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RESEARCH ARTICLE

Large spatio-temporal variations of size-resolved particulate matter and volatile organic compounds in urban area with heavy traffic

Nguyen Hong Phuc[1] · Nguyen Thi Kim Oanh[1]

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Abstract

A monitoring campaign, the first of this kind for a heavy traffic urban area of Vietnam, was conducted which generated nearly 200 daily filter samples of $PM_{2.5}$, PM_{10}, and black carbon (BC), 1300 online hourly PMx (PM_{10}, $PM_{2.5}$, and PM_1), 900 hourly/bi-hourly BTEX data, 700 h of traffic counts, and online meteorology records. PMx and BTEX levels show large horizontal gradients across this small urban area of 300 m width suggesting that the pollution data should be generated with sufficient spatial resolutions for assessment of the exposure and health effects. This paper focuses on analyzing PMx with reference to the previously published BTEX to provide a more complete picture of the traffic-related pollution in the area. Spatio-temporal variations of pollutants are analyzed in relation to traffic flows and fleet compositions, weekday-weekend effects, local and regional meteorology. PM_{10} and BTEX levels had larger variations between the sites indicating their stronger associations with the traffic activities than the finer particles. Twenty-four-hour (24 h) $PM_{2.5}$ levels ranged between 19 and 191 μg/m^3 with high $PM_1/PM_{2.5}$ ratios of above 0.8 at ambient site (AA) and above 0.7 at roadsides. Multivariate relationship analysis (PCA) for the bi-hourly datasets of meteorology, traffic flows, and pollutant levels indicated overwhelming influence of on-road traffic fleet compositions on the roadside pollutants levels. At AA, PCA results showed a complex interaction between local emissions, meteorological conditions, and regional/long-range transport. Higher pollution levels were associated with the airmass types having the continental origin and pathways.

Keywords Roadside · Ambient air · Pollutant variations · Traffic fleet composition · Hanoi

Introduction

High levels of air pollution observed in the developing countries are of pressing health concern (Hopke et al. 2008; Tai et al. 2020; UNEP 2019). Scientific evidence indicates that exposure to air pollution can result in a wide range of acute and chronic health effects. Worldwide, in 2019, there were 6.67 million premature deaths attributed to air pollution, with 4.14 million due to ambient fine particulate matter or $PM_{2.5}$ (particles with aerodynamic diameter ≤ 2.5 μm), 365,000 due to ozone, and 2.31 million due to household pollution (HEI 2020). Over 90% of the premature deaths associated with air pollution occurred in low- and middle-income countries with the highest number of deaths found in the Western Pacific and South East Asian regions (WHO 2014).

A steady growth of vehicle population in developing Asian cities induces heavy traffic congestions that disproportionately increase emissions of toxic pollutants. Accordingly, the large on-road vehicle fleets are reported to be the leading causes of ambient air pollution in big cities (Huy et al. 2020a; Kim Oanh et al. 2018; Phuc and Kim Oanh 2018). For example, in Vietnam, during the period of 2005 and 2015, the number of total in-use vehicles had increased by almost 3.5 times (OICA 2016). High growth rates over the same period were also reported for other South-East Asian countries, i.e., by 2.5, 1.8, and 1.6 times in Indonesia, Malaysia, and Thailand, respectively (OICA 2016). Although there is a wide array of urban transport modes, the dominance of private cars and motorcycles (MC) is still common in many Asian countries (UN-ESCAP 2015). In Hanoi, the capital of Vietnam, MC shared

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✉ Nguyen Thi Kim Oanh
kimoanh@ait.ac.th

[1] Environmental Engineering and Management, School of Environment, Resources and Development, Asian Institute of Technology, Pathumthani, Thailand

between 70%- and 90% in the on-road traffic fleet, followed by cars with 4%-–15% (ADB 2015). In a busy urban road in Bangkok, Thailand, the modal shares were 18–25% for MC and 30–33% for private cars (Kim Oanh et al. 2013). A slow expansion of the road area coupled with a rapid growth in vehicle population has resulted in frequent traffic jams, especially during rush hours that lead to high levels of air pollution along heavily traveled roads (Giang and Kim Oanh 2014; Truc and Kim Oanh 2007). Furthermore, a large number of old and poorly maintained vehicles are still in circulation which further increases the emission rates to worsen air quality (Trang et al. 2015). Exposure to the high air pollution levels results in serious adverse health effects which have been reported in many epidemiology and toxicology studies (Cohen et al. 2017; Ha Chi and Kim Oanh 2021).

Hanoi capital city is located within the Hanoi Metropolitan Region (HMR) which had a population of over 8 million and an average population density of 2410 people/km^2 as of 2019 (GSO 2019). Similar to many other cities in Asian developing countries, Hanoi is facing with serious air pollution problems during the last two decades as a consequence of rapid urbanization and massive increase of vehicular population, notably a fast growth of private vehicles (MONRE 2014). By the end of 2018, the accumulated registered MC fleet in HMR was 5.7 million, i.e., a 1.25-fold higher than 2015, while the private car fleet was 362,000 that is 1.64-fold higher than 2015 (VR 2017). The drastic increase of the vehicle fleets by far surpassed the handling capacity of the road network. Moreover, the space limitation of the inner city areas could accommodate only narrow roads hence inducing more traffic congestions during rush hours (ADB 2011). The large MC fleet, not properly equipped with exhaust control devices (Kim Oanh et al. 2012; Tung et al. 2011), moving in streets with heavy traffic jams is a major cause of the high levels of traffic-related pollutants, including $PM_{2.5}$ and the volatile organic compounds of benzene, toluene, ethylbenzene, and xylenes (BTEX) measured at roadsides (Hai and Kim Oanh 2013; Truc and Kim Oanh 2007). Children, elderly, and the poor living in the heavy traffic areas would be the most affected by the traffic-related air pollution in Hanoi (Hung 2010; Luong et al. 2018). As a way of example, Nhung et al. (2019) analyzed the 2007–2016 data for Hanoi and revealed a positive, stable, and solid association between the increased daily ambient levels of PM_{10} (particles with aerodynamic diameters ≤ 10 μm) and the length of hospital stay of children aged 0–5 years due to lower-respiratory infection.

This paper presents the results of a monitoring campaign for traffic-related pollutants in a congested traffic area in Hanoi during dry months. The key pollutants included daily levels of $PM_{2.5}$ and black carbon (BC), measured by 24 h Minivol filter-based sampling, and hourly levels of PMx: PM_{10}, $PM_{2.5}$, and PM_1 (particles with aerodynamic diameters ≤ 1.0, μm), monitored by real-time GRIMM recording. The BTEX levels, previously reported for the sites (Phuc and Kim Oanh 2018), were also referred to in the PM data analysis to show a more complete pollution picture in the area. This study is a part of our efforts to assess the effects of on-road traffic to the air pollution in Hanoi to provide scientific information to further quantify the change in health risks to local people in the various scenarios of the modal shift to the public transport.

Methodology

Hanoi is influenced by the tropical monsoon climate typical for Northern Vietnam with two main seasons, i.e., winter (November to March) and summer (May to September), and two transitional periods in between: spring (April) and autumn (October) (Ngu and Hieu 2004). In winter, high-pressure ridges are frequently observed extending from the central China to Northern Vietnam that bring in cold weather spells with associated low mixing heights and calm wind, typically for areas under influence of stable high pressure systems. When prevalent, these conditions enhance accumulation of ambient air pollution to considerably high levels during winters in the city (Hai and Kim Oanh 2013; Hien et al. 2011; Kim Oanh et al. 2006).

We conducted monitoring for air pollutants, traffic flow, and meteorology at two representative roads and one ambient air (AA) site. The monitoring periods included a dry season period of 13 Jan–9 Feb (winter with stagnant air, dry weather and less wet removal) and a transitional autumn period of 13 Oct–9 Nov (also in dry season but with more rainfall) in 2015. The monitoring aimed to capture both the day-to-day fluctuations of 24 h PMx and BC, as well as the diurnal variations of PMx. During the winter period, totally at the 3 sites, 79 samples of $PM_{2.5}$ were collected by Minivol for mass and BC measurements, 517 hourly PMx data (averaged from 5-min measurements) and 494 h of traffic video records. In the transitional period, the corresponding numbers were 68, 765, and 193, respectively, with an additional 47 samples of PM_{10} collected by Minivol, as detailed in Table S1, Supplementary Information (SI). The pollutant data were statistically analyzed to reveal the relationship between traffic volume and the pollution levels at the sites. The association between the daily PM and BC levels with airmass types arriving at AA was investigated using the airmass backward trajectories to identify the potential of the regional air pollution transport.

Sampling sites

This study aimed to comprehend the spatio-temporal variations of the pollutant levels within a small area, i.e., with the maximum distance of 300 m between the monitoring sites, in a heavy traffic zone of Hanoi. The monitoring area was located along the congested Truong Chinh (TC) road which crosses the populated Dong Da and Thanh Xuan urban districts (Fig. S1, SI). Two roads were selected to represent typical traffic conditions in the city: TC road was an inner ring road with heavy traffic and Nguyen Ngoc Nai (NN) was a small and less traveled residential street. Each road had two traffic lanes (inbound and outbound) and both had an east–west orientation, with the road angle of 107° to the north–south line (Fig. 1). This orientation induced one upwind and one downwind side of the road when either the north-east monsoon (winter) or south-west monsoon is prevalent in the city.

With the aim to represent the normal traffic flows, the monitored sections of the roads were selected to be away from any road crossing or bus stations. The sampling segment of the residential road of NN (8 m wide) had both sides bordered by residential buildings with the average height of 9 m, hence inducing a street canyon configuration. TC (15 m wide) had a semi-street canyon configuration with an open space looking toward the AA site. Roadside monitoring was done with the equipment placed at 2.0 m above the ground level and 1–1.5 m away from the traffic lane (Fig. S1, SI). The AA site (21° 0′ 0.51″ N; 105° 49′ 33.0″ E) was surrounded by mixed institutional and residential areas, and is located in between the two roads, about 150 m away from each road. The monitors at AA were placed on the rooftop of an office building, at 12 m above the ground. To ensure the data quality, the QA/QC was carefully implemented in both sampling and chemical analysis stages.

Air pollution monitoring

Filter-based PM and BC monitoring

The daily (24 h) PM samples, starting at around 6:00 a.m. each day, were collected on glass fiber filters (Advantec GB-140 47 mm, retention 0.4 µm) using the Minivol Portable Air Samplers (Model 5.0) of Airmetrics (http://airmetrics.com//minivol). Two Minivol samplers were placed at two opposite sides of each road for simultaneous sampling. During the winter period, only $PM_{2.5}$ samples were collected while during the transitional period, both PM_{10} and $PM_{2.5}$ were collected. The sampling was done on every monitoring day at AA, simultaneously with the sampling at either TC or NN road. The Minivol samplers were calibrated at the sites to ensure the designated flow rate of 5L/min. The Airmetric flow rate calibration kit (http://www.airmetrics.com/products/calibration.html) was used which included a digital manometer (Serries 447AV by Dwyer Instrument), one calibration orifice with a louvered inlet, and 1-m transparent Polyethene tube. After sampling, each sampled filter was placed in a separate petri dish, wrapped with aluminum foil, and then put in an airtight bag. The bags containing samples were refrigerated at 4 °C during the transportation (using an icy box) and storage. The pre- and post-weight of every filter were determined, repeated 3 times, by a 7-digit Analytical Balance of Mettler Toledo following the procedure described in the United States Enviornmental Protection Agency (USEPA) (1998). Before weighing, the filters were desiccated for 24 h at temperature of 22 ± 2 °C and relative humidity of 40 ± 5%. All the mass concentrations in this paper are reported at the standard conditions of 25 °C and 1 atm.

BC loaded on the $PM_{2.5}$ sampled filters was measured using an optical transmissometer (Sootscan Model OT21 Optical Tranmissonmeter) of Magee Scientific (https://mageesci.com/mproducts/sootscan-model-ot21-optical-transmissometer). The device measures the attenuation of transmitted light through the aerosol layer deposited on the filter, simultaneously at 2 wavelengths IR (880 nm) and UV (370 nm). The light absorption at 880 nm was interpreted as BC while that at 370 nm was interpreted as UVPM. The latter may be interpreted as brown carbon

Fig. 1 The sampling area (satellite view) and sketch showing the sampling sites in a busy urban area of Hanoi (detailed in Fig. S1, SI)

and is thought to be able to relate to the presence of aromatic organic compounds (Jeong et al. 2004). The limit of detection is 0.075 μg/m^3 and the limit of quantification is 3.1 μg/m^3 for the samples collected on a 47 mm diameter filter at 16.7 L/min over 24 h (Magee Scientific 2021). In our study, due to the high pollution load, BC and UVPM were detected in all the sampled filters. The OT21 measurements were done 3 times for each filter and the average was used to calculate mass of BC and UVPM, respectively, using the corresponding attenuation coefficient and the PM collection area.

Online PM measurement with GRIMM

Real-time measurements of PM_x at a 5-min interval were done using two GRIMM Aerosol Technik devices (GRIMM Model 1.107 spectrometer) which measure the particle numbers based on the light scattering principle. The number concentrations are converted to the mass concentration using the universal particle density of 1.67 g/cm^3 (Baldwin et al. 2015). We used the GRIMM Model 1.107 devices with non-heated inlets which would minimize the loss of the volatile particulate components. At low relative humidity (RH), the GRIMM device operated without inlet heating has been reported to produce the total $PM_{2.5}$ mass comparable to a reference method of Rupprecht and Patashnick Co. (R&P) filter dynamic measurement system (FDMS). At high RH, however, the GRIMM results for $PM_{2.5}$ mass were slightly higher than FDMS which may be attributed to the increase in the particle volume associated with water-bound. To better quantify the PM mass, one GRIMM device (GRIMM#1) was collocated with a Minivol at AA, 11 days for PM_{10} and 20 days for $PM_{2.5}$. The collocated monitors showed good linear relationships of the mass results with $R^2 = 0.98$–0.99 (Fig. S2a, SI). We used the regression equations (see Fig. S2a, SI) to convert the measured GRIMM#1 mass concentrations to the equivalent Minivol concentrations and this would help account for the actual density of particles in the study area and the RH effects.

To harmonize the data produced by two devices (GRIMM#1 and GRIMM#2), they were collocated for 3 days. The results showed good linear relationships between the data series produced by the two devices with $R^2 = 0.95$ for PM_{10} and higher ($R^2 = 0.99$–0.998) for the finer PM fractions of $PM_{2.5}$ and PM_1, respectively (see detail in Fig. S2b, SI). The obtained regression equation between the readings of the two devices was used to synchronize the records for every size range. The measurements produced by GRIMM#2 were adjusted to the equivalent values of GRIMM#1 to produce consistent GRIMM data for comparison of PMx levels between the sites.

Traffic flow and meteorology

The video records of the traffic flows, taken by cameras simultaneously with the roadside air pollution sampling, were replayed for manual counting to get the traffic volume and composition. Totally, there were 687 h of the traffic flow data collected (Table S1, SI). To represent 1-h traffic data, we analyzed the 15-min record of the hour and multiplied the results by four (4). A portable meteorological station (Vantage Pro II, DAVIS Instrument) was deployed at AA to record, at 5-min interval, the meteorological variables (temperature, humidity, wind speed, wind direction, and barometric pressure) at 15 m above the ground in every sampling day. The processing of meteorological data has been detailed previously by Phuc and Kim Oanh (2018) which included the data screening to remove erroneous data points, and calculation of hourly and daily average. To account for the circularity effects of wind direction, its hourly average (mean angle) was calculated from 5-min measurements using the so-called directional statistics (Mardia and Jupp 2000).

Data analysis

First, the 5-min data produced by GRIMM were used to calculate hourly and 24 h PMx concentrations which were then synchronized with the Minivol mass data using the linear regression equations (Fig. S2, SI). The hourly traffic flows were sorted into 4 major categories based on their fuel usage and type/size commonly used in Hanoi (Trang et al. 2015). These included the following: (1) MC&DTC (delivery tricycle with MC engine), all gasoline powered; (2) passenger car (PC: Sedan car and MPV/SUV: multi-purpose vehicle/sport-utility vehicle MPV/SUV with 7 seats and above) and taxi, above 93% gasoline powered with the restdiesel-powered; (3) Van&Pick (van with 12–16 seats and pick-up), 50% gasoline and 50% diesel-powered; and (4) diesel (all buses: inter-city bus and city public bus, and all trucks: light and medium truck with ≤ 6 wheels and heavy-duty truck with ≥ 10 wheels), all diesel-powered.

The multivariable relationship between air pollution concentrations, the vehicle flows of 4 categories, and meteorological conditions was evaluated by Principal Component Analysis (PCA) using the IBM Statistical Package of Social Science (SPSS 2010). The data set for PCA of two roadsides included bi-hourly air pollution data of $PM_{2.5}$ and coarse fractions ($PM_{2.5-10} = PM_{10} - PM_{2.5}$) produced by GRIMM, the individual BTEX species, monitored following NIOSH (2003) previously presented in Phuc and Kim Oanh (2018) with the detail on the monitoring method and QA/QC given in Textbox 1 (SI), and meteorological variables of temperature (Temp), relative humidity (RH), barometric pressure (P), and wind speed (WS). For the AA site, the wind direction was also included which was converted to the wind direction index (*WDI*) to overcome the discontinuity in the wind direction scale, i.e., $WDI = 1 + \sin(\theta + \pi/4)$, where θ is the wind direction angle. The traffic data used in the AA dataset was the sum of the traffic flows in both roads. Note that TC had overwhelmingly larger traffic flows (Fig. 2); hence, the days without traffic counting data in TC were not included in the PCA dataset for AA. The consecutive hourly data series of traffic flows, pollutants (PMx and BTEX), and meteorology were used to produce the corresponding bi-hourly data series for PCA.

The final dataset for PCA consisted of 14 variables for each TC and NN road, and 15 variables for AA that yielded a total of 1,100–1,260 data points. The roadside Minivol sampling for PM and BC, as well as for BTEX, was done simultaneously at two opposite sides of each road to produce a pair of samples in each monitoring day. We used the average of every pair in the data analysis, including PCA, to account for the difference in pollution levels between windward and leeward sides of a road with street canyon configurations (Kim Oanh et al. 2008).

The potential contribution of regional pollution transport to the area was analyzed using the backward trajectories, considering both origins and pathway of airmass arriving at the ambient site (AA) in every monitoring day. The HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) 3-day backward trajectories (https://www.scribd.com/document) were constructed using the final analyzed meteorological data (FNL) from Global Data Assimilation System (GDAS), starting at 1,000 m above the AA site coordinates and at 00:00 UTC or 07:00 at local time each day. The pollution levels observed within different airmass types were comparably analyzed. The 3-day backward trajectories were used previously for BTEX (Phuc and Kim Oanh 2018) to account for the relatively short atmospheric life of the species (only benzene has about 9 days, other species < 2 days). Therefore, this study also used the 3-day trajectories to analyze both PMx and BTEX data together.

Results and discussion

Traffic volume, fleet composition, and diurnal variations

The diurnal variations of hourly traffic counts and shares of the 4 categories, averaged for weekdays and

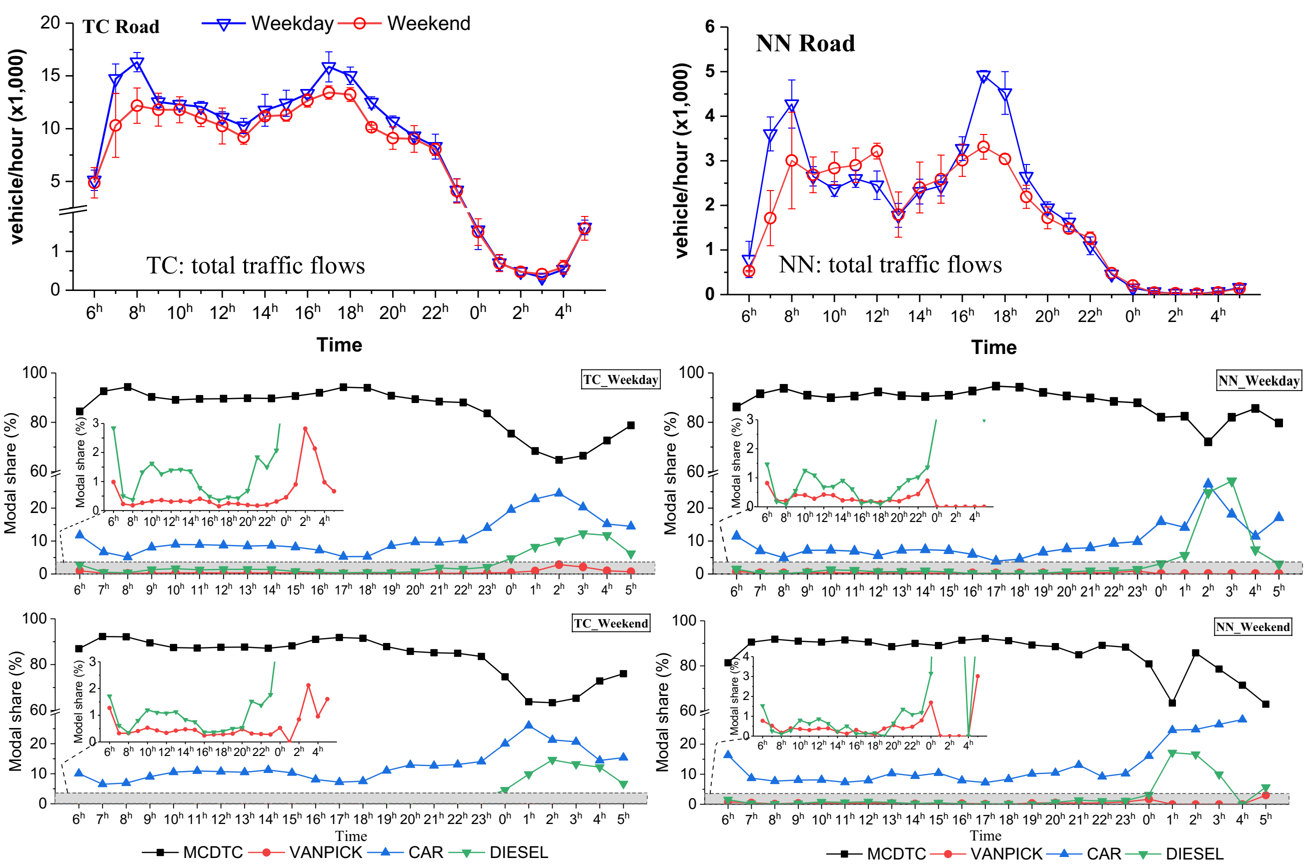

Fig. 2 Diurnal variations of total traffic flows and traffic composition in weekday and weekend at TC and NN roads (inserted are scaled up graphs for diesel and VanPick categories)

weekends, respectively, in two roads are presented in Fig. 2. This study captured both daytime and nighttime traffic flows fully for 20 days, with 12 days in TC (8 weekdays and 4 days in weekend) and 8 days in NN (6 weekdays and 2 days in weekends), and additional rush-hour traffic records at both roads. The total traffic volume on the larger ring road TC was about 4 times higher than that in the residential road NN. For example, in the winter period, the hourly average traffic flow in TC was 8,971 ± 5,395 veh/h compared to 2,122 ± 1492 veh/h in NN. In each road, the hourly traffic volume was not statistically different between the two monitoring periods, i.e., with p value = 0.18 for TC and 0.23 for NN (Table S2). The p value is the statistical significance, which was taken to be 0.05 in this study, i.e., the probability of wrongly rejecting the tested hypotheses of ≤ 5% was considered acceptable.

In both roads, the hourly traffic flows on weekdays were higher than weekends, especially during the rush hours, morning 7:00–8:00 and afternoon 17:00–18:00, with larger relative differences found for NN. The difference between the weekday and weekend traffic flows was statistically significant at $p \leq 0.05$ at NN, while for TC, the p value was slightly higher, i.e., 0.06 (Table S2, SI). The traffic flows were significantly different between daytime and nighttime at both roads, i.e., at nighttime after 22:00, the traffic volume at both roads reduced substantially on both weekdays and weekends (Fig. 2).

The diurnal variations of the 4 vehicle categories generally show similar patterns of the fleet composition between the two roads, as well as between weekdays and weekends on each road. The period 6:00–23:00 had the on-road traffic fleet mainly composed of MC&DTC with a more than 90% modal share, followed by the PC&Taxi with around 8–10%, while the other two types collectively contributed less than 2%. Late evening and early morning hours saw a reduction in the MC&DTC share reaching a minimum of about 70% at around 01:00–02:00. The diesel fleet share increased in the evening, reaching a maximum of about 10% between 01:00 and 03:00 which was consistent with the regulation allowing medium and heavy-duty trucks to ply in the city only from 21:00 to 6:00 the next day. This is an important factor affecting the pollution levels at the monitoring sites discussed later.

Hourly levels and diurnal PMx variation in relation to traffic flow and composition

Hourly levels

The hourly levels of PMx, calculated from 5-min measurements by GRIMM after being adjusted against Minivol mass, showed much higher PM_{10} at the roadsides than AA, especially at busy road TC (Fig. 3). The residential road NN had comparable high PM_{10} levels with TC during winter (i.e., reaching a maximum above 400 μg/m^3) but significantly lower during the transitional period (maximum of 216 μg/m^3), most probably due to the more intensive rain during the monitoring days at NN (Fig. 4). The lowest hourly PM_{10} levels were observed at AA with the peaks of around 150 μg/m^3 in both periods. The ratio of PM_{10} morning peaks during the winter period between both roadsides and AA, respectively, was about 2.9 times, while that during the transitional period reduced to 1.3–1.8.

The concentrations of fine PMx, i.e., $PM_{2.5}$ and PM_1, were more uniformly distributed among the sites than PM_{10} (Fig. 3). The highest levels were still seen at TC with the hourly peaks during the winter reaching about 150 μg/m^3 and 120 μg/m^3 for $PM_{2.5}$ and PM_1, respectively, which were well above the corresponding values in the transitional period, i.e., 110 μg/m^3 and 90 μg/m^3, respectively. The second highest $PM_{2.5}$ levels in the winter period were at NN, with morning hourly peak reaching120 μg/m^3. However, the remaining peaks at NN, i.e., the PM_1 peak during the winter period (90 μg/m^3), and both $PM_{2.5}$ (94 μg/m^3) and PM_1 (74 μg/m^3) in the transitional period were lower than the corresponding values at AA. The levels at AA were quite similar between two periods, with the hourly $PM_{2.5}$ peak reaching around 100 μg/m^3 and PM_1 peak of around 90 μg/m^3. The ratio of the morning peaks between TC and AA for $PM_{2.5}$ and PM_1 was lower than that for PM_{10}, i.e., 1.1–1.4, while that between NN and AA was only 0.85–0.95, meaning AA actually had higher finer PMx than NN (Fig. 3).

PM_1 had high shares in $PM_{2.5}$ mass with AA having the $PM_1/PM_{2.5}$ ratio of 0.82–0.87, which was higher than the roadsides (0.71–0.75), as detailed in Table S3 (SI). The obtained $PM_1/PM_{2.5}$ ratio in this study was quite similar to those reported at traffic sites in other Asian cities (Lee et al. 2006; Zhang et al. 2018), but was relatively higher than that reported by Hien et al. (2021) of 0.60 ± 0.05 for a residential site in Hanoi. The difference may be explained by the fact that the ratio obtained in our study was for more polluted months in a heavy traffic area, whereas Hien et al. (2021) reported the average ratio over a longer period of 10 months (including also early summer) for a residential site in Hanoi.

Similar to the case of $PM_1/PM_{2.5}$, at AA, we obtained the $PM_{2.5}/PM_{10}$ ratio > 0.60, which is also higher than the roadsides (0.40–0.45). The higher fraction of coarse particles ($PM_{10\text{-}2.5}$) at roadside implied a substantial contribution from the road dust resuspension. For comparison, the ratios between simultaneously measured 24 h $PM_{2.5}$ and PM_{10} at each site are also given in Table S3 (SI) which showed the values in the same range as those calculated based on hourly GRIMM values. In fact, the ratios obtained at AA site were similar to the general

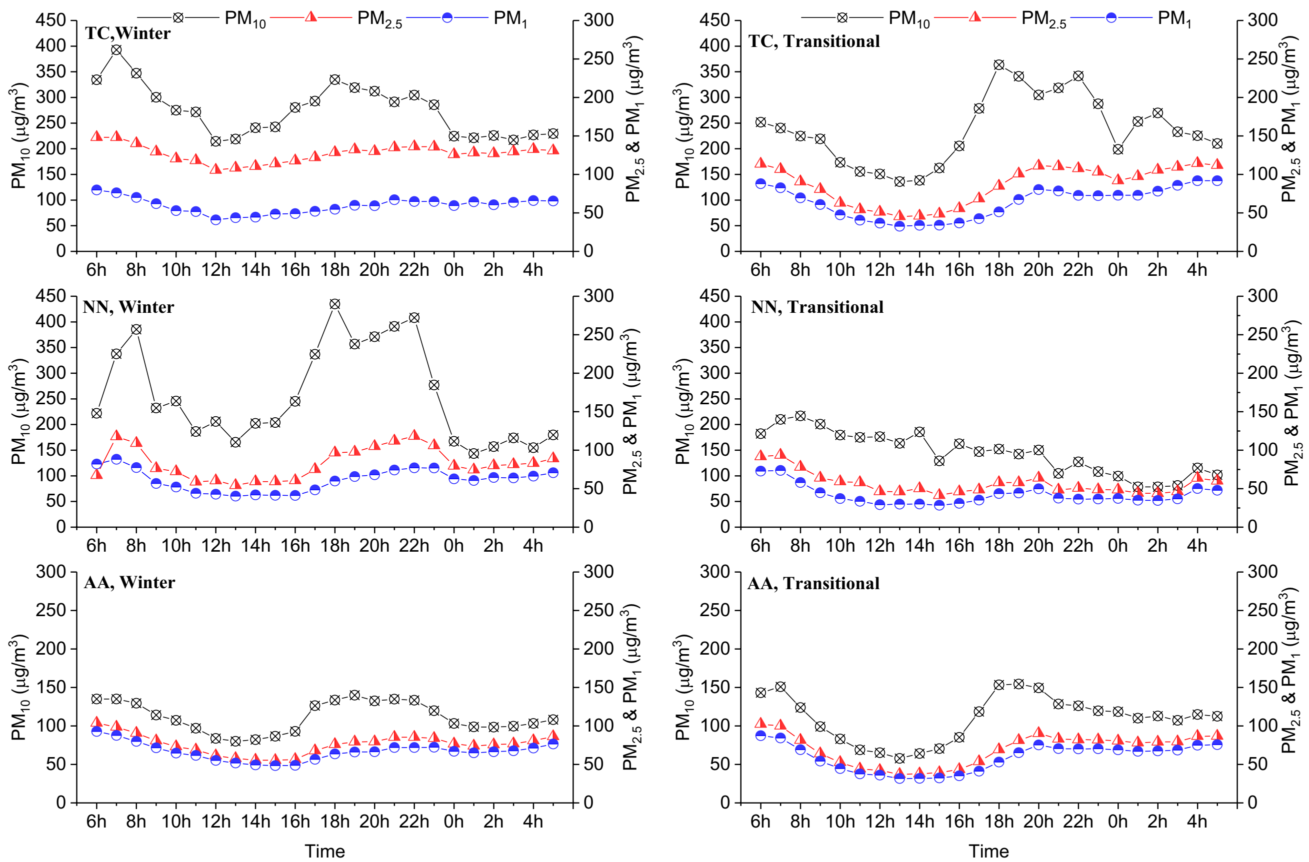

Fig. 3 Diurnal variations of hourly average of PM_x by GRIMM (note the difference in scale of Y1 between AA and roadsides)

values reported for six Asian cities (i.e., ≥ 0.6) (Kim Oanh et al. 2006). Overall, the higher $PM_{2.5}/PM_{10}$ ratios obtained for AA can be explained by its ambient location, not close to any traffic lane and a higher sampling intake (12 m above the ground). Larger fraction of coarse particles (smaller ratio) at roadsides may be attributed to the road dust resuspension and also other intensive residential activities such as those along NN. The linear regressions between hourly $PM_{2.5}$ and PM_{10} had low coefficients of determination (R^2), lower at roadsides with $R^2=0.31$–0.46 and higher at AA with $R^2=0.54$–0.70. The linear regressions between $PM_{2.5}$ and PM_1 had significantly higher R^2, 0.83–0.94 at roadsides and 0.98–0.99 at AA, which in turn suggested their similar fluctuation patterns that may be linked to the similarity in influencing sources/factors.

Diurnal variations

It is remarkably shown that both PM_1 and $PM_{2.5}$ had very similar diurnal variation patterns at every site. Between the 3 sites, these fine fractions also had more similar diurnal variation patterns than that of PM_{10} (Fig. 3). The diurnal patterns of all PMx reflected well the morning traffic rush hours with peaks appeared at 7:00–8:00. In the evening, however, the hourly fine PMx levels continued rising after 19:00 (traffic peak) and remained high with less pronounced peaks appearing about 3 h later (20:00–22:00) when the total traffic volume was minimum in both roads. Thuy et al. (2018) reported a similar diurnal variation of $PM_{2.5}$ obtained for an ambient site in a university campus and showed, for example, a clear morning rush hour peak appearing at about 7:00 a.m. while the high $PM_{2.5}$ levels remained from 19:00 onward. The evening peaks of PM_{10} obtained in our study in fact were more consistent with the rush hours at AA and TC with peaks appearing at about 17:00–18:00, than at the residential road NN. Other factors than the total traffic activities may also influence the PM_x diurnal variations, e.g., non-traffic activities and diurnal changes in meteorological conditions, e.g., lower mixing height at nights (Hien et al. 2002). In particular, the residential activities in the evening along the NN road, such as cooking in the morning and evening and street sweeping, would affect the diurnal variations. It is worth mentioning that cooking with the polluting honey coal briquettes was

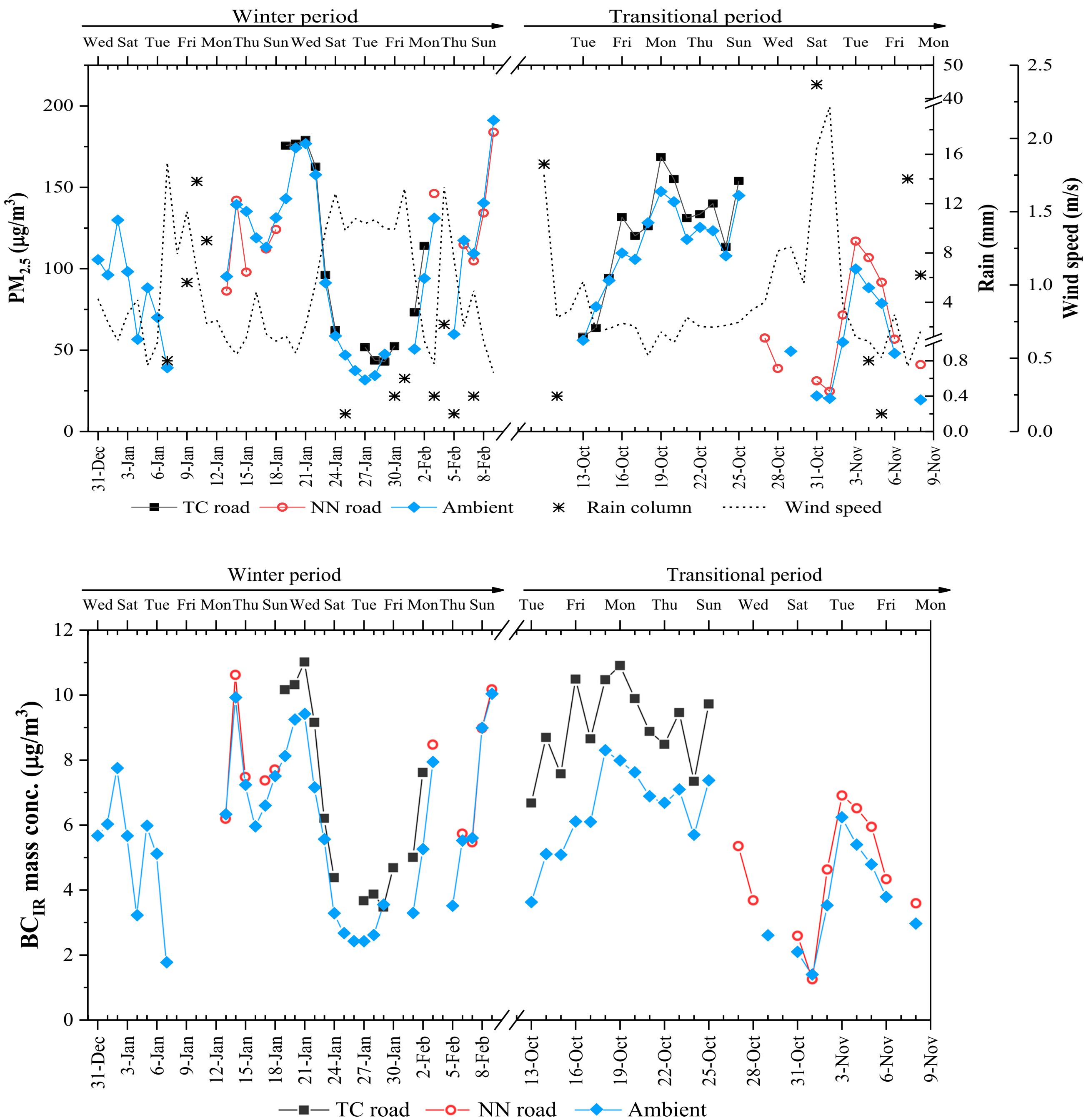

Fig. 4 Variation in daily levels of $PM_{2.5}$ and BC at the monitoring sites

still practiced in the study area during the monitoring time which may be a significant source of fine particles (Huy et al. 2020b).

An important factor to be considered was the change in the traffic composition during a day, with higher shares of heavy diesel trucks at nighttime after 21:00, when they were allowed plying the roads. The share reached the maximum at about 3:00 a.m. then reduced after 5:00 a.m. (Fig. 2). The movement of heavy trucks on the roads would induce more road dust resuspension which may be evident from another PM_{10} peak in TC road at 22:00. The exhaust of these diesel vehicles emits fine PMx with a high BC portion (Kim Oanh et al. 2009). Note that because the public buses in Hanoi generally stopped operation from 22:00 until 5:00, the high share of diesel-powered vehicles in the roads after 22:00 would be mainly from medium and heavy-duty trucks. Thus, even with a considerable reduction in the total traffic volume at nighttime, the fleet after 21:00 consisted of a higher number of heavy trucks that induced large amounts of PMx emissions, both directly from the exhausts with mainly fine particles and from the road dust resuspension with mainly coarse particles. Particularly, less trucks (and no buses all day) operated in NN could be a reason that the fine PMx levels at this road dropped faster than that at TC.

The diurnal variation patterns of the fine fractions ($PM_{2.5}$ and PM_1) in fact were quite similar to those reported for BTEX reported for the sites (Phuc and Kim Oanh 2018). BTEX also had prolonged evening hours with high levels, extending to 21:00–22:00, which was mainly due to the meteorological conditions. For PMx at the sites, the rush hour peaks are explained by the traffic activities, more clearly for PM_{10}, while the mid-day lows are associated with low traffic volume and elevated mixing height with stronger wind, i.e., reaching about 1.5–2.0 m/s (Phuc and Kim Oanh 2018). The high PMx levels during prolonged evening hours may be explained by the contributions of residential cooking emissions, and meteorological conditions with low wind at nighttime, i.e., speeds ≤ 0.5 m/s (Phuc and Kim Oanh 2018), and low mixing height (Hien et al. 2002). The reasons for high levels of fine particles during the midnight and early morning in Hanoi, especially at AA, still need further investigation, in particular, to get detailed information on diurnal variations of PMx chemical compositions.

Daily levels of PM and BC in relation to local meteorology

Levels of PM and BC

The 24 h PM Minivol sampling results showed high but fluctuating levels at 3 sites (Fig. 4). AA had the largest number of 24 h data points with 55 valid daily samples. TC and NN had 25 and 20 pairs of samples, respectively, with each pair representing the two simultaneously collected samples on both opposite sides, i.e., TC1 and TC2, and NN1 and NN2, as shown in Fig. 1 and Fig. S1 (SI). Each data point of the daily concentration of $PM_{2.5}$ and BC at the roadsides presented in Fig. 4 is the average of a corresponding pair. Overall, similar fluctuating patterns of the $PM_{2.5}$ and BC are seen between the 3 sites, but the BC levels at TC and NN roadsides were clearly higher than those at AA. The fluctuations in the daily BC levels in each site were mostly similar to those of $PM_{2.5}$. However, the relative difference in BC levels between the sites was larger than that of $PM_{2.5}$, i.e., TC had remarkable higher BC levels than AA, and this may be attributed to the larger diesel vehicles fleet, the main BC emitters, in this busy road.

Compared to the Vietnam national ambient air quality standard (NAAQS) for 24 h $PM_{2.5}$ (50 μg/m^3), the exceedance percentage was the largest in TC with 92% during both periods, higher than NN with 84% and AA with 77%. For 24 h PM_{10} (measured only during the transitional period), the percentage of exceedance of the NAAQS (150 μg/m^3) was 100% at TC, 40% at NN, and 46% at AA. Exposure to these high levels of PM suggests potential health effects.

The period average of daily pollution levels of $PM_{2.5}$, BC, and UVPM measured at AA was not statistically different between the two monitoring periods, i.e., *p* values: 0.24–0.38 (Table 1). Considering the combined data of both periods, the range of $PM_{2.5}$, BC, and UVPM, respectively, at 3 sites was largely overlapped, although TC still had the highest average values ($PM_{2.5}$ = 113, BC = 7.9, and UVPM = 12.9 μg/m^3). Statistical comparison between pollutants measured at AA and roadsides is detailed in Table S4 (SI), for both separated and combined data series of the monitoring periods. Note that the comparison would be more justified between AA and each road because the 24 h measurements were taken simultaneously at these sites, but not between the two roads due to the fact that the measurements were not done on the same days (Fig. 4).

Overall, the 24 h levels of the pollutants at the busy road TC were higher than those at NN and AA. Similar to the GRIMM measurement results above, the 24 h $PM_{2.5}$ also exhibited a more uniform spatial distribution than PM_{10}. As for BC, which was mainly emitted from the diesel-powered vehicles, the busy TC road had statistically higher levels than AA. Less diesel vehicles (no buses allowed) in the small NN road induced lower BC levels at this roadside than TC, as expected. The fact that the coarse PM fractions at roadsides were largely contributed by the road dust resuspension suggested that the actions such as water spraying or banning of dusty vehicles from entering the roads can effectively reduce the PM_{10} pollution. There are also non-exhaust vehicle emissions of PM, for example, break wear and tire wearing, mainly in the coarse fraction of PM_{10} size (Grigoratos and Martini 2015), hence can also contribute to the high levels in busy roads which need further studies to quantify.

The UVPM values (measured at 370 nm), although cannot be considered as the real mass concentration (Jeong et al. 2004), were well correlated with BC (Fig. S3, SI). UVPM levels were consistently higher than BC for every sample (Fig. S3, SI and Table 1) and the difference "delta" = (UVPM – BC) would indicate an enhanced optical absorption at the wavelength. Jeong et al. (2004) reported that the measurements at this wavelength can be used for identifying the presence of UV absorbing organic compounds such as polycyclic aromatic hydrocarbons (PAHs). On average, the delta values in our study were 5–6 μg/m^3 at every site (Table 1) and this might indicate the abundancy of absorbing organic compounds in the air. Hansen (2005) suggested that UVPM may serve as an indicator for the biomass burning emissions and the enhanced optical absorption may indicate the presence of "brown carbon" aerosol associated

Table 1 Statistics of 24 h levels of pollutants (μg/m^3) during two sampling periods in comparison with other studies

Site	Sampling period	PM_{10}	$PM_{2.5}$	BC	UVPM
This study **AA** ($n=55$)	Winter ($n=33$)		100 ± 45 (32 ÷ 191)	5.8 ± 2.4 (1.8 ÷ 10.0)	11.5 ± 4.2 (4.6 ÷ 18.9)
	Transitional ($n=22$)	137 ± 60 (35 ÷ 240)	89 ± 41 (19 ÷ 147)	5.3 ± 2 (1.4 ÷ 8.3)	10.4 ± 3.7 (3.1 ÷ 16.3)
	Comparison between periods: p value		*0.245*	*0.383*	*0.317*
	Combined (winter & transitional)		96 ± 43 (32 ÷ 191)	5.6 ± 2.2 (1.4 ÷ 10.0)	11.1 ± 4.0 (4.6 ÷ 18.9)
This study **TC** ($n=25$ pairs)	Combined (winter & transitional)*	240 ± 37 (155 ÷ 293)	113 ± 46 43 ÷ 179	7.9 ± 2.5 (3.5 ÷ 11)	12.9 ± 3.8 (5.6 ÷ 19.3)
This study **NN** ($n=20$ pairs)	Combined (winter & transitional)*	138 ± 56 (58 ÷ 234)	94 ± 43 (86 ÷ 184)	6.2 ± 2.4 (5.5 ÷ 10.6)	11.2 ± 4.0 (3.2 ÷ 18.1)
Roadside, Hanoi [a]	Winter, average, Dec–February, 2007		76 ± 32 (26 ÷ 143)	1.5 ÷ 4.9	
Ambient, Hanoi [b]	Winter, monthly average Dec, 2017		70 ÷ 76		
Ambient, Hanoi [c]	Winter, daily average, 1998–1999		36.1 ± 1.3 (25 ÷ 150)		
Roadside, Bangkok[d]	Dry season, daily average PM, 2010		57 ± 13[d]	17.9 ± 6.6[e]	
Ambient&roadside, Beijing[f]	Winter & summer, urban area, 2003–2008		59 ÷ 132		

p values are the statistical significance of independent t test (2-tailed) of mean values between winter and transitional period; n, number of 24 h samples; bold values are statistically different at confidence interval level of 95%. The results of p values are based on 1,000 bootstrap samples. * – 24 h PM_{10} measured in transitional season only

[a]Hai and Kim Oanh (2013), [b]Thuy et al. (2018), [c]Hien et al. (2002), [d]Kim Oanh et al. (2013), [e]Hung et al. (2013), [f]Yang et al. (2015)

with the biomass combustion (https://mageesci.com/mproducts/sootscan-model-ot21-optical-transmissometer). It is worth mentioning that the most noticeable biomass open burning activity around HMR is the rice straw open burning. In Hanoi area, this activity is most intensive around May–June and October–November harvesting months when the weather is dry (Dong 2013; Lasko and Vadrevu 2018). Accordingly, it would be expected that the rice straw burning emissions may be reflected in UVPM values and the enhanced absorption of "delta" would be higher in the transitional period. However, as seen in Table 1, the average "delta" value at AA was actually slightly lower in the transitional period (5.1 μg/m^3) compared to winter (5.7 μg/m^3). This may be partly due to more intensive rain observed during the transitional period which would suppress the open burning activity. It is worth emphasizing that statistically, both BC and UVPM, respectively, were not different between the two periods (with $p > 0.3$, Table 1). Specifically designed monitoring program for BC and UVPM in a rice straw burning plume may better reveal the association between "delta" and the emissions. Other PM compositions, such as organic carbon, water soluble ions, and elements, should also be monitored to quantify the source contributions by receptor modeling (Hai and Kim Oanh 2013).

A comparison between weekdays and weekends showed that only at NN, the levels of 24 h PM_{10} in weekdays were significantly higher than weekends ($p = 0.004$, Table S5, SI) and this consistently reflected the weekday-weekend difference in the traffic flows in this street (Table S2, SI). The daily levels of all pollutants at TC and AA were not statistically different between weekdays and weekends, i.e., with p values ranged from 0.25 to 0.97 (Table S5, SI). This is consistent with the vehicle activities at TC that were not statistically different between weekdays and weekends (Table S2, SI).

The 24 h $PM_{2.5}$ levels obtained in our study for AA are presented in Table 1 along with those reported in Hanoi and other Asian cities. Our results are above those reported for winter in Hanoi about 15 years ago by Hien et al. (2002), both in the average and the range. Our results are also higher than the $PM_{2.5}$ levels measured at a mixed site along a busy road in Hanoi during winter about 8 years ago (Hai and Kim Oanh 2013). The higher levels found in this study may reflect an increasing trend of pollution in Hanoi over the years. More recently, Thuy et al. (2018) reported the $PM_{2.5}$ levels in Hanoi in 2017, based on online sensor records, and showed the highest monthly average levels of around 76 μg/m^3 observed in Dec, i.e., lower than our average winter level at AA of 100 μg/m^3. This is largely explained by their measurement site, which was a university microenvironment, compared to the congested area of our study. The roadside levels reported in our study were quite similar with those measured in Ho Chi Minh city, 97 ± 31 μg/m^3 (Giang and Kim Oanh 2014) or Beijing (roadside and ambient), 59–132 μg/m^3 (Yang et al. 2015). However, the roadside levels in this study were almost two times higher than the roadside levels reported in other Asian cities, such as in Bangkok, Thailand, 57 ± 13 μg/m^3 (Kim Oanh et al. 2013), or Hong Kong, 52 ± 18 μ/gm^3 (Lee et al. 2006).

The obtained BC levels in our study were higher than those measured at a mixed site along a busy road in Hanoi reported in Hai and Kim Oanh (2013) with elemental carbon (EC) of 1.5–4.9 μg/m^3. Our BC results, however, were lower than those measured at a busy roadside in the Bangkok city center, 17.6 ± 6.6 μg/m^3 (Hung et al. 2014), or downwind of an interstate highway in Los Angeles of 21.7 μg/m^3 (Zhu et al. 2002). On average, the BC values obtained in our study constituted about 6–7% of the $PM_{2.5}$ mass at all three sites. BC is a significant component of $PM_{2.5}$ emitted from diesel-powered vehicles (Kim Oanh et al. 2009) but the traffic in the selected streets was dominated by MC and other gasoline powered vehicles (Fig. S2, SI) which may be a reason for the lower BC levels in our study area compared to other cities.

Influence of local meteorological conditions

The variations in 24 h levels of pollution can be linked to the day-to-day variations in the emission sources and meteorological conditions that influence both the local and regional transport of pollution, as well as the wet/dry removal and formation of secondary pollutants. The similar ranges and variations patterns of 24 h $PM_{2.5}$ and BC at the 3 sites, discussed above, suggested that there were other influencing factors than the traffic emissions in the roads alone. For example, the daily traffic activities in the studied roads were significantly higher during weekdays than weekends, especially at rush hours (Fig. 2), and the difference was statistically significant at $p < 0.05$ for NN and at $p = 0.06$ for TC (Table S2, SI). However, the difference between the weekend and weekday average $PM_{2.5}$ and BC, respectively, was not statistically significant (Table S5, SI).

On the other side, the local weather conditions of rainfall and wind speed showed clear associations with the variations in the daily pollutant levels (Fig. 4). Low $PM_{2.5}$ and BC levels were observed during the days with rain and/or higher wind speed. Note that the samples collected during heavy rainy days were considered invalid and excluded from the data analysis and the monitoring results are not shown in Fig. 4 (blank spaces). During 30 winter monitoring days with valid samples, there were 6 days recorded with fine dizzy rainfall (< 2 mm daily, with a total of 4 mm for 6 days) that is typical for winter in Hanoi (Hai and Kim Oanh 2013). Out of 25 monitoring days with valid samples in the transitional period, there were also 6 rainy days but with a relatively high rainfall, i.e., totaled at 67 mm over 6 days. Besides, days with rain were concurrently observed with higher wind speeds; