Real-Time Characterization of Particle-Bound Polycyclic Aromatic Hydrocarbons at a Heavily Trafficked Roadside Site

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ABSTRACT

A continuous photo-electric aerosol sensor (PAS) was used to characterize the particle-bound polycyclic aromatic hydrocarbons (p-PAHs) content at a heavily trafficked roadside site in Hong Kong. The measurements at the roadside station were performed for nine months from January to September 2005. For the total quantified p-PAHs, the PAS output shows good correlations to the data obtained with the offline quartz-fiber aerosol filter collection method. The average concentration of corrected p-PAHs was 5.3 ± 3.7 ng/m^3, ranging from 0.2 to 22.0 ng/m^3. The p-PAHs concentrations increased in the daytime in accordance with the heavy amount of road traffic, and decreased in the nighttime due to less traffic. Hourly p-PAHs concentrations had high correlation coefficients with all goods vehicles (> 0.9) and large buses (~0.8), showing that diesel vehicle emissions are the primary source of p-PAHs. The day-to-day variation of p-PAHs is significant, because it was influenced by various factors (e.g., traffic levels, mixing height, and potential pollution sources).

Keywords: Polycyclic aromatic compound; PAS; Black carbon; Roadside measurement.

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are one of major concerns in air quality control because some compounds have been identified as carcinogen and mutagen (IARC, 1983). Arising from incomplete combustion and high temperature pyrolysis process of fossil fuels and other organic materials (Baek *et al.*, 1991), PAHs present in the atmosphere as both of gas- and particle-phases (Bidleman *et al.*, 1986) based on their molecule weights. Carcinogenic PAHs are mainly found in the particle phase (Lyall *et al.*, 1988) and the inhalation of airborne particle-bound polycyclic aromatic hydrocarbons (p-PAHs) poses a lung cancer risk in humans (Lan *et al.*, 2004).

A few studies on p-PAHs have been previously conducted in ambient atmosphere of Hong Kong. Vehicle exhaust is well known to be the most significant source for p-PAHs

in Hong Kong (Zheng *et al.*, 1997; Panther *et al.*, 1999; Lee *et al.*, 2001; Ho and Lee, 2002; Ho *et al.*, 2002; Guo *et al.*, 2003). In addition, cooking fume can contribute to the p-PAHs emission due to characteristics of Chinese stir-frying cooking style (Chen *et al.*, 2007; Huang *et al.*, 2011). In order to advance the understanding of p-PAHs emitted from vehicles, real-world vehicle emission factors for p-PAHs were determined (Ho *et al.*, 2009). The vehicle emission factors for the main p-PAHs including pyrene, fluoranthene, chrysene, phenanthrene, and naphthalene accounted for ~70% of total measured p-PAHs (Ho *et al.*, 2009).

Previous studies on p-PAHs were mainly applied conventional filter-based sampling, followed by the gas chromatography/mass spectrometry (GC/MS) analysis. However, it is currently impractical to perform better time resolution data using traditional time-integrated particulate samplers. Continuous monitors has been attracted more and more attentions because they can not only capture the properties of particles in better resolutions but also are more economical to be operated by reducing frequency of site visits and eliminating the needs of laboratory facilities and the analytical costs. At present, a steadily increasing in amount of literatures describing the development and

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evaluation of continuous Photoelectric Aerosol Sensor (PAS) has emerged over the past decade (Agnesod *et al.*, 1996; Junker *et al.*, 2000; Chetwittayachan *et al.*, 2002a, c; Leutwyler *et al.*, 2002; Ott and Siegmann, 2006). The previous studies found that the PAS provides satisfactory precision in most sampling locations. In this study, the PAS was applied to investigate the temporal variations of p-PAHs at a roadside environment of Hong Kong. The potential impact factors such as vehicle numbers and meteorological parameters were examined as well. At the same time, comparison experiment of p-PAHs was conducted between the photoelectric method and conventional method.

METHODOLOGIES

Sampling Site

The p-PAH monitoring was carried out at the Air Quality Supersite (22.30°N, 114.17°E, Fig. 1) built in the campus of the Hong Kong Polytechnic University (PU), Hung Hom. The campus locates in a street canyon in a combined residential and commercial area near the Victoria Harbor. The Supersite is ~1 m away from the curb of Hong Chong Road, where is approximately 30 m in width with four lanes for two-ways directions, representing the busiest cross-harbor tunnel in Hong Kong. The monitoring station is ~400 m away from the tunnel entrance. Vehicle numbers, speeds and types were counted at the toll gates. Besides restaurants and household cooking activities, industrial emission or other anthropogenic sources were not found in the vicinity. This location is a typical roadside site area of Hong Kong (Lee *et al.*, 2006).

The average traffic speed in the lanes near the monitoring station was slow due to periodic switching of traffic lights. Braking was also frequent near the site because of the presences of public bus stations (100 meters to the southwest),

tolls (200 meters to southeast), and intersection (200 meters to the southwest). However, all vehicle engines were at a warming-up status. The Hong Chong Road and its surrounding streets are paved. The toll data provides counts for nine vehicle classes, including (i) private car, (ii) moto cycle, (iii) private/public light bus, (iv) private/public single-decked bus, (v) private/public double-decked bus, (vi) ≤ 5.5 ton goods vehicle, (vii) 5.5–24 ton goods vehicle, (viii) > 24 ton goods vehicle, and (ix) taxi. The private cars and moto cycles were fueled with gasoline; > 90% taxis were fueled with liquid petroleum gasoline (LPG); and the remains were almost fueled with diesel. The daily traffic number is around 120,000 on average.

According to the toll traffic statistic, there are two rush sections with one in the morning (8:00–10:00) and the other in the evening (17:00–19:00). The diurnal variations of vehicle fractions are quite similar over the sampling days on this road, with higher traffic number in daytime and lower values at nighttime. During daytime, over 6,000 vehicles per hour passed the sampling site, of which 30% are diesel-fueled vehicles, 60% are gasoline-fueled vehicles, and 10% are LPG-fueled taxis. Most of diesel-fueled vehicles are light-duty goods vehicles (≤ 5.5 ton) (~90%) and the remaining (10%) are medium- and heavy-duty goods vehicles (> 5.5 ton). The average hourly gasoline-fueled vehicles ranged from 35% (minimum at 11:00–12:00) to 60% (maximum at 19:00–20:00) and diesel-fueled vehicles varied from 10% (minimum at 22:00–23:00) to 50% (maximum at 11:00–12:00).

Online Measurements for p-PAHs

Five-minute p-PAHs were measured by PAS (Model 2000, Ecochem, League City, TX, USA) from 15 January to 31 September 2005. The PAS measures total p-PAHs adsorbed on carbon particles. With using an Excimer lamp,
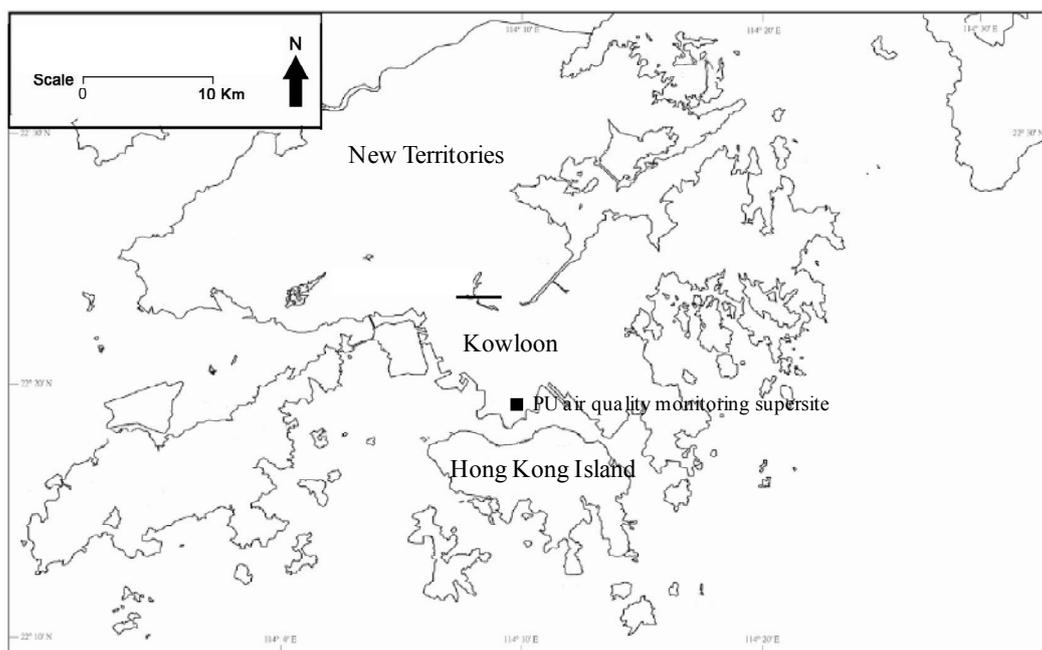
Fig. 1. Sampling location, PU air quality monitoring Supersite.

the aerosol flow is exposed to UV radiation. The lamp offers a high intensity, narrow band source of UV radiation. The aerosol particles which have PAH molecules adsorbed on the surface emit electrons when they are exposed to UV radiation. The electrons are subsequently removed when an electric field is applied. The remaining positively charged particles are collected on a filter inside an electrometer, where the charge is measured. The resulting electric current establishes a signal which is proportional to the concentration of total p-PAHs.

Offline Sampling and Chemical Analysis for Individual p-PAHs.

In this study, three-hour aerosol samples were concurrently collected onto the quartz-fiber filters by the Desert Research Institute (DRI, Reno, NV, USA) portable source sampler, parallel with the PAS continuous monitoring. Aerosol samples were collected on two parallel 47 mm quartz-fiber filters (Whatman, UK), with a flow of 113 L/min through a Bendix 240 cyclone $PM_{2.5}$ inlet. The sampling durations were 3 h (08:00–11:00 and 15:00–18:00). Traffic counts, temperature, and relative humidity (RH) were monitored during each of the sampling period. Six pairs of aerosol samples were collected on pre-baked quartz-fiber filters and pair samples were taken average to represent one.

Three- to seven-rings PAHs in the roadside samples were analyzed using the in-injection port thermal desorption (TD)-GCMS method described in our previous papers (Ho and Yu, 2004; Ho *et al.*, 2008; Ho *et al.*, 2011). National Institute of Standards and Technology (NIST) SRM 2260A and SRM2266 were used as calibration standards for target PAHs. A filter cut of 3 cm^2 was used for the roadside samples. The analysis details have been described in our previous papers (Ho and Yu, 2004; Ho *et al.*, 2008; Ho *et al.*, 2011). Briefly, portions of quartz-fiber filter samples were cut into smaller portions inserted into the TD tube, which was custom-fabricated to be the same dimensions as the Agilent 6890 GC (Santa Clara, CA, USA) split/splitless injector liner. The temperature of the injector port was lowered to 50°C before analysis. The injector port temperature was then raised to 275°C for desorption in a splitless mode while the GC oven temperature was kept at 30°C. After the injector temperature reached 275°C, the GC analysis began. The GC oven program was initially held at 30°C for 2 min, increased to 120°C at 10 °C/min, then to 310°C at 8 °C/min, and finally held at 310°C for 20 min. An HP-5 ms capillary column (5% diphenyl/95% dimethylsiloxane, 30 m × 0.25 mm × 0.25 μm, J&W Scientific, Folsom, CA, USA) was used. The MS was operated in scan mode from 50 to 550 amu. Identification was achieved by comparing the mass spectra and retention times of the chromatographic peaks with those of authentic standards. Quantification was based on peak area ratios of select fragment ions characteristic of individual NPOCs to those of internal standards (Ho *et al.*, 2008; Ho *et al.*, 2011).

RESULTS AND DISCUSSION

Comparison of Online and Offline p-PAHs

Concentrations

On-line concentrations of the total p-PAHs measured with the PAS 2000 were obtained, based on the photo-electric signals emitted by real-world p-PAHs that coated on the surface of carbon aerosols. The electric signal is characteristics of site-specific and time-dependent as different locations are characterized by a different mix of PAHs sources. Source-specific calibration curves can be generated where the monitor output is compared to an analytically determined p-PAHs concentration. Previous studies (Chetwittayachan *et al.*, 2002b) have revealed a good correlation [determination coefficient (R^2) ≥ 0.82] for a wide variety of sources, including oil burner exhaust, parking garage air, cigarette smoke, burners, and urban aerosols, indicating that it is possible to calibrate the PAS signal to analytically determine p-PAHs concentrations in different environments. Unfortunately, as only 11–15 p-PAHs were determined in the previous studies, a large number of specific p-PAHs were uncounted. Few studies have also demonstrated the correlations of the PAS signal with specific p-PAHs, leading to have a better understanding on how the PAS signal responds to a variety of p-PAHs.

Fig. 2 shows the correlations between the three-hour average PAS signals and the sum of 16 U.S.EPA Priority p-PAHs and 34 most abundance p-PAHs, respectively, quantified on the filter samples. Good linear regressions (R^2 = 0.76 and 0.77) were demonstrated in the online and offline comparisons. In a certain degree, a slightly better correlation was obtained with more p-PAHs species taken into account from the offline method, showing the capability of photo-ionized of the PAS is unconfined for the 16 Priority p-PAHs.

The correlations of continuous PAS signals and individual PAHs concentration determined by the offline approach were examined (Table 1). Good agreements were found for eight PAHs with R^2 > 0.70. This suggests that the capability of photo-ionized of the PAS is sensitive enough for the eight PAHs including acenaphthylene, phenanthrene, fluoranthene, pyrene, perylene, 9-methylanthracene, methylfluoranthene, and cyclopentapyrene. Pyrene, fluoranthene, phenanthrene, chrysene, and 9-methylanthracene are major p-PAHs in the roadside environment, accounting for ~68% of the total quantified 34 p-PAHs. Except for chrysene, the PAS offers good correlations for the top five most abundant p-PAHs, validating the monitoring of the PAS in this environment at a certain degree. However, long-term comparison of PAS signals and individual PAHs concentration should be performed in order to make a solid conclusion.

After correction with online measurements, the hourly p-PAHs concentrations measured by the PAS ranged from 0.2 to 22.0 ng/m^3 at the PU roadside station from 15 January to 20 September 2005, with an average and median values of 5.3 and 4.6 ng/m^3, respectively. The average concentrations of p-PAHs were consistent in summer (5.9 ± 3.7 ng/m^3), spring (5.7 ± 3.9 ng/m^3), and winter (5.0 ± 3.6 ng/m^3). It is noted that the raw data (uncorrected data) by the p-PAHs will be used in the rest parts of this paper due to small data size used for the correlation equation, unless declaration of the corrected data.
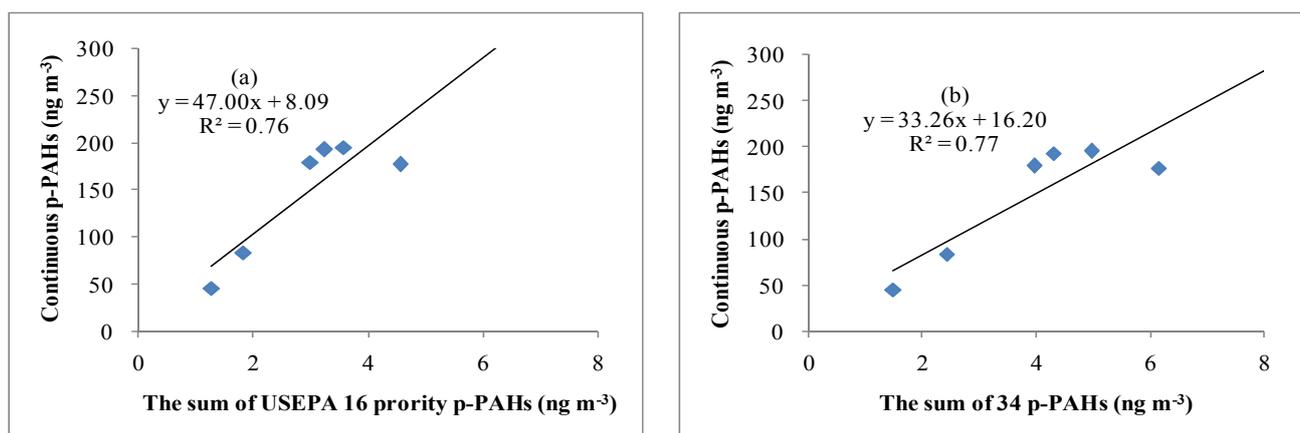
Fig. 2. Correlation between the three-hour average p-PAHs and analytically determined total p-PAHs concentrations that was calculated based on (a) the USEPA 16 Priority PAHs and (b) the sum of total measured 34 p-PAHs, respectively.

Table 1. Slopes, intercepts, and correlation coefficients of all regression equations between continuous p-PAHs and individual species.

Compound	Formula	Molecular weight	Slope	Intercept	R^2
acenaphthylene	$C_{12}H_8$	152.2	2823.1	20.4	**0.81**
acenaphthene	$C_{12}H_{10}$	154.2	6138.2	86.6	0.07
fluorene	$C_{13}H_{10}$	166.2	1546	94.1	0.29
phenanthrene	$C_{14}H_{10}$	178.2	382.9	17.4	**0.81**
anthracene	$C_{14}H_{10}$	178.2	1082.4	−70.9	0.59
fluoranthene	$C_{16}H_{10}$	202.3	182.9	12.8	**0.78**
pyrene	$C_{16}H_{10}$	202.3	130.8	14.2	**0.86**
benzo[a]anthracene	$C_{18}H_{12}$	228.3	1732.9	1.9	0.38
chrysene	$C_{18}H_{12}$	228.3	428.2	−9.3	0.58
benzo[b+k]fluoranthene	$C_{20}H_{12}$	252.3	381.5	114.8	0.16
benzo[a]fluoranthene	$C_{20}H_{12}$	252.3	1696.2	78.7	0.39
benzo[e]pyrene	$C_{20}H_{12}$	252.3	615.5	92.9	0.44
benzo[a]pyrene	$C_{20}H_{12}$	252.3	1922.2	51.8	0.48
perylene	$C_{20}H_{12}$	252.3	3710.1	28.3	**0.90**
2,6-dimethylnaphthalene	$C_{10}H_6(CH_3)_2$	156.2	971.8	108.4	0.6
9-fluorenone	$C_{13}H_8O$	180.2	757	26.2	0.64
9-methylanthracene	$C_{15}H_{12}$	192.3	452.9	50.9	**0.74**
anthraquinone	$C_{14}H_8O_2$	208.2	571.3	46.2	0.49
methylfluoranthene	$C_{17}H_{12}$	216.3	1314.6	36.1	**0.86**
retene	$C_{18}H_{18}$	234.3	1300.5	89.1	0.34
cyclopenta[cd]pyrene	$C_{18}H_{10}$	226.3	8059.6	−29.5	**0.76**
methylchrysene	$C_{19}H_{14}$	242.3	−1911.5	196.3	0.39
benzonaphthiophene	$C_{16}H_{10}S$	234.3	1434.8	63.1	0.37
Total USEPA 16 Prority PAHs			47	8.1	**0.76**
Sum of total 34 p-PAHs			33.3	16.2	**0.77**

Note: species below detection limit were not shown.

Diurnal Variation of p-PAHs

The average weekly cycles of p-PAHs (Fig. 3) suggest that p-PAHs concentrations are proportional to the hourly statistic of diesel vehicles, with an $R^2 = 0.94$. The diurnal variations of p-PAHs were approximately similar for every sampling day. As seen in Fig. 3, the concentrations of p-PAHs had obvious diurnal variations, with high values in daytime (9:00–17:00) and low values at mid-night (1:00–5:00). One dominant peak for p-PAHs occurred at 9:00–12:00, and one minor peak occasionally occurred at 15:00–

17:00. During weekdays, the time period between 9:00 to 12:00 was recorded as the maximum level of diesel-fueled vehicle (~3000 vehicles per hour) in a day, accounting for > 40% of total on-road traffic counts during this period. The lowest concentrations were measured at mid-night (1:00–5:00) when a minimum number of diesel vehicles (< 500 vehicles per hour) in a day was recorded.

The correlations between the p-PAHs concentrations and traffic counts for different classes of vehicles were examined by calculating the R^2 from the hourly data sets, as shown in
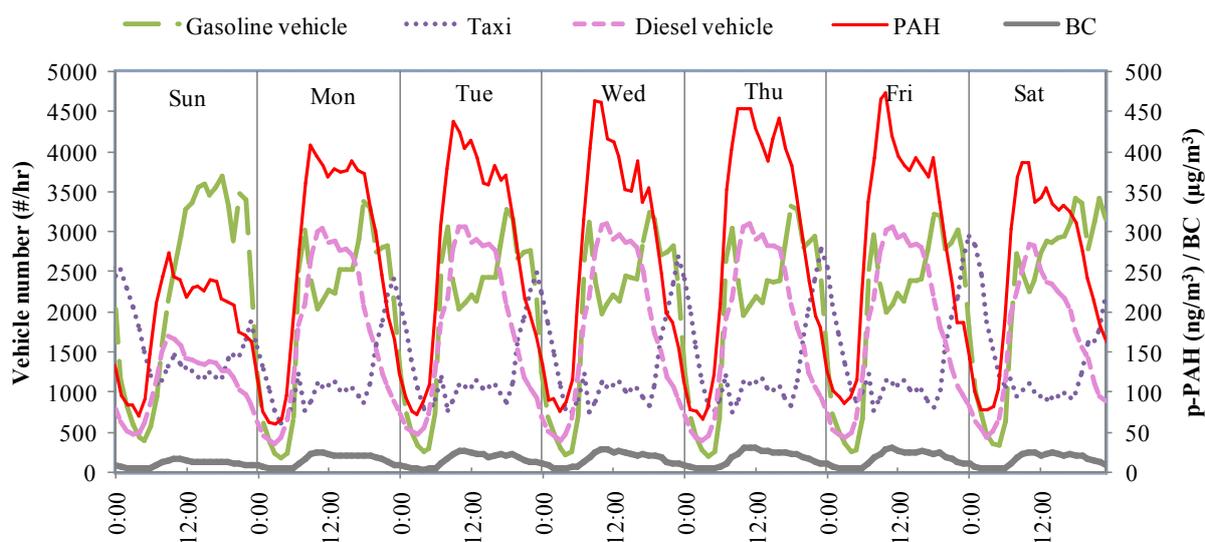
Fig. 3. weekly cycles of p-PAHs, BC, and traffic numbers.

Table 2. The best correlations were found for p-PAHs with diesel vehicles, followed by gasoline vehicles. The exhausts from LPG-fueled taxis contribute very little to particles (Cheng *et al.*, 2010), thus the poorest correlation was observed. Except for light buses, all categories of diesel-fueled vehicles including big buses and all goods vehicles had positive correlations with the p-PAHs concentrations. The diurnal variation of light buses was reverse from other classes of vehicles, having a higher traffic counts in nighttime but lower in daytime. Moto cycle fueled with gasoline also contributed to the emission of p-PAHs.

Average p-PAHs concentrations (raw data) along the whole sampling period are summarized in Table 3, focusing on the 24-h average (from 00:00–23:59), daytime (8-hr average from 09:00–17:00), and nighttime (4-hr average from 01:00–05:00) concentrations. The average concentrations over the period of observation in the present study were higher than that of Tokyo, Japan (Chetwittayachan *et al.*, 2002b), Bangkok, Thailand (Chetwittayachan *et al.*, 2002b), Bastel, Switzerland (Junker *et al.*, 2000), and Aosta, Italy (Agnesod *et al.*, 1996), which is due to a high diesel vehicle traffic volume on the road in Hong Kong. Black carbon (BC), the most abundant fraction in roadside fine particles, was also associated with diesel-fueled vehicles (Gertler *et al.*, 2001; Cheng *et al.*, 2010). The average daytime concentration of p-PAHs was *ca.* 4–5 times of the nighttime value, which is similar with the day/night ratio of BC. This indicates that both p-PAHs and BC are affected by the similar emission source of diesel vehicles.

Day-to-Day Variation of p-PAHs

As shown in Fig. 4, the daily p-PAHs concentrations fluctuated significantly from day to day throughout the entire sampling period, which is potentially due to the daily traffic volume variation (especially diesel vehicles) on the road. Clear weekly cycles of higher values on normal working days and low value on Sunday was obtained, relating to a higher traffic number and a smaller traffic number (especially less diesel-fueled vehicles), respectively. Consistent patterns between p-PAHs and BC also supports this observation because both arise from the combustion source. Diesel fuel combustion from vehicles has been reported as the major contributor to the p-PAHs and BC in

Table 2. Correlation coefficients (R^2) of p-PAHs and vehicle numbers from the weekly average.

Vehicle categories	p-PAHs	BC
Private car	0.58	0.60
Private/Public light bus	−0.68	−0.64
Moto cycle	**0.82**	**0.75**
Private/Public single deck bus	**0.81**	**0.67**
Private/Public double deck bus	**0.84**	**0.78**
Goods vehicle (≤ 5.5 ton)	**0.92**	**0.91**
Goods vehicle (5.5–24 ton)	**0.90**	**0.83**
Goods vehicle (> 24 ton)	**0.92**	**0.88**
Gasoline vehicle	0.63	0.64
Taxi (liquefied petroleum gas)	−0.49	−0.36
Diesel vehicle	**0.97**	**0.93**

Table 3. Statistic summary of p-PAHs concentrations over the periods of measurements.

	Period	24 h (00:00–23:59)	8 h (09:00–17:00)	4 h (01:00–05:00)	Day/night ratio
p-PAHs (ng/m^3)	Whole period	260.0	273.4	82.0	4.6
	Weekday	277.2	405.2	81.0	5.0
	Weekend	217.0	294.0	124.6	2.4
BC ($\mu g/m^3$)	Whole period	15.7	23.3	5.7	4.1
	Weekday	16.6	24.9	5.7	4.4
	Weekend	13.3	19.1	5.7	3.4
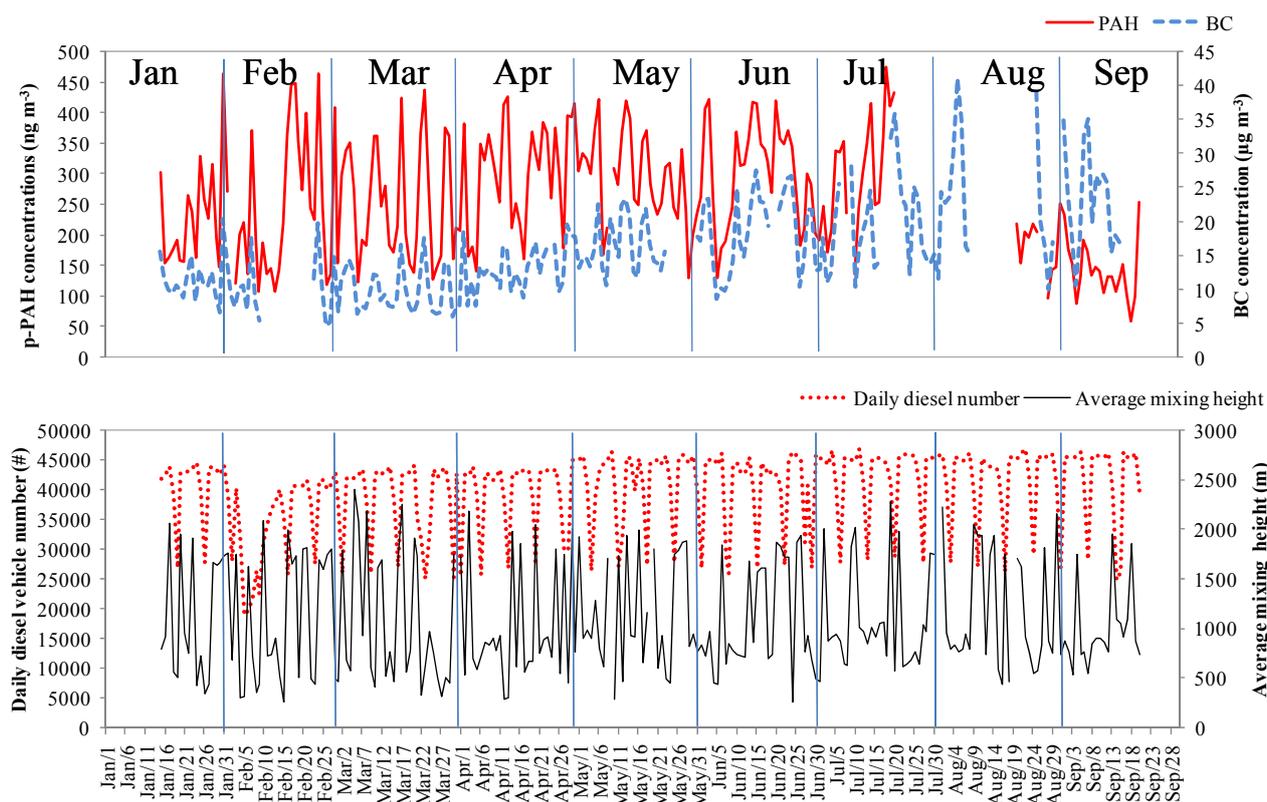
Fig. 4. Temporal variations of p-PAHs and BC.

