12° 18' 25" N latitude and 76° 38' 58" E longitude. Altitude of the city is 765 m above mean sea level. According to the 2011 national census of India, the population of Mysore is 1,119,031 and it has the second largest population density after Bangalore metropolitan city. The city is well connected to the neighboring states through road transport and rail network. The district is bordered by four districts of the Karnataka state and one district of the Kerala state. The Mysore city has a warm, cool and salubrious climate throughout the year. The minimum temperature in winter is 15°C and the maximum temperature in summer is 35°C. Mysore gets most of its rain during the monsoon with an annual average of 782 mm [30]. Urbanization in Mysore city increases recently due to new establishment of industries, educational institutions, and transportation facilities. Mysore heritage centers also attract many tourists to come from other cities. Increase in public amenities leads to increase in traffic density and thus the vehicular emissions. Like many other Indian cities, Mysore city has high vehicular growth and emissions problem as well. It has over 523 thousand vehicles registered in the city in 2015 and is projected to expand about 120% in 2020 [31].

Sampling site of PM emission study was divided in three different categories namely commercial area, industrial area and residential area (Fig. 1). Two locations were selected in commercial area; one is located in Irwin Road (Site 1: 12° 18′ 51.36″ N, 76° 39′ 15.12″ E) and the other one is near Gandhi Square (Site 2: 12° 18′ 40.6″ N, 76° 39′ 14″ E). One spot in Metagalli industrial area (Site 3: 12° 21′ 51.6″ N, 76° 37′ 48.78″ E) was taken as the study site for the second category. Two sites was chosen in residential area; one is in Mysore University campus (Site 4: 12° 18′ 57.7″ N, 76° 37′ 16″ E) and the other one is in 3rd stage Vijayanagar, a newly developed village (Site 5: 12° 18′ 56.6″ N, 76° 36′ 28.6″ E). These three site categories have different vehicle traffic condition as well as pedestrian and population density. Irwin Road located in the city center has the highly narrow and congested traffic lane in Mysore city compared to other sites and was loaded by huge number of high storey buildings, but less green space (Fig. 2).

2.2 Sampling Protocols

Euroclean XForce vacuum air pump was used for collection of particulate matter. The sampler contains one inlet and outlet with two types of exchangeable filters (bag filter and screen filter). One end supplies the air to the vacuum pump with auto adjustable speed, and the pump pushes the air to the other end with an adjusted flow rate of 25 L/min. The inlet allows the admission of ambient air containing different size of particulate matter, whereas the outlet releases clean atmospheric air after passing through a step-wise filtering system. The sampler was set up exactly on the roadside to allow full collection of particulates originated from vehicular emissions. The sampling was carried out in the daytime when traffic density was relatively high, usually about five to six hours per day from morning to evening. PM entrapped in the vacuum pump filters were then taken out thoroughly and stored to the laboratory for further physical and chemical analyses.

Outdoor temperature, humidity, heat index, barometric pressure and wind speed were measured at all sampling sites. A weather center named 'AcuRite®' model 00615 was used to record those meteorological parameters. The instrument consists of a sensor and a digital display unit; both were connected using wireless transmission signal that can be monitored in a range up to 330 feet (or 100 meters). A sensor containing anemometer was placed on the top of the building to avoid any obstruction to wind flows when measuring wind speed. The instrument readings displayed in the LCD screen were noted down every half an hour. Daily average values for each weather parameter were calculated after recording the data in the daytime similar to time period of PM sampling.

Road traffic survey was conducted to analyze the site traffic condition at different study areas. In the survey, vehicles passing through the sampling points were counted at peak hours from

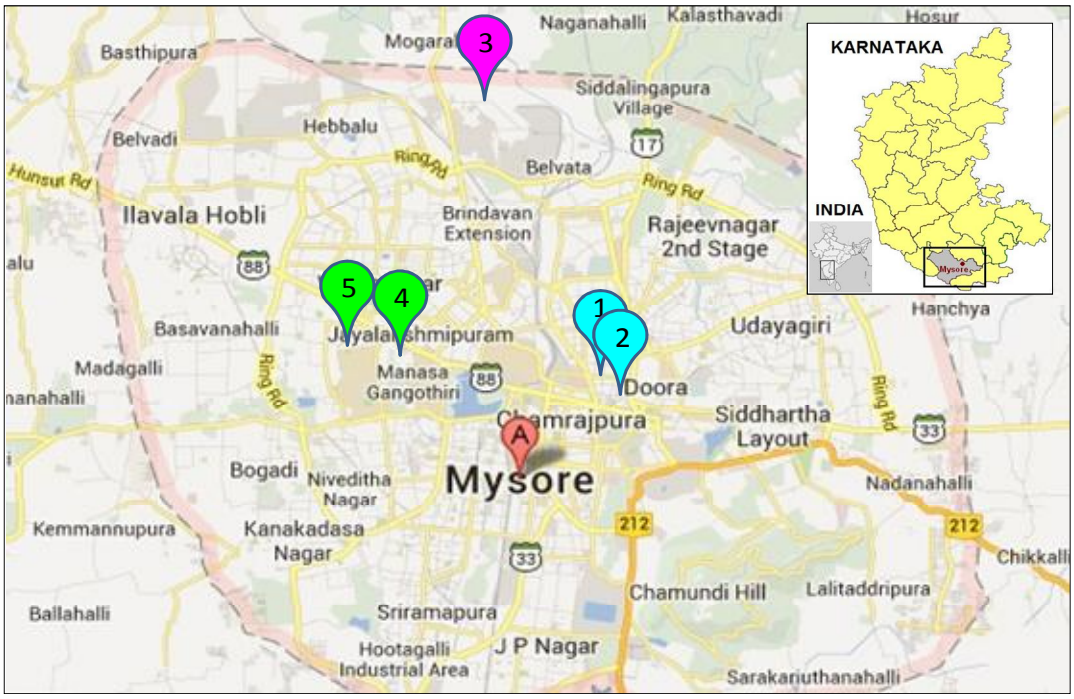


Fig. 1. Five study sites in three different categories: commercial (cyan), industrial (pink) and residential (green) area. Inset: Map of Mysore city in Karnataka, India

(a) **(b)** **(c)**

(d) **(e)**

Fig. 2. Panoramic views of (a) Site 1 (b) Site 2 (c) Site 3 (d) Site 4 and (e) Site 5

morning to evening to determine the average number of vehicles per hour that usually emit particulate pollution to the urban roadways. Vehicles were categorized into six groups namely two-wheelers (2W), three-wheelers (3W), four-wheelers (4W), buses, light commercial vehicles (LCV), and heavy commercial vehicles (HCV). Each category has different emission factor. The factors depend on the types of vehicles, engine cylinder volume, vintage, and fuel technology used [32,33]. Emission factors calculated in the previous work [34] were used in this study. Vehicular $PM_{2.5}$ and PM_{10} emissions [34-36] were estimated by the following equation:

$$E_i = \sum_{j=1}^{6} \left(N_j \times f_j \times d_i \right) \quad (1)$$

where, subscript *i* represent the study site and *j* is the respective vehicle category;

E_i = emission of particulate matter, either $PM_{2.5}$ or PM_{10}, in study site *i* ($g \cdot h^{-1}$);
N_j = number of vehicles in category *j* (h^{-1});
f_j = emission factor of vehicle category *j* ($g \cdot km^{-1}$);
d_i = distance of road segment in study site *i* (km).

$PM_{2.5}$ emission factors for two wheelers, three wheelers, cars, buses, LCVs, and HCVs are 0.038; 0.075; 0.06; 0.773; 0.332; and 1.095 g/km, respectively, whereas PM_{10} emission factors are 0.083; 0.188; 0.175; 0.566; 0.239; and 0.477 g/km, respectively. Distances or lengths of road segment in Site 1, 2, 3, 4 and 5 were measured to be in the order of 1.51; 0.48; 0.5; 0.19; and 1.63 km, respectively.

2.3 Laboratory Analyses

The physical analysis was performed by Shimadzu AX200 analytical balance with 0.1 µg accuracy. The balance was used in order to obtain the gravimetric analysis of the sampled filter mass. The mass concentration of suspended particulate matter (SPM) collected by the sampler [19] is given by:

$$C = \frac{m}{Q \times T} \quad (2)$$

where,

C = particulate concentration in ambient air (mass/volume);
m = net mass of particles collected in the sampler filter (mass);
Q = flow rate of ambient air pumped to the sampler (volume/time);
T = total duration of particle collection (time).

Particulate matter measured by the sampler is considered as total suspended particles, because no specific attempt was taken to separate the collected particles in different size categories. Relations between SPM concentrations and other factors like meteorological parameters, traffic flows and vehicular PM emissions were analyzed statistically using SPSS version 16.0 and Data Analysis Tool from Microsoft Excel.

Particle size distribution was determined by dynamic light scattering (DLS) method. The technique measures the random changes in the intensity of light scattered from a liquid suspension. Particle samples were dispersed in a specific liquid, vortexed to provide a homogeneous solution, and transferred to the DLS instrument in a square cuvette. The instrument model employed in this work is Nanotrac Wave™. DLS instrument works by measuring the dynamic thermal motion of the particles in the liquid suspension, known as the Brownian motion. Applying an AC electric field creates an electrophoretic mobility. Combining this with Brownian motion enables the system to measure the velocity distribution of charged particle movement [30]. The technique uses a laser that penetrates the suspension and illuminates the sample in the cell. Scattering takes place with the encountered particles. The backscattered light mixed with the incident light returns to the photodetector. Light scattering intensity is sampled at regular intervals as a raw photodetector signal and converted into a frequency power spectrum. Recording this data will result in accurate particle size distribution calculated either in nm or µm.

A scanning electron microscope was used to capture an image of particle samples by scanning them with a focused electron beam over their surface. The electrons in the beam interact with the sample and produce various signals that can be used to obtain information about surface topography, morphology, composition as well as number and volume-size distribution of particles [22]. SEM can produce very high-resolution images of a sample surface to thousand times the magnification limit of the best light microscopes. In this work, the high definition Zeiss EVO LS15 SEM was employed. It has magnification range from <5 to 1,000,000× and is capable to produce images at resolution as small as 2 nm. The images captured by SEM were stored into digital format for detail analysis. The digital images were then processed in a computer to adjust their contrast and brightness, and to quantify their size (length or diameter), area, perimeter, and circularity. The circularity, also known as degree of roundness, is dimensionless. It represents the shape or form factor of particles [28,37] and is defined as:

$$Circularity = 4\pi \frac{Area}{Perimeter^2} \quad (3)$$

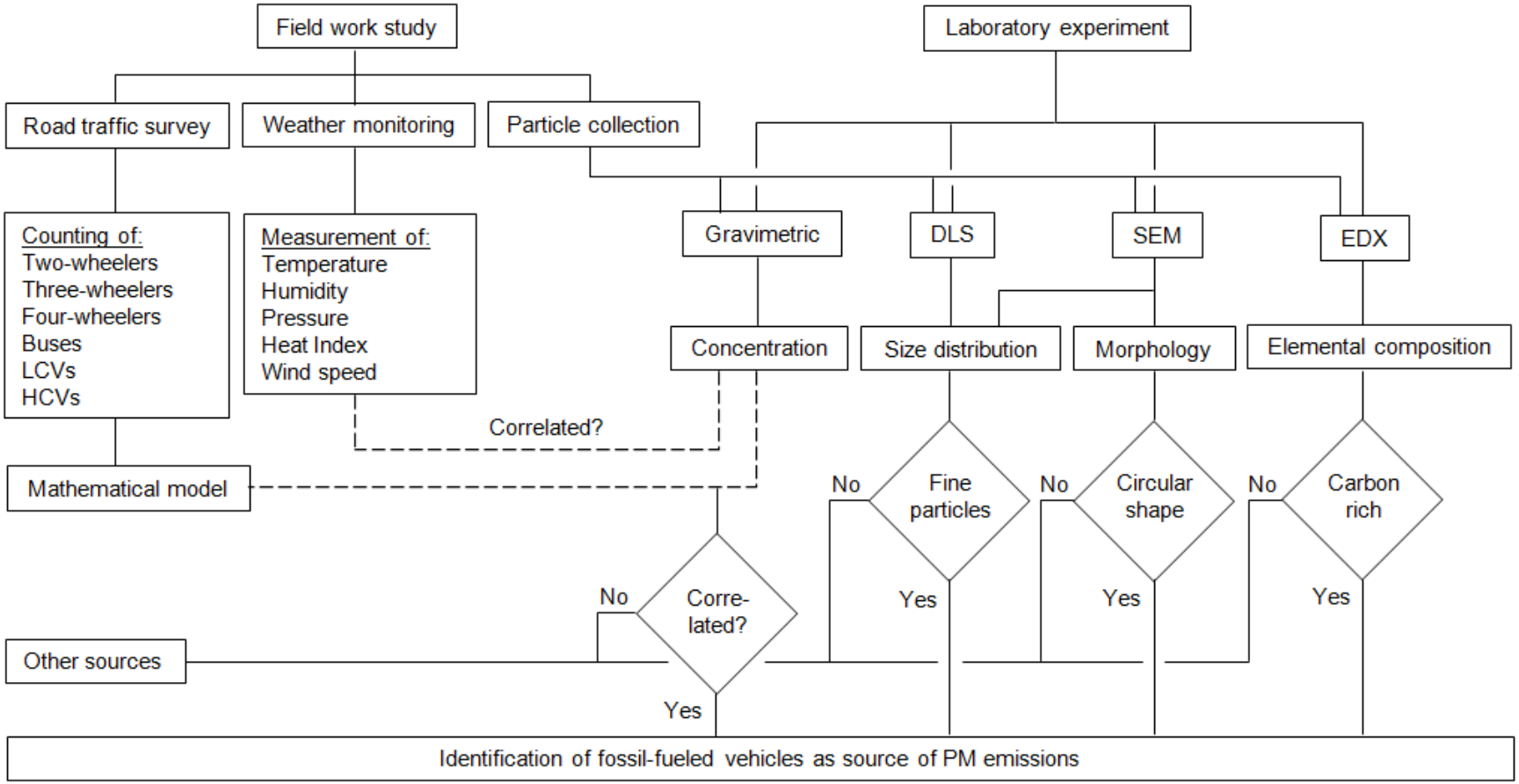


Fig. 3. Diagram of research study in identifying the source of PM emissions in urban ambient environment

By using this formula, a perfectly rounded particle sphere should have circularity value of 1. Liquid droplets or aerosols are almost nearly spherical. Sielicki et al. [22] suggested that particles of unburned fuel are considerably circular. Irregularly shaped particles will have less circularity values. Scientists usually use the term 'equivalent or aerodynamic diameter' to characterize the shape of irregular particles. In this study, length, area, and perimeter of particles were precisely measured by employing specific image processing software (Digimizer version 4.6.0) before being exported to Microsoft Excel spreadsheet to calculate their size distribution and circularity values.

Elemental composition of PM was investigated by Energy Dispersive X-Ray (EDX), also referred to as Energy Dispersive Spectroscopy (EDS) analysis. The instrument model used is NORAN System 7 X-ray Microanalysis. The technique detects X-rays emitted from the sample during bombardment by an electron beam to balance the energy. The data generated by EDX instrument consists of spectra showing peaks corresponding to the specific elements. The count number of emitted X-ray in Y-axis versus their energy in X-axis is evaluated to determine the true elemental composition of the sampled volume being analyzed. The X-ray energy was converted to voltage pulse and recorded in voltage units (usually in keV). The height of the peaks represents the relative abundance of X-rays emitted by the elements. The higher the peak, the more X-rays emitted by the element, thus the abundance of that element presents in the sample will also be higher. The data were then converted into composition units in weight percentages after calculating the atomic weight of each element. All methods used in this study are described clearly in a schematic diagram presented in Fig. 3 explaining the full concept of the research.

3. RESULTS AND DISCUSSION

3.1 PM Concentrations and Related Factors

Gravimetric analysis of the sampled particles found that ambient air in Site 1 located in commercial areas has the highest PM concentration than any other areas (Table 1). The concentration was about three times of Site 2 located in the same area type. This is due to the sampling location of Site 2 is around the road circle and has more spacey area than of Site 1. Roadway in Site 2 is about ten meters in width, whereas in Site 1 is only six meters. Wider space gives more medium for the dispersion of particles to the atmosphere, hence reduces the concentration. Ambient air in industrial and residential areas was found to have quite similar concentrations of particulate matter, even though the composition might be different. The meteorological data of all sites are also presented in the same table.

It is noted that using statistical data analysis, almost no significant correlation was found between any meteorological parameter and PM concentration in this current study, except for outdoor temperature (Fig. 4a). The ambient PM concentrations may be influenced by outdoor temperature as they considerably correlated (r = 0.9, $p < 0.05$; 4 d.f). The relation may be due to less vegetation exist in commercial area resulting in less shaded area and higher outdoor temperature. Less vegetation cover also minimizes the occurences of ambient particulate trapping by leaves stomata. Moreover, commercial area usually has higher traffic volume. Ambient temperature in high traffic area is relatively higher due to heat generated by the vehicles' engine smokes.

This correlation may be different to other studies with different location types and different source of particles. Study at construction jobsites found that wind speed has significantly correlated to PM concentration compared to other weather variables [19]. The reason may be due to the site locations are relatively large and widely open allowing wind to disperse the suspended particles easily to the ambient air. The condition is somehow different with urban roadways that are more congested with vehicles and surrounded by rows of high storey buildings in the left and right side of the road, particularly in the case of Site 1 (Fig. 2). Wind speed may not affect PM concentration in such location due to lots of obstruction in particle dispersion.

Number of vehicles presented in urban roadways was found more in commercial areas (Table 2). The number was about ten-fold compared to such in industrial or residential areas. Among all vehicle categories, two-wheelers (2W) contributed more to the traffic crowd. In fact, the two-wheelers dominated all road spaces in any type of habitation, either in commercial, industrial, or residential area. Passenger auto rickshaws (3W), buses, and passenger cars (4W) – including also jeeps, vans, and taxi – were abundant in commercial areas. Light commercial

vehicles (LCV) such as luggage auto rickshaws and tempos were widely found in commercial and industrial areas. Only heavy-duty trucks owning six tires or more (HCV) were found more in industrial area.

Although two-wheelers have the least emission factor (f_j) compared to other vehicle types, but because of their significant numbers in urban roadways, they contributed more to the particulate emissions in almost all sampling sites, either to $PM_{2.5}$ or PM_{10} emissions. Due to high traffic density, Site 1 received the largest particle emissions from urban vehicles followed by Site 2 located at the same area type. On the contrary, vehicles were found less in residential areas; therefore the areas receive lesser PM emissions not more than 10 g/h. $PM_{2.5}/PM_{10}$ ratios were almost nearly half in all sampling sites, except in industrial area that measured about 0.89. It is because particles calculated in PM_{10} category also include particles in $PM_{2.5}$ size and most of the study sites are located in urban areas. These ratios are quite relevant to the studies reported by the other scientists elsewhere [7].

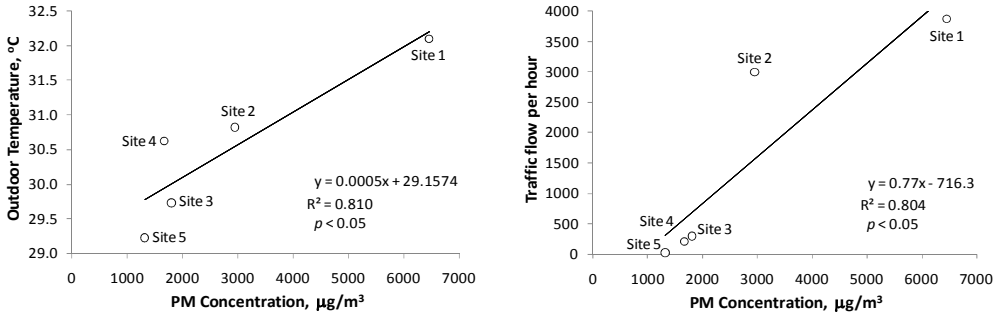
Considerable correlation was found between PM concentrations and site traffic density (r = 0.897, p < 0.05; 4 d.f). Commercial areas located in the city centre with high traffic flow per hour have higher PM concentrations (Fig. 4b). Conversely, Site 4 and 5 located in residential area was found to have less concentration of ambient particulates. Industrial area with relatively low traffic volume as compared to commercial area still have adequate particle concentration, because of high number of diesel-fueled vehicles (HCV) that contributed more particulate emission due to their high emission factors. Industrial area also received non-vehicular particles emitted by industrial smoke stacks from neighboring metal and rubber manufacturing industries located not too far from the sampling site.

Linear regression analysis found that ambient PM concentrations measured experimentally by air sampler are significantly correlated with PM emissions emitted by motor vehicles, either $PM_{2.5}$ or PM_{10} (Fig. 5). High amount of $PM_{2.5}$ emissions will increase ambient PM concentrations (r = 0.99, 99% CI from 6.7 to 18.8 g/h, p < 0.005; 4 d.f). Similarly, increase in PM_{10} emissions will also affect to the high amount of ambient PM concentrations (r = 0.996, 99% CI from 4.6 to 8.8 g/h, p < 0.0005; 4 d.f). It clearly explains that ambient particulates presented in urban roadways are mostly generated from vehicles' tail end pipe emissions. Complete descriptions in correlation values between particulate pollution related factors are presented in Table 3. The relatively similar profiles between PM concentrations, traffic flows, vehicular $PM_{2.5}$ emissions, and vehicular PM_{10} emissions in all sampling sites are described in Fig. 6.

Table 1. PM concentration and weather condition* records in five sampling points

Site	Concentration µg/m^3	Temperature °C	Humidity %	Pressure kPa	Heat index °C	Wind speed kph
1	6464	32.09 ± 0.58	23.82 ± 1.11	1.0110 ± 0.0003	30.55 ± 0.45	3.73 ± 0.52
2	2954	30.82 ± 0.35	18.73 ± 0.97	1.0107 ± 0.0003	29.18 ± 0.30	5.91 ± 0.44
3	1808	29.73 ± 0.51	22.09 ± 0.94	1.0103 ± 0.0004	28.36 ± 0.36	10.27 ± 0.80
4	1674	30.62 ± 0.40	42.00 ± 0.95	1.0117 ± 0.0002	30.92 ± 0.35	5.08 ± 0.46
5	1322	29.22 ± 1.10	31.11 ± 4.24	1.0114 ± 0.0005	29.57 ± 0.37	3.56 ± 0.84

** All weather values are mean ± S.E.M.*

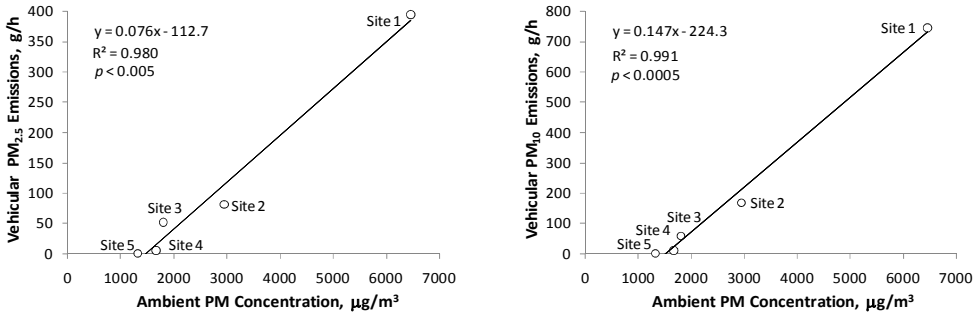


(a) **(b)**

Fig. 4. Relations between PM concentrations and (a) temperature (b) traffic flow

Table 2. Traffic condition and hourly emissions of $PM_{2.5}$ and PM_{10}

Study site *i*	1	2	3	4	5
Traffic flow per hour					
2W	2475	2072	224	188	12
3W	794	601	4	1	1
4W	464	244	25	12	3
Bus	73	4	1	0	0
LCV	59	67	25	1	1
HCV	3	4	11	1	0
Total *Nj*	**3868**	**2992**	**290**	**203**	**17**
$PM_{2.5}$ emissions (g/h)					
2W	142.02	37.79	13.87	3.57	0.09
3W	89.92	21.64	0.49	0.04	0.01
4W	42.04	7.03	2.45	0.36	0.03
Bus	85.21	1.48	1.26	0.00	0.00
LCV	29.58	10.68	13.53	0.17	0.06
HCV	4.96	2.10	19.63	0.55	0.00
Total E_i	**393.72**	**80.72**	**51.23**	**4.68**	**0.20**
PM_{10} emissions (g/h)					
2W	310.19	82.55	30.30	7.80	0.19
3W	225.40	54.23	1.23	0.09	0.04
4W	122.61	20.50	7.13	1.05	0.10
Bus	62.39	1.09	0.92	0.00	0.00
LCV	21.29	7.69	9.74	0.12	0.05
HCV	2.16	0.92	8.55	0.24	0.00
Total E_i	**744.05**	**166.97**	**57.88**	**9.30**	**0.37**
$PM_{2.5}/PM_{10}$	**0.53**	**0.48**	**0.89**	**0.50**	**0.54**



(a) **(b)**

Fig. 5. Relations between ambient PM concentrations and vehicular emissions of (a) $PM_{2.5}$ (b) PM_{10}

Table 3. Correlation coefficients (*r*) between PM concentrations, PM emissions, traffic flows and weather parameters

	C	TF	E2.5	E10	T	H	P	HI	W
C	1								
TF	0.897*	1							
E2.5	0.990**	0.837	1						
E10	0.996***	0.858	0.998***	1					
T	0.900*	0.864	0.849	0.865	1				
H	-0.398	-0.586	-0.393	-0.381	-0.181	1			
P	-0.160	-0.263	-0.190	-0.152	-0.005	0.858	1		
HI	0.360	0.193	0.323	0.354	0.551	0.677	0.813	1	
W	-0.341	-0.298	-0.297	-0.345	-0.326	-0.357	-0.773	-0.741	1

C = PM concentration, TF = traffic flow, E2.5 = $PM_{2.5}$ emission, E10 = PM_{10} emission, T = outdoor temperature, H = humidity, P = atmospheric pressure, HI = heat index, W = wind speed.
** $p < 0.05$, ** $p < 0.005$, *** $p < 0.0005$.*

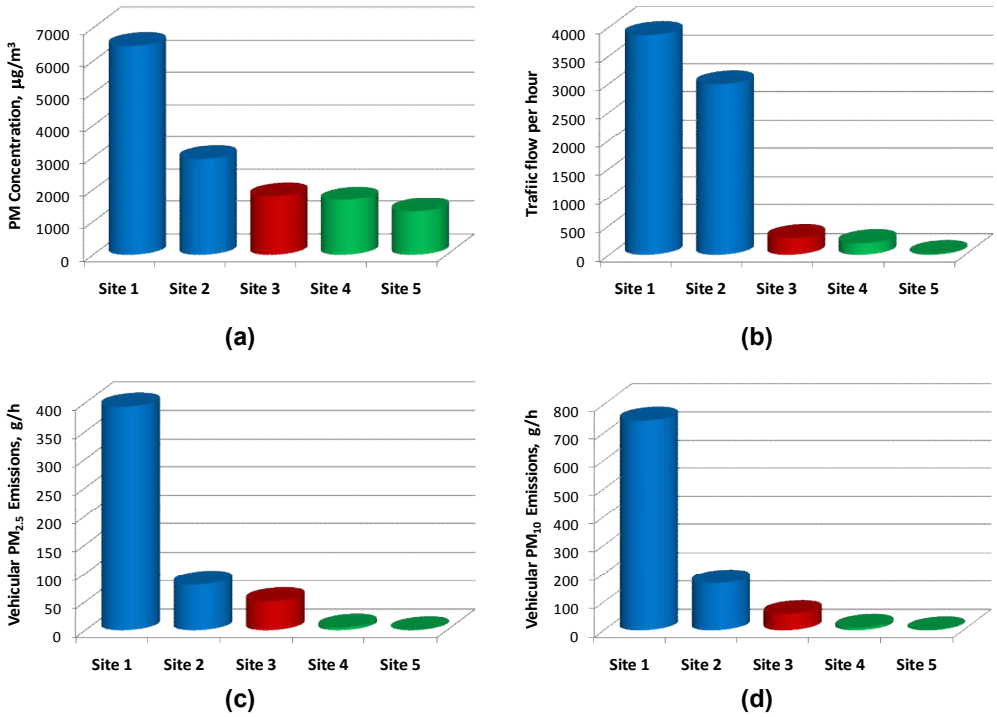


Fig. 6. Variations in (a) PM concentrations (b) Traffic flows (c) $PM_{2.5}$ emissions (d) PM_{10} emissions in each study site

3.2 Particle Size Distribution

The dynamic light scattering method was used to identify the size variation of particles by suspending the samples in a specific liquid contained in the cell. Viscosity of the solution after mixing with the samples was 0.819 cp recorded at the temperature of 28.76°C. Conductivity of the solution was 888 µS/cm, while the pH was relatively neutral (7). The zeta potential of the particles that is proportional to the modulated power spectrum signal was -8.6 mv with negative polarity. The output result obtained from DLS instrument is presented in the form of particle size distribution graph calculated in nanometer (Fig. 7).

The results showed that ambient particulates entrapped in the sampling chamber were mainly fine particles ($PM_{2.5}$) with size less than 2.5 µm or 2500 nm (Table 4). The larger the size, the less percentage of particles was identified. The size distribution presented in the table is certainly higher for particles in ultrafine category or $PM_{0.1}$ (size of <100 nm in diameter) and much lower for PM_{10} category (size of 2.5 to 10 µm in diameter). Analysis from DLS instrument found that only 0.12% of sampled particles have aerodynamic diameter above 2,500 nm or in the size of PM_{10} (coarse particles). It means that the remaining 99.88% have diameter less than 2.5 µm or in the category of fine particles ($PM_{2.5}$). It is relevant to other studies carried out in other worldwide cities that ambient air in urban traffic areas contain mostly fine and ultrafine particles represent fossil-fueled vehicles as the main source of emissions [11,12].

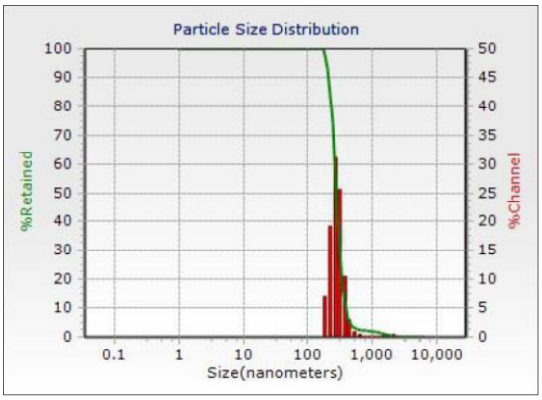


Fig. 7. Particle size distribution obtained from DLS analysis

The DLS machine has recorded (i) maximum number of particles suspended in the sample

container was in the size of 318 nm, (ii) maximum area of scattered particles has a mean diameter of 1,010 nm, and (iii) maximum volume of suspended particles has an average diameter of 1,728 nm. Moreover, the peak summary report obtained from the instrument also indicated that 97.4% of the total sample volume contained particles with a mean aerodynamic diameter of 275.8 nm. The remaining 2.6% were relatively larger particles with an average diameter of 1,505 nm or still below 2.5 microns.

Table 4. Size of particles measured in percentiles

Percentile	Size above (nm)
10	377.9
20	335.7
30	311.9
40	293.6
50	277.7
60	263.1
70	248.6
80	232.5
90	211.8
95	197.8

All results in DLS analysis explain that particles sampled in Mysore urban city roadways belong to the $PM_{2.5}$ category. Ratio between $PM_{2.5}$ and PM_{10} measured in the DLS analysis is relatively higher and different with ratios reported by Table 2, because in DLS analysis ratio was measured based on particle number, not on weight as calculated by numerical model of PM emission estimation.

3.3 SEM Image Interpretation

SEM analysis produces black and white or pseudo color images that can be saved in different image formats and at various pixel resolutions. Image resolutions provided by Zeiss EVO LS15 SEM can be manually selected up to 3072×2304 pixels. In this study, particle samples were saved in TIFF-image format at resolution of 1024×768 pixels. The magnification of the images varied from 1,000× to 15,000× (Fig. 8). The particle objects (n = 100-200) displayed in the SEM photomicrographs was digitized in the computer by using Digimizer software to measure particle length (or equivalent diameter) required for size distribution analysis and to identify area and perimeter of particles required for circularity value measurement. Bar scales provided on the bottom-left corner of the SEM images ranging from 1 to 10 µm were used as references for computational length measurement in the image processing software.

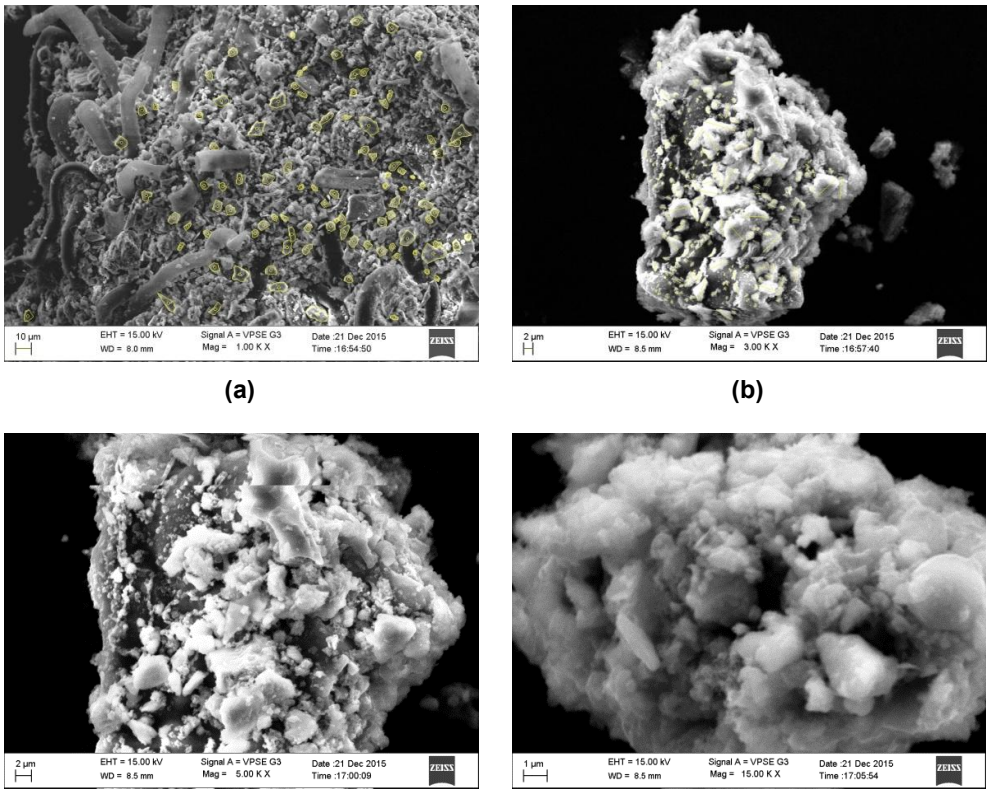
Computational graphic analysis of SEM images has resulted in particle size distribution (Fig. 9) and circularity value of the particles (Fig. 10). It was found that under scanning electron microscope, the mean size of the particles collected at the sampling site was 1.66 µm. In details, they were distributed in a variety of size, mainly <1 µm (31%), <1.5 µm (26%), <2 µm (11%) and <2.5 µm (10%). It again indicates that majority (83%) of roadway PM in Mysore city were in the size of $PM_{2.5}$ category or fine particles, even though a few of them (17%) has aerodynamic diameter above 2.5 microns, but due to resolution constraint, smaller particle objects size below 0.5 microns – including ultrafine particles or $PM_{0.1}$ – should be more than they have identified in the analysis. Higher magnification and pixel resolution will enable the identification of tiny particles that, in turn, will increase the percentage of $PM_{2.5}$ category. The domination of fine and ultrafine size in particle samples indicates that automobiles are the main source of airborne PM emissions [11,12].

Particles generated from unburned fuel of vehicles tend to have circular shape [22]. The shape of the sampled particles in this study was found close to circular; even they were not exactly spherical. Therefore, it is suggested that the analyzed samples were traffic-related particles. Most of the particles have circularity values up to 0.85 (31%), 0.8 (27%) and 0.9 (13%). However, the percentage of more circular objects could be higher if using higher resolution or better contrast images and considering not only large size particles in the shape evaluation [28]. High difficulty was found in the computational analysis when digitizing the smaller objects or random stack of particles. Better analysis would be achieved if the particles were dispersed in a very thin and spacey area while taking photomicrographs by the electron microscope. Moreover, presence of non-particulate objects in the photomicrographical images – like cotton fibers brought originally from the sampler's filter bag, somehow, obstructed the computational image analysis [22].

3.4 Elemental Composition

The EDX instrument readings described spectrum charts showing a number of peaks that

correspond to the chemical elements present in the particulates (Fig. 11). The result showed that ambient particulate matter collected at Mysore city center either on summer season (from mid February to mid May) or monsoon season (from mid May to mid August) comprised elements mostly in the form of carbon (C, above 50%) and oxygen (O, above 30%). Other elements were found in smaller fraction such as silicon (Si), iron (Fe), aluminium (Al), calcium (Ca), sodium (Na), potassium (K), and magnesium (Mg); each below 4% in weight (Table 5, Fig. 12).



(a) **(b)**

(c) **(d)**

Fig. 8. SEM digital images of PM by magnification of (a) 1,000× (b) 3,000× (c) 5,000× and (d) 15,000×. Yellow lines and polygons show objects in the digitation process

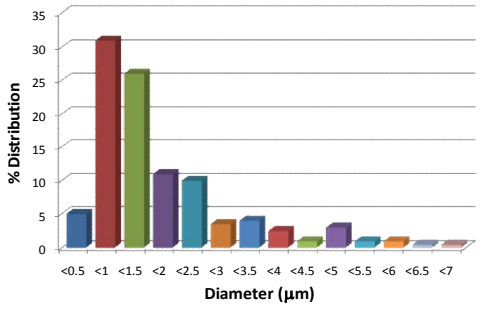


Fig. 9. Size distribution of roadway PM analyzed from SEM digital images

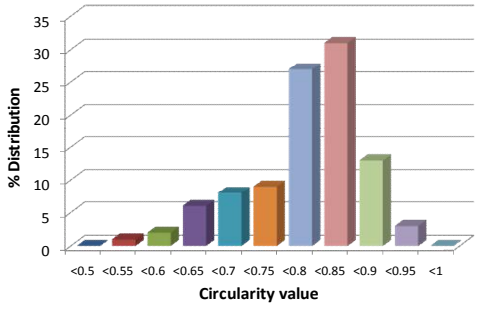
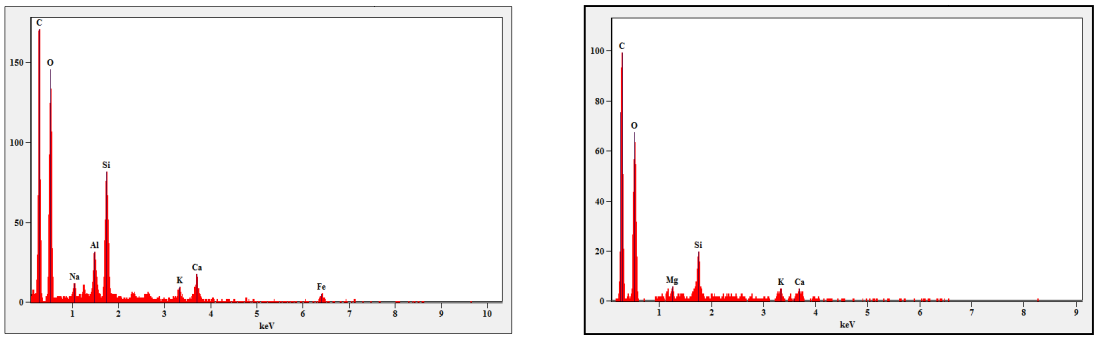


Fig. 10. Classification data for the circularity values of particulate matter



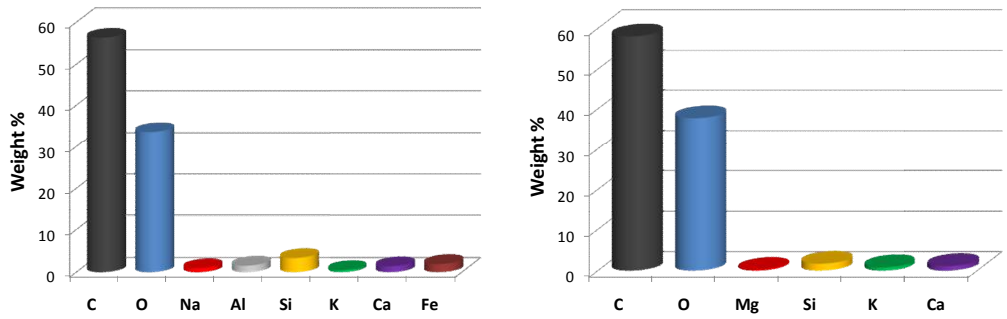
(a) (b)

Fig. 11. EDX spectra showing peaks corresponding to the elements present in particulate matter collected on (a) summer season (b) monsoon season

Table 5. Percentage of elements detected by EDX instrument in the sample of particulate matter collected at Mysore urban area

Collection time	Percentage of elements								
	C	O	Si	Fe	Al	K	Ca	Na	Mg
Summer	56.38	33.66	3.39	1.97	1.59	0.56	1.46	0.99	nd
Monsoon	58.13	37.78	1.76	nd	nd	0.90	1.18	nd	0.25

nd = not detected



(a) (b)

Fig. 12. Weight percentage of elements present in particulate matter collected on (a) summer season (b) monsoon season

In nature, particles may present in the form of single elements or chemical compounds. The only disadvantage of EDX technique is the unavailability data about which compound that may compose the particulate matter [22]. The result obtained from the instrument only provides elementary basis. In the sampled particles, single elements may be found in the form of elemental carbon, iron, and aluminium, whereas compounds are combination of different elements that can be assigned as C-rich, Fe-rich, Si-rich, Fe/Si-rich, Al-rich, Ca-rich, S-rich, K-rich, or other metal-rich particles [21,29]. Oxygen element is abundant in the analysis because most of the compounds usually require one or more oxygen atoms as electron receptors needed to form anions in their chemical structures, except for compounds with halogen anions.

Chemical compounds that may constitute in the particulate matter can be estimated based on their metal or metalloid cations. There are many possibilities of chemical structures, but the simple estimation is often made according to the formulae that are widely known and most

available in nature. For instances, iron may be assigned as iron rust or iron oxides (FeO or Fe_2O_3); aluminium as alumina (Al_2O_3); magnesium as magnesium carbonates ($MgCO_3$); calcium as lime (CaO, $CaCO_3$, $Ca(OH)_2$) or gypsum ($CaSO_4$); sodium and potassium both as chloride compounds or sea salts (NaCl and KCl, respectively); or other possible forms of compound like oxides, hydroxides, chlorides, carbonates, sulfates, phosphates, etc. Silicon can be assigned as pure silica (SiO_2) or as complex compounds such as aluminosilicates (Al_2SiO_5), $CaAl_2Si_2O_8$, or $(Na,K)AlSi_3O_8$ [29].

Based on EDX particle characterization and particle morphology, Ebert et al. [38] differentiated particulate aerosols collected from urban and rural environments into various forms such as silica, aluminosilicates, marine salts, ammonium sulfates, calcium sulfates, metal oxides or hydroxides, soot, carbon-rich particles, carbon/sulfate-mixed particles, and biological particles. In other study Geng et al. [39] has divided atmospheric particles into several possible compounds including primary soil-derived particles (SiO_2-containing particles, AlSi-containing particles, $CaCO_3$, and $CaSO_4$), secondary soil-derived particles, marine particles (raw or reacted sea salts, or mix of both), organic particles, carbon-rich particles, K-containing particles, Fe-containing particles, fly ash, and droplets rich in C, N, O, and S.

Referring to the above categorizations, near-roadway particles collected either on summer or monsoon season mostly consist of carbon-rich particles and a considerable amount of silica. Summer collection also obtained aluminosilicates. Both silica and aluminosilicates were identified as soil-derived particles. Other soil-derived particles may also be assigned as gypsum, lime, and magnesium carbonates. Small amount of particle constituents may be derived from marine aerosols (or sea salts) and metal oxides or metal- (potassium and iron) containing particles. Ambient particles rich in elemental black carbon indicated that sources of airborne particulate matter entrapped in the sampler's filters were mainly from motor vehicles that often emit carbonaceous agglomerates as waste products of diesel and gasoline fuel combustion [13,14].

4. CONCLUSION

Although the emissions of PM air pollution are largely from natural sources, like forest fires and volcano eruptions that incidentally occur, but the recent evidences in medical and toxicological research found that daily respiratory health problems related to air pollution in world public also take place in urban areas where industrialization and motorized transportation develop relatively higher. Despite these concerns, large proportion of urban people in Asian countries is still unaware to the situation. As part of the initial action, the examination of PM dispersed in the ambient environment is somehow needed to find the exact source of PM emissions in urban area, thus the framework in mitigating the further effects of airborne PM pollution will have the focus target.

There is no standard methodology adapted for dust analysis. This work brings a set of methodology to identify the source of PM emission in ambient air, taking into consideration different factors like spatial, seasonal, and meteorological condition. The methodology combines both experimental and numerical data taken from field work study, laboratory experiment, and computational analysis that are useful for quantifying and characterizing the airborne PM. It employed energy dispersive X-ray spectroscopy, scanning electron microscopy, and dynamic light scattering techniques to derive the size-resolved particle distribution, morphology, and composition of atmospheric aerosols.

DLS has measured the sampled particles to be in the category of fine particles ($PM_{2.5}$). Interpretation of SEM images also provided the same result similar to DLS with addition of findings on nearly circular shape of particles indicating the automobile traffic as the main source of PM emissions. Analysis of elemental composition using EDX also generated important findings that majority of urban roadway particles were carbon-rich particles – widely known as black carbon – that emitted mostly from fuel combustion of vehicles. All findings come to the same point that emissions of motor vehicles are the main sources of particulate pollution in urban environment. These findings are certainly important for policymakers to give more concerns in mitigating the health effects of vehicular PM pollution rather than to focus only on PM emitted from natural sources.

Apart from those valuable results, however, there is still a need for further studies in the effects and examination of non-vehicular particulate matter such as from domestic and industrial practices.

The present study may be useful as a basic framework to replicate the same methods in different types of particles or different emission sources using different models for correlating the findings between laboratory analysis and numerical study.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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