Chapter 5

The Air Quality Influences of Vehicular Traffic Emissions

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Additional information is available at the end of the chapter

http://dx.doi.org/ 10.5772/64692

Abstract

The number of automobiles has been steadily increasing in cities as a consequence of rapid urbanization and economic growth. It has been widely reported that vehicular emissions are strongly correlated with the level of urban air pollution. The major primary air pollutants that are linked to direct emissions from on-road vehicles include soot (black carbon), carbon monoxide (CO), and nitric oxide (NO). Human exposure to these air pollutants is of health concern. Therefore, it is important to investigate air pollutants of traffic origin (e.g., BC, CO, and NO) in ambient air at different locations of cities and to assess the effects of vehicles on the urban air quality. With this goal in mind, we carried a systematic study in Singapore (the fourth most densely populated country in the world) with concurrent measurements of BC, NO, and CO in ambient air at four different locations having variations in traffic flows and meteorology. We then assessed the relationship between traffic flows and prevailing levels of the three air pollutants, and studied the association of these air pollutants among each other and with diverse meteorological conditions. The major outcomes of the study are discussed.

Keywords: urban air quality, traffic emissions, aerosol black carbon, carbon monoxide, nitric oxide, diurnal variations

1. Introduction

Aerosol black carbon (BC) is a widespread environmental pollutant, which is generated from combustion processes of carbonaceous materials at high temperature. Being the principal light absorbing aerosol species with specific absorption coefficients ranging from 11 to 12 $m^2 g^{-1}$ at 650 nm, BC absorbs radiation that lowers the single scattering albedo [1, 2]. As a result, the amount of reflected radiation is reduced, and the radiation absorbed by the atmosphere is increased. Due to the nature of higher porosity, BC adsorbs other species from the vapor phase,

especially organics that are potentially mutagens or carcinogens [3, 4]. For example, BC adsorbs polycyclic aromatic hydrocarbons (PAHs) with four rings or more, those are carcinogenic in nature [5, 6]. In the environment, BC is inhaled frequently by humans that can be deposited in the lungs or other airways causing severe health effects on a long- or short-term basis [7, 8]. In addition, BC provides surfaces that may catalytically promote certain other reactions in the atmosphere [9]. This can be seen in the environment with higher levels of BC that affect ozone (O_3) and nitrogen oxide (NO_x) concentrations due to heterogeneous destruction of O_3 molecules on particles [10]. For example, Dasch and Cadle [11] observed that oxidation of sulfur dioxide (SO_2) is also catalyzed by BC aerosol.

During the past two decades, research on BC has been increasing rapidly due to its role on local air quality causing visibility problems and adverse health effects, regional air quality affecting cloud microphysics, and global climate change due to its positive radiative effects [12–18]. Previous studies have also reported that BC of local origin can be transported over a long distance due to its long lifetime (order of several days to several weeks depending on meteorology) in the atmosphere [19]. Major anthropogenic sources of BC include on-road vehicles, domestic heating, industrial activities, and refuse burning, among which vehicular emissions are particularly dominant [20–22].

Human exposure to ambient BC, carbon monoxide (CO), and nitric oxide (NO) is of concern in urban environments due to a high population of vehicles [23, 24]. Such higher exposure to BC, CO, and NO causes several health effects, including myocardial infarction and pneumonia, elevated inflammatory markers of cardiovascular disease, diminished heart rate variability, and ventricular tachyarrhythmias. [23–27].

In general, two types of studies are performed to characterize the vehicular emissions, i.e., direct tests on engines and experiments in the ambient air. In the past, several studies focused on engine particle emission through engine test sites or chassis dynamotors [28–30]. However, to get the real atmospheric particulate characterization, it is essential to perform field measurements. Although some studies [31, 32] were performed under stationary measurements in recent years, these studies were, however, confined to a particular measurement site. Therefore, ambient characterization with on-road measurements under real moving traffic conditions are the best experiments that can provide better scientific knowledge on traffic emissions.

In the past, several detailed studies were undertaken in Singapore to assess the status of air quality and particulate matter characterization [33, 34]. However, these studies did not address the effects of vehicles on aerosol BC and association of NO and CO with BC. The latest study by Kalaiarasan et al. [35] investigated traffic-generated airborne particles in naturally ventilated multistorey residential buildings of Singapore and the potential health risk at different vertical heights. To get a clear picture on the role of vehicles on ambient air quality, it is essential to get the status of air quality at different urban locations with varying traffic loads. The status of ambient air quality at human breathing levels (2 m above ground) can be augmented to health risk models to assess the risk due to location of a particular area.

http://dx.doi.org/ 10.5772/64692

Some limited studies have been carried out in the urban areas of developed countries recently to assess the effects of vehicles on ambient air quality [36–40]. These studies monitored the air quality status (BC, CO, and NO) near locations of heavy and low traffic flows for comparative assessment of air quality. However, no such studies were undertaken in tropical areas within Southeast Asia. We undertook a preliminary study in Singapore for the first time. Being a heavily motorized country, the traffic system in Singapore (the fourth most densely populated country in the world) is of particular interest in the region of Asia Pacific from air pollution point of view [35, 41, 42]. In connection to health problems, bronchial asthma is one of the common respiratory disorders in Singapore and is commonly observed that about 1 of 5 children is asthmatics [35]. Therefore, there is a strong need to investigate traffic-generated pollutants (BC, CO, and NO) in ambient air at different locations and to assess the effects of vehicles on the ambient air quality.

The present study was designed with the following objectives: (1) measurements of levels of BC, NO, and CO in ambient air at four different locations having variations in traffic flows and meteorology, (2) assessment of the relationship between traffic flows and prevailing levels of the three air pollutants, and (3) examination of the association of these air pollutants among each other and with the meteorology.