a typical roadside environment (Gertler *et al.*, 2001; Ho *et al.*, 2002; Ho *et al.*, 2009; Cheng *et al.*, 2010). As an indicator of atmospheric stability, the mixing height was the other factor influencing the concentrations of p-PAHs. Good dispersion by high mixing heights often reduces p-PAHs concentrations and vice versa (Fig. 4).

The scattering plot of daily p-PAHs versus BC was examined in order to further investigate potential sources for p-PAHs and BC. In Fig. 5, the data can be grouped into three different sampling periods: group 1 from January to April; group 2 from May to July; and group 3 from August to September, in 2005. The best correlation ($R^2 = 0.80$) and lowest intercept (close to 1) were found for group 1,

suggesting the behavior of p-PAHs and BC follow each other in winter (January to April), confirming they have the same source of vehicle emissions. In contrast, moderate correlations and high intercepts were found for group 2 and 3, indicating that other sources have contributions to either pollutant. Ferry or container ship emissions had been previously reported to enhance the downwind roadside BC concentrations in summer (Lee *et al.*, 2006). In addition, the p-PAHs concentrations in group 3 were much lower than those in group 1 and 2. It could not be well-explained with current available data. Further work (e.g., conducting a long-term monitoring) is needed to clarify the potential sources or obtain a better understand of the emissions.

CONCLUSIONS

The present study continuously measured p-PAHs concentrations by the PAS at the typical roadside station in Hong Kong for nine months from January to September 2005. The data was validated with the traditional offline sampling and analytical method. The diurnal profiles of the p-PAHs were consistent throughout the sampling periods. The p-PAHs concentrations were the highest at 09:00–11:00 in working days (Monday through Saturday) when the number of diesel vehicles was at a maximum level of 3179 per hour, suggesting that people living around the location are exposed to a high number of diesel combustion particles enriched with p-PAHs coatings. In addition, the p-PAHs concentrations were found to depend upon the mixing height on a daily basis.

Fig. 5. Scattering plot of p-PAHs and BC.

ACKNOWLEDGMENTS

DISCLAIMER

Reference to any companies or specific commercial products does not constitute an endorsement, recommendation or favouring by the Company.

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