2. Materials and methods

2.1. Characteristics of study area

The study was conducted in Singapore, which is located at the tip of the Malayan Peninsula (1°09'N to 1°29'N and 103°36'E to 104°25'E) with areas of about 699 km^2 and population of about 4.7 million [35]. Singapore's geographical location and maritime exposure, uniform temperature, pressure, and high humidity characterize its climate. The temperature ranges from 32°C for a high and 24°C for a low with a daily mean humidity of 84.4%. Singapore is the fourth most densely populated country in the world with a population density of 6369 persons/km^2 as at 2008 and can be considered as a land scarce country [35]. Early to 1995 and after independence, Singapore's public transport systems were managed separately, but in 1995 most land transport functions were brought together by the Land Transport Authority (LTA) [43]. With the plans and policies for implementation to provide a world class transport system, since 1998 onwards Singapore is known to be a heavily motorized country with the most developed road system in the Asia-Pacific region [44].

In Singapore, the number of automobiles has been steadily increasing as a consequence of rapid urbanization and economic growth. As an example, based on LTA data, the total number of vehicles in Singapore were 688,811 in 1999 and 956,704 in 2011 (LTA, 1999; LTA, 2011) [45, 46]. In the present study, we have studied the effects of vehicles on local air quality at the human breathing level in 2010. We give the vehicle details in the break-up of different categories for 2009, as described below. As per the data of LTA (1999), cars accounted for 61% of the total vehicle population. These cars are all gasoline driven and a majority of them have catalytic converters. Goods vehicles, running on gasoline and diesel, accounted for

18%. The gasoline driven two wheelers, i.e., motorcycles and scooters accounted for approximately 16% of the vehicle population. Buses and taxis accounted for 2% and 3%, respectively, and these vehicles run with diesel fuel. Although the percentage composition of the buses and taxis in the total population is low, the overall vehicle kilometers traveled by them on road are quite high.

Singapore's strategy for reducing air pollution from vehicles focuses on two main aspects: improving the fuel quality to reduce emissions and management of traffic to control the increase in the number of vehicles [47]. The high expense of owning and operating a vehicle in Singapore has effectively controlled the growth of vehicles. In addition, car owners pay annual road taxes based on the engine capacity of their vehicles. "Singapore has also introduced a full-fledged use of the Electronic Road Pricing System, commonly known as ERP, for vehicles on major expressways. According to this system, vehicles traveling on certain expressways at certain time periods have to pay a toll" (LTA, 2011). The Singapore government introduced a quota system known as Certificate of Entitlement (COE), in January 1990. Under this system, a vehicle entitlement is valid for 10 years from the date of registration of the vehicle. If the owner wishes to continue using the vehicle on expiration of the vehicle entitlement, he has to pay a revalidation fee.

2.2. Details of the measurement program

To achieve the objectives of this study, field measurements were carried out at four different sites: (i) road side in the National University of Singapore campus (NUS), (ii) road side in the vicinity of expressway (EXW), (iii) central express tunnel (CTE), and (iv) a remote site near coastal region (RME). These measurement sites were selected on the basis of intensity of traffic flow. The parameters of measurement included were air quality (BC, CO, and NO), meteorology (wind speed and wind direction), and traffic volumes (car, bus, taxi, motorbike, and others). **Table 1** presents the details of the field measurement program with information of the monitoring sites.

Measurement site	Site code	Characteristics of sites	Details of field measurement			
			Air quality	Meteorology	Traffic survey	Duration of study
Road side in National University of Singapore campus	NUS	Medium traffic volume	BC, CO, NO	Wind speed, wind direction	Car, bus, taxi, bike, etc.	13–16 May, 2010
Road side of expressway	EXW	Higher traffic volume	BC, CO, NO	Wind speed, wind direction	Car, bus, taxi, bike, etc.	22–23 July, 2010
Central express (CTE) tunnel	CTE	Higher traffic volume	BC, CO, NO	Wind speed, wind direction	Car, bus, taxi, bike, etc.	02–04 June, 2010
Remote site near coastal region	RME	Negligible traffic influence	BC, CO, NO	Wind speed, wind direction	Car, bus, taxi, bike, etc.	05 May, 2010

Table 1. Monitoring sites and details of the measurement program.

To get insights into the behavior of air pollution patterns during weekdays and weekends, measurements at the NUS site were conducted from Thursday to Sunday. Automated equip-

http://dx.doi.org/ 10.5772/64692

ment was maintained in an air conditioned mobile enclosure with the sampling tubes protruding out from the window of the van. A video camera was used to capture the traffic flows and to characterize the composition of the vehicles traveling at each measurement site. In the tunnel site, CTE, the monitoring point was at the center of the tunnel, so that the field measurements could be carried out under smooth flowing traffic conditions. The wind speed and wind direction were measured with digital wind vane and anemometer, respectively.

2.3. Measurements of BC, CO, and NO

In this study, field equipment was placed at a distance of 5.0 m from the road at all measurement sites except the tunnel site, CTE. Air pollutants were measured at approximately 2.0 m height above the ground except the NUS site. Measurements at NUS were done on the rooftop of the Atmospheric Research Station, which is about 15 m above the ground level.

For the measurement of BC in ambient air, the optical and thermal techniques are widely used. In the past, various research groups used these techniques to estimate the BC concentration [48]. However, aethalometer is the only equipment that is used for the real-time measurement of BC [49]. To get measurement information with short-term peaks in quasi-real time, aethalometer is the best option. The model AE–20UV Aethalometer (Magee Scientific Company, Berkeley, CA) was used to measure BC in real time for this study. The sample flow rate was 5.0 L/min, and sampling time base was 5 min. The principle of measurement method is based on the optical attenuation of light by particles collected on the quartz fiber filter, which is summarized as follows: (i) light from a stabilized lamp is split and passes the sampling portion and a reference blank portion of the filter, (ii) the intensity of the transmitted light is determined by the light sensors placed after the filter, (iii) thus, changes of light transmitted through the sampling portion of the filter due to collected absorbing aerosols are detected and recorded as changes in optical attenuation, and (iv) finally, assuming a constant specific attenuation cross-section of BC, the concentration of BC is calculated. We used the original laboratory calibration factor of 17 m^2/g, as recommended by the manufacturer.

CO was measured by a real-time CO analyzer, Thermo Environmental Instruments (TEI), Model 48C. The instrument is based on the principle that CO absorbs infrared radiation at the wavelength of 4.6 μm. The model 42C, NO–NO_2–NO_X analyzer from TEI, was used to measure NO based on the principle of chemiluminescence. It is based on the principle that NO and O_3 react to produce a characteristic luminescence with intensity linearly proportional to the NO concentration.

3. Results and discussions

3.1. Overall results

Table 2 presents the overall results of the ambient levels of BC, CO, and NO at four measurement sites. On the basis of 1 h observations, the mean BC varied as the lowest at the RME site (1.7 $\mu g/m^3$) to the highest at the CTE site (45.6 $\mu g/m^3$). Similarly, CO and NO varied from 336.3

ppbv (parts per billion by volume) at the RME site to 8322.4 ppbv at the CTE site and 60.0 ppbv at the NUS site to 100.2 ppbv at the EXE site, respectively. Concurrent measurements of NO at CTE and RME were not possible due to instrumental problems. From the results obtained at the four sites, it can be clearly seen that the tunnel, CTE, experienced the highest air pollution followed by the expressway site, EXW, and the site with the least influence of vehicles (RME) experienced the lowest air pollution. Therefore, the contribution of vehicles to the ambient levels of these air pollutants is significant. The highest levels of BC and CO measured at the CTE site can be attributed to the high traffic flow of vehicles of different types with reduced ventilation as well as little homogeneous mixing of air pollutants inside the tunnel. The relation between various types of the volume of vehicles and the resulting levels of air pollutants during the measurement period can provide better insights into the role of vehicles on the profiles of the measured concentrations of air pollutants. This aspect of the study is discussed in a subsequent section.

Measurement site*	BC (μg/m³)			CO (ppbv)			NO (ppbv)		
	Mean	SD	*N*	Mean	SD	*N*	Mean	SD	*N*
NUS	6.2	3.8	94	582.9	404.3	95	60.0	58.3	87
EXW	7.2	1.9	45	1384.8	325.4	46	100.2	37.1	47
CTE	45.6	9.6	14	8322.4	886.6	15	NA	NA	NA
RME	1.7	0.5	23	336 .3	162.5	24	NA	NA	NA

*Experimental results are based on average of 1 h observations.
SD: standard deviation; *N*: number of observations; NA: not applicable.

Table 2. Overall results of BC, CO, and NO during the measurement period.

To compare the results obtained from this study with those from other appropriate studies reported in the literature, **Table 3** presents the relevant data on BC, CO, and NO. In general, the ambient concentrations of BC, CO, and NO in Singapore are comparable to those reported for other urban areas of the world.

Location	BC (μg/m³)	CO (ppbv)	NO (ppbv)	Particular of site	Reference
Essen East, Germany	NA	1 921.4	60.0	Urban	[50]
Düsseldorf–Mörsenbroich, Germany	NA	2707.4	111.2	Urban	[50]
Helsinki, Finland	1.5	NA	NA	Sub Urban	[21]
Los Angeles, USA	4.4	230	NA	Urban	[38]
Toronto, Canada	NA	NA	70.5	Urban	[37]
Aachen, Germany	9.4	NA	NA	Urban	[36]
Aachen, Germany	1.5	NA	NA	Rural	[36]
Barcelona, Spain	3.6	NA	14.2	Urban	[40]

NA: not available.

Table 3. Ambient concentrations of BC, CO, and NO reported for different parts of the world.

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3.2. Diurnal variations of BC, CO, and NO

To get a better insight into the diurnal variations of BC, CO, and NO, the measurement data obtained for every 5-min interval observations were averaged during all hours of the day. In this section, we have considered two measurement sites (NUS and EXW) for examining diurnal variations, because of the similar pollution status of BC at these two sites. However, the tunnel site, EXW, and the remote site, RME, experienced different pollution status and traffic flows (**Table 2**). Therefore, in this paper, we discuss the pollution and traffic patterns at the CTE site in a separate section. The site, RME, did not show any specific trends of air pollution. **Figure 1** shows the diurnal variations of BC, CO, and NO with hourly average observations at NUS. BC showed pronounced peaks in the morning traffic hours (7:00–11:00) with a maximum concentration of 8.9 μg/m^3 at 9:00, indicating that vehicular emission is an important source of BC (**Figure 1**). Similarly, CO and NO showed pronounced peaks in the morning traffic hours (7:00–

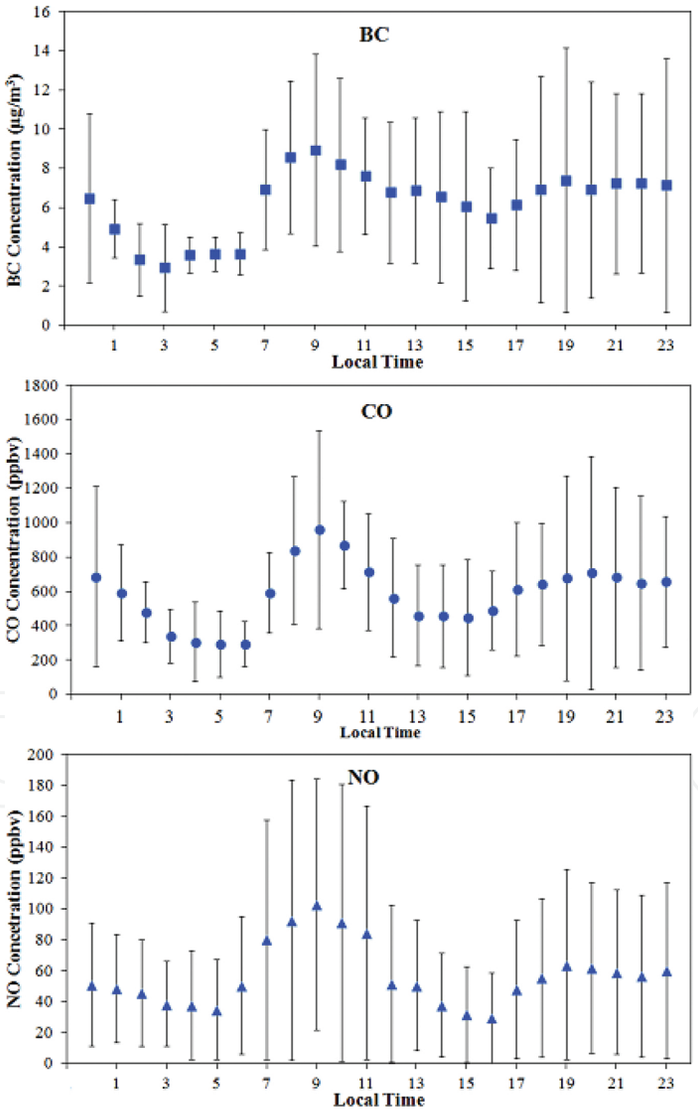

Figure 1. Diurnal variations of concentrations BC, CO, and NO at the NUS site. The error bars represent the corresponding standard deviations.

11:00) with maximum concentrations of 959.7 and 102.8 ppbv, respectively, at 9:00, indicating that vehicular emission is an important source of CO and NO (**Figure 1**).

The concentrations of BC, CO, and NO remained low in the afternoon. There were no significant evening rush hour peaks, which could be probably due to unstable atmospheric conditions induced by long hours of sunshine and hence improved dispersion of traffic emissions during the afternoon. Moreover, being a tropical country, the solar radiation in Singapore remains high even during the evening rush hour, thereby resulting in the enhanced vertical mixing of air pollutants. This leads to dilution of BC, CO, and NO concentrations. Relatively higher concentrations in the early morning could be attributed to low mixing heights and reduced dispersive conditions. Being a coastal city, Singapore's air quality is also influenced by land breeze and sea breeze. Land breeze blowing during early morning brings in contaminated air, whereas the sea breeze that blows during afternoon brings in relatively clean air. In addition, the traffic flow of heavy-duty/utility vehicles is relatively higher in the morning than during other periods of the day.

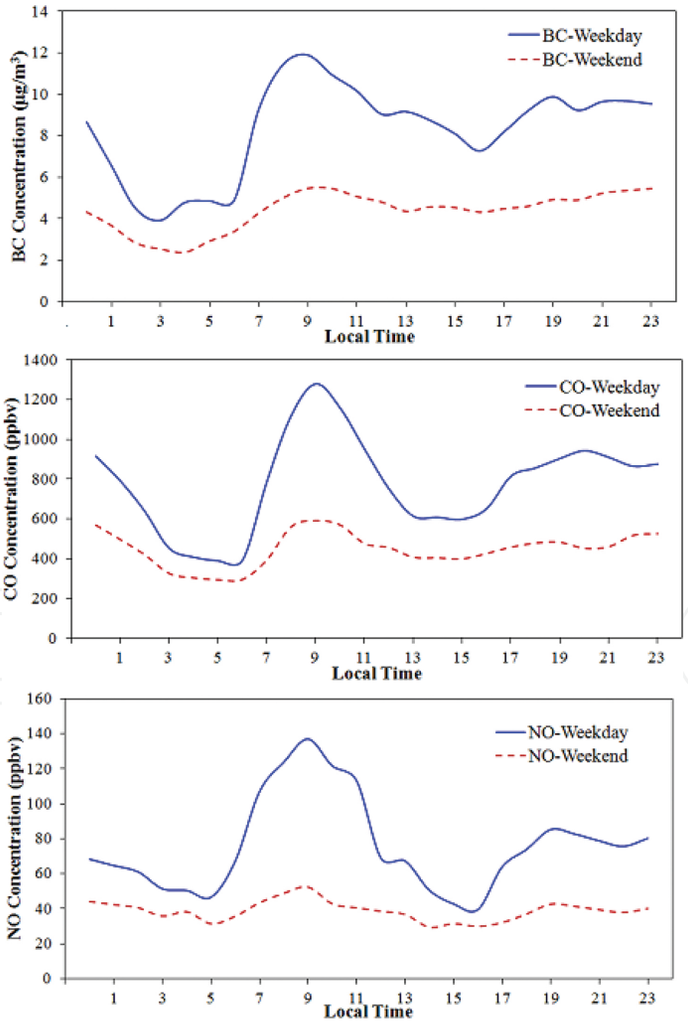

Figure 2. Diurnal variations of concentrations of BC, CO, and NO at the NUS site during weekday and weekend.

In the case of the expressway site, EXW, it was observed that the traffic volume remained high on the expressway even in the noon; there was no decrease in the concentration in the afternoon in contrast to the diurnal profiles of BC, CO, and NO concentrations in the ambient air at NUS. Furthermore, the effect of dispersion on the concentrations of air pollutants does not play a significant role at this site because the measurements were performed at the ground level.

To examine the effect of rush hour traffic, the mean diurnal variations of BC, CO, and NO were divided into weekdays (Thursday and Friday) and weekends (Saturday and Sunday). This is to be noted that the measurement at the NUS site was done for 4 days with two weekdays and two weekends. **Figure 2** shows the diurnal trends of these pollutants during weekdays and weekends. The morning peak, which is usually seen between 7:00 and 11:00 on weekdays, does not exist on weekends. The peak concentrations on weekdays were 11.9 μg/m^3, 1279.6 ppbv, and 137.1 ppbv for BC, CO, and NO, respectively. However, during weekends, the peak values seemed to be less than half of respective peaks of weekdays (the peaks on weekend were 5.5 μg/m^3, 590.8 ppbv, and 52.1 ppbv for BC, CO, and NO, respectively). From these observations, it can be confirmed that vehicles play a major role in making significant contributions to the prevailing air pollution levels in Singapore.

In the eastern United States, at Uniontown, PA, the diurnal variation of BC concentrations was measured in 1990 [49]. A clear peak was observed between 6:00 and 10:00. There was no significant evening rush hour peak, but instead slightly elevated concentrations were observed from 20:00 to 23:00. Pakkanen et al. [21] analyzed in Helinski reported that the peak in hourly average BC concentrations was observed during the morning and evening rush hours on weekdays. Weekends showed relatively stable hourly average concentrations. Therefore, based on these comparisons, it appeared that our observations on BC due to traffic emissions were consistent with those from the other studies reported in the literature.

3.3. Measurements in the tunnel site

The field measurements at CTE were conducted under varying traffic composition to examine the effect of traffic volume and the composition on the levels of BC and CO measured in the traffic tunnel. Air sampling and traffic surveys were performed on 2 days from 13:40 to 19:25 on day 1 and from 9:45 to 16:10 on day 2. The average traffic count on the first day was 3900 vehicles per hour, and the vehicle fleet comprised 8% motorbikes, 3% heavy-duty vehicles, 24% pickups and vans, 49% cars, 1% buses, and 15% taxis. On the second day, the traffic count was around 4400 vehicles per hour, almost 10% higher as compared to the first day. The traffic composition was similar to the first day consisting of 6% motorbikes, 3% heavy-duty vehicles, 24% pickups and vans, 48% cars, 1% buses, and 18% taxis. Heavy-duty vehicles, pickups and vans, buses, and taxis are driven by diesel fuel, whereas the fuel used in motorbikes and cars are with gasoline. Based on the classification by fuel types, the fraction of diesel-driven vehicles on the first day was 0.43 and on the second day was 0.46.

The concentration of BC was 40.9 μg/m^3 on the first day and 49.9 μg/m^3 on the second day. The concentration of CO was 8394.5 ppbv on the first day and 8247.3 ppbv on the second day. Average concentrations of the air pollutants at different sampling locations are also shown in **Table 2**. As can be seen from **Table 2**, the concentration of air pollutants in the CTE was

observed to be much higher than those at other sampling sites. Such higher concentrations at CTE site could be due to a combination of three factors: (1) measurements in the tunnel were performed very close to the emission source (vehicles), (2) there was a substantially high volume of traffic flow in the tunnel, and (3) there was a limited vertical dispersion of BC and CO in the confined environment in the tunnel.

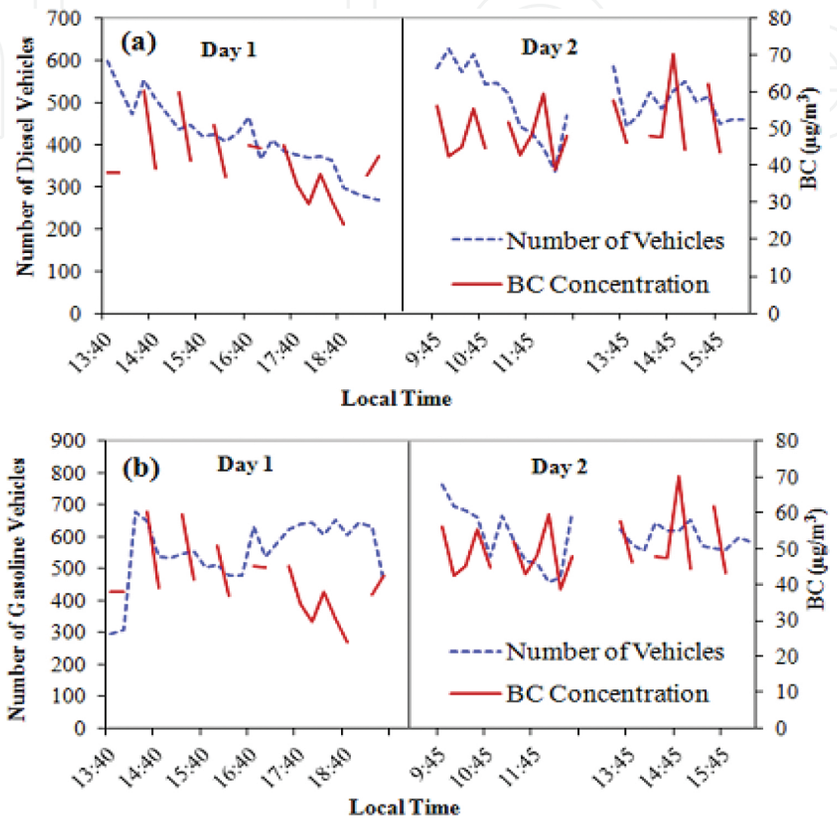

Figure 3. Variations of BC concentration with the number vehicles: (a) diesel-driven vehicles, and (b) gasoline-driven vehicles.

Figures 3(a) and **(b)** show the variations of BC with the total number of diesel-driven vehicles and the total number of gasoline-driven vehicles, respectively, for 2 days. It should be noted that the concentration plots were made based on the observations of 5-min interval. The concentration of BC could not be continuously obtained in the tunnel during these 2 days as the instrument was automatically set in the calibration mode for every hour. Hence, the variations of BC in **Figures 3(a)** and **(b)** do not show their continuous profiles. The increase in the frequency of calibration is especially important as the instrument tends to be saturated with such a high concentration of BC in ambient air of the tunnel. The variations of CO concentration with traffic are shown in **Figures 4(a)** and **(b)**.

The variation in the concentration of BC followed closely the trend in percentage of diesel-driven vehicles. However, such a similarity in the trends of concentration of BC and gasoline vehicles was not seen. For example, on the first day of the air sampling in the tunnel, there was a gradual decrease in the number of diesel-driven vehicle between 16:40 and 19:40, which was

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accompanied by a similar decrease in the concentration of BC. On the other hand, the number of gasoline vehicles remained almost the same. The link between the number of diesel-driven vehicles and the corresponding change in the concentration of BC is further strengthened by the variability in the concentration of CO. In other words, the concentration of CO follows a trend very similar to the number of gasoline vehicles, which is in contrast to the pattern of BC.

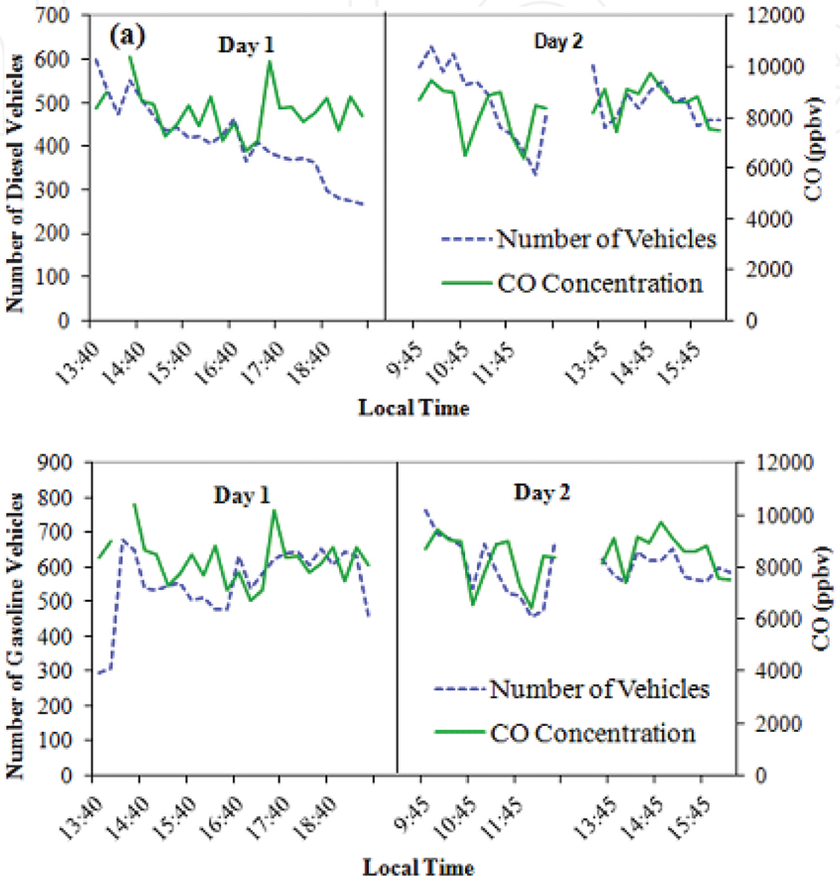

Figure 4. Variations of CO concentration with the number of vehicles: (a) diesel-driven vehicles, and (b) gasoline-driven vehicles.

This finding strongly suggests that diesel-driven vehicles emit much higher levels of BC than gasoline-driven vehicles. This observation is consistent with the previous studies, which have indicated that BC is more abundant in heavy-duty diesel-fuelled exhaust than in light-duty gasoline-fuelled vehicle exhaust [51, 52]. The amount of BC emitted is highly variable, and depends on the vehicle condition, its age, quality of the fuel used, maintenance, the speed of vehicles, and the operating modes of drivers. Miguel et al. [52] did the measurement of CO and BC in two bores of the Caldecott tunnel in California: one bore was influenced by heavy-duty diesel truck emissions; a second bore was reserved for light-duty vehicles. Miguel et al. [52] found that concentration of BC in truck-influenced bore was higher by a factor of 5 in spite of higher traffic volume in other bore. The concentration of CO was higher in the bore dominated by light duty vehicles. According to Miguel et al. [52] light-duty gasoline-driven vehicles and heavy-duty diesel trucks, emitted, respectively, 30 ± 2 mg and 1440 ± 160 mg of

fine BC particles per kg of fuel burned. Gray and Cass [53] estimated that diesel-driven vehicles were responsible for 60% of total BC emissions. Steiner et al. [54] showed that the contribution of BC to total suspended particulate matter (TSP) emitted from a spark ignition engine is 11%. On the other hand, the contribution of BC to TSP in the case of particles emitted from diesel engine ranges from 50% to 80%.

3.4. Correlation of BC with CO and NO

From the time-series plots of BC and CO with vehicles (in Section 3.2), it has been observed that the traffic flow makes a major contribution to the existing levels of these air pollutants. To further confirm this trend, we reviewed the relevant literature on studies done in various parts of the world. From the literature review, it could be concluded that vehicular emissions are strongly correlated with the levels of CO and NO under normal conditions and these pollutants are indicators of traffic emissions in the urban environment [55, 56]. From the estimation of USEPA [57], it is clear that transportation sources were responsible for nearly 72% of total CO emissions, 40% of NO_x, and 31.5% of hydrocarbons in the United States in 1991. In the UK west Midlands conurbation, of which Birmingham is the major city, 98% of CO and 85% of NO_x emissions arise from road traffic. In Australia, the local transport was responsible for up to 63% of NO_x emissions and 95% of CO emissions. In Mexico City, traffic accounts for 99% of the CO and 70% of NO_x [58].

Therefore, to provide further evidence that BC is influenced by traffic flows and to estimate the contribution of traffic flows toward the total ambient levels of BC, the statistical correlation of BC with CO and NO were examined. In this interpretation, we considered the data from the sites of NUS and EXW as we made observations of air pollutants in the open ambient air at

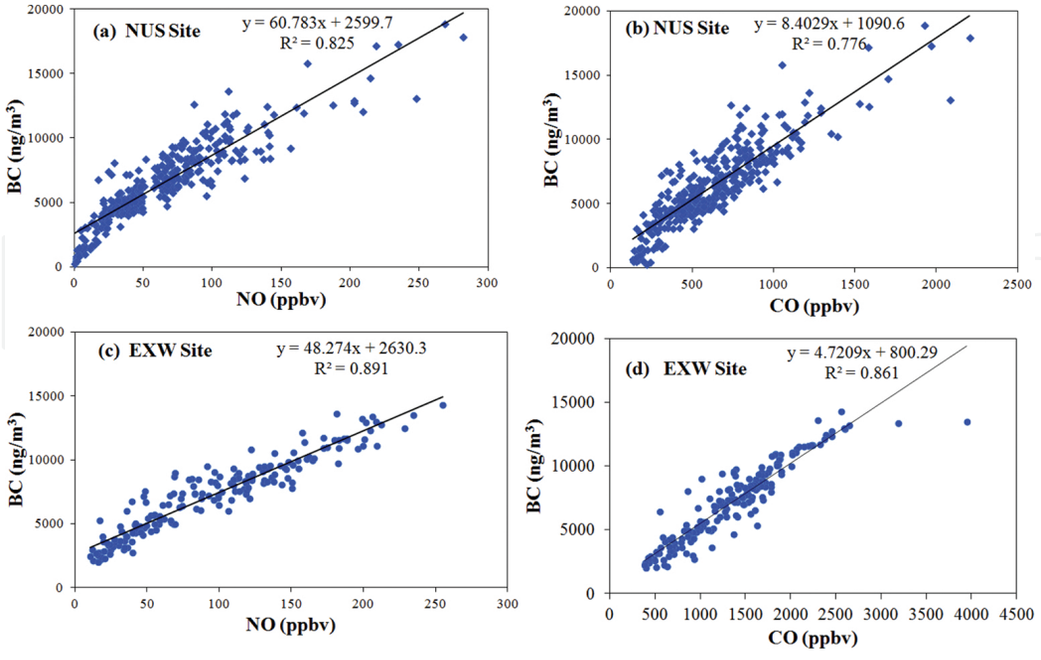

Figure 5. Correlation plots of BC with NO and CO at the NUS and EXW sites: (a) BC vs. NO at NUS, (b) BC vs. CO at NUS, (c) BC vs. NO at EXW, and (d) BC vs. CO at EXW.

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these measurement sites with substantial traffic volumes. Linear regressions of 15 min observations of BC versus NO and BC versus CO were performed from the data of these two sites individually. High correlations of BC with NO ($R^2 = 0.825$, $n = 343$, $p < 0.01$) and with CO ($R^2 = 0.776$, $n = 352$, $p < 0.001$) were observed at NUS (**Figures 5a** and **b**). Similarly, the EXW site also experienced high correlations of BC with NO ($R^2 = 0.891$, $n = 183$, $p < 0.001$) and with CO ($R^2 = 0.861$, $n = 181$, $p < 0.001$) as shown in **Figures 5(c)** and **(d)**, respectively. These high correlations suggest a strong association between vehicular emissions and levels of BC. With these highly significant correlations, we have attempted to assess the contribution of vehicles to the BC concentration in the ambient air using the linear regression equations obtained from the plots of BC and CO in **Figures 5(b)** and **(d)** at the sites, NUS and EXW.

A linear regression of BC with CO yielded the following equations at NUS (Eq. 1) and EXW (Eq. 2):

$$BC = 8.4 \times CO + 1090.6 \tag{1}$$

$$BC = 4.7 \times CO + 800.2 \tag{2}$$

where BC is in ng/m^3 and CO is in ppbv. To estimate the contribution of BC from vehicular sources, we assumed that 90% of the total emission of CO is traffic generated. Therefore, the background CO in the absence of traffic would be 58.3 and 138.5 ppbv at NUS and EXW, respectively (i.e., 10% of 1 h average CO concentration; see **Table 2**). For these values of CO, Eqs. (1) and (2) gave an estimate of BC concentration of 1.6 and 1.4 μg/m^3 at NUS and EXW, respectively, in the absence of traffic. The average ambient concentrations of BC at NUS and EXW were 6.2 and 7.2 μg/m^3 (**Table 2**). Thus, the average concentrations of BC due to traffic were 4.6 and 5.8 μg/m^3 at NUS and EXW. Finally, the contributions of vehicular traffic to the total BC concentration were estimated to be 74% and 80% at NUS and EXW, respectively. While EXW is close to the road, NUS is relatively far from the road. Consequently, the roadside environment involving human exposure to higher BC emissions could cause more health effects. It should be noted that the approach to estimate the traffic contribution toward air pollutants through the regression analysis has been used by other investigators. For example, Lim et al. [59] estimated the contribution of traffic to PAHs concentration using a regression equation.

3.5. Correlation of vehicles with BC, CO, and NO

As described in Section 3.3, the traffic flow showed a significant influence on the levels of BC and CO. It is also evident from the past studies [55, 56] that vehicles have a dominant influence on the ambient levels of NO concentration. Therefore, to examine the role of vehicles on the levels of BC, CO, and NO, correlation coefficients were estimated between various species using Minitab 15 English. In this approach, we classified the entire observations into two parts with respect to the site characteristics (results are presented in **Tables 4a** and **b**). It should be noted that the observations were made in the ambient air at the sites, NUS and EXW. However,

in the case of CTE, the observations were made inside the tunnel. In the correlation analysis, all pollutant concentrations and their corresponding traffic numbers (diesel-driven and gasoline-driven) were included at both NUS and EXW sites. The correlation analysis at CTE was made separately to ensure that the assessment of correlation was based on traffic flows only.

	BC	**CO**	**NO**	N_d	N_p
BC	1.00				
CO	**0.85**^{III}	1.00			
NO	**0.92**^{III}	0.21	1.00		
N_d	**0.84**^{III}	**0.29**^{II}	**0.85**^{III}	1.00	
N_p	**0.27**^{II}	**0.82**^{III}	**0.88**^{III}	**0.49**^{III}	1.00

N_d: number of diesel-driven vehicles; N_p: number of gasoline-driven vehicles.
Bold marks are statistically significant. Superscripts II and III denote that correlation is significant at $P < 0.01$ and $P < 0.001$.

Table 4(a). Correlation matrix for NUS and EXW sites.

	BC	**CO**	N_d	N_p
BC	1.00			
CO	**0.91**^{III}	1.00		
N_d	**0.92**^{III}	**0.56**^{II}	1.00	
N_p	**0.53**^{II}	**0.94**^{III}	**72**^{III}	1.00

N_d: number of diesel-driven vehicles; N_p: number of gasoline-driven vehicles.
Bold marks are statistically significant. Superscripts II and III denote that correlation is significant at $P < 0.01$ and $P < 0.001$.

Table 4(b). Correlation matrix for CTE site.

Table 4(a) presents the correlation matrix of the sites of NUS and EXW. The observations from this correlation analysis are summarized as follows: (1) BC concentration showed significant correlations with CO and NO, (2) BC concentration showed strong and significant correlations with the number of diesel-driven vehicles monitored during that period, (3) CO concentrations showed a strong and significant correlation with the number of gasoline-driven vehicles observed during that period, and (4) NO showed strong and significant correlations with both gasoline-driven and diesel-driven vehicles. Hence, it could be concluded that the BC concentrations were mainly influenced by the diesel-driven vehicles. However, the CO concentrations were predominantly influenced by the gasoline-driven vehicles.

Table 4(b) presents the correlation matrix of the site, CTE. It should be noted that the parameter, NO concentration, was not included due to unavailability of observational data. Overall, it can be seen that the concentrations of BC and CO were influenced by diesel-driven and gasoline-driven vehicles, respectively.

http://dx.doi.org/ 10.5772/64692

3.6. Effect of wind speed and wind direction on pollutants

In general, the variability of pollutant concentration levels strongly depends on the origin of the air masses arriving at the sampling site and the concentration of pollutants in the ambient air is influenced by the direction from which wind blows. In this study, we did not find any specific trends between wind directions and the concentrations of air pollutants at the measurement sites. For example, at the NUS site, we observed that winds mainly blew from the Northeast during the morning rush hour. However, on the following day, there was a change in the wind direction. To assess the role of meteorology in the variation of ambient levels of BC, CO, and NO, we estimated correlation coefficients between pollutant levels, wind speed, and wind direction using Minitab 15 English. As explained in Section 3.5, we considered the NUS and EXW sites in a single platform for correlation analysis and further interpretation. **Table 5** presents the correlation matrix of BC, CO, NO, wind speed, and wind direction. It was observed that the wind direction had lower correlations with the air pollutant concentrations during the measurement period, indicating that there was little change of air pollution levels with the change in wind direction. The reason for such observations could be due to the fact that winds blowing from the South bring in clean marine air resulting in relatively low concentrations of air pollutants; however, the northerly winds originated from land air mass. Being influenced by human activities, the land air mass led to the enhancement in the level of air pollutants.

	BC	CO	NO	WS	WD
BC	1.00				
CO	**0.85**^{III}	1.00			
NO	**0.92**^{III}	0.21	1.00		
WS	**-0.68**^{III}	**-0.72**^{III}	**-0.67**^{III}	1.00	
WD	**0.14**^{I}	**0.16**^{I}	**0.15**^{I}	**0.13**^{I}	1.00

WS: wind speed; WD: wind direction.
Bold marks are statistically significant. Superscripts I and III denote that correlation is significant at $P < 0.05$ and $P < 0.001$.

Table 5. Correlation matrix meant for meteorology at NUS and EXW sites.

In general, wind speed is considered as one of the important parameters affecting the concentration of air pollutants. It determines the time taken to travel from a source to a given receptor and the total area over which the air pollutant would be dispersed. The wind speed showed strong and significant negative correlations with BC, CO, and NO. Higher wind speeds result in better mixing of air pollutants, causing their dilution. As the wind speed gets lower, air pollutants tend to get accumulated due to poor dispersion of air. Therefore, the concentration of BC < CO, and NO increased at low wind speeds. Harrison et al. [7] compared the daily mean elemental carbon concentration and wind speeds at Birmingham, and found quite a similar relationship as the one observed in this study.

4. Conclusions

The present study investigated the on-road emissions of BC, CO, and NO under real moving traffic conditions at four different measurement sites (NUS, EXW, CTE, and RME) of Singapore, an urban environment in the Asia-pacific region. On the basis of 1 h observations, the mean BC varied from the lowest value at the RME site (1.7 μg/m^3) to the highest one at the CTE site (45.6 μg/m^3). Similarly, CO and NO varied from 336.3 ppbv at the RME site to 8322.4 ppbv at the CTE site and 60.0 ppbv at the NUS site to 100.2 ppbv at the EXE site, respectively. At the NUS site, BC showed pronounced peaks in the morning traffic hours (7:00–11:00) with a maximum of 8.9 μg/m^3 at 9:00, and CO and NO showed pronounced peaks in the morning traffic hours (7:00–11:00) with maximum values of 959.7 and 102.8 ppbv, respectively, at 9:00, indicating that vehicular emission is an important source of BC, CO, and NO. The concentration of air pollutants in the tunnel site, CTE, was observed to be much higher than those at other measurement sites. The study revealed that diesel-driven vehicles had a major influence on the ambient BC concentration. However, gasoline-driven vehicles had more influence on ambient CO concentrations. The contribution of on-road vehicles to the total BC concentration was estimated to be 74% and 80% at NUS and EXW, respectively. The statistical analysis of data obtained in this study showed significant correlations between BC, CO, and NO, confirming that the on-road vehicles were the dominant source of these air pollutants. A significant negative correlation between wind speeds and concentrations of BC, CO, and NO was observed, confirming that the lower wind speed was mainly responsible for the accumulation of air pollutants in the sampling location due to poor dispersion of air.

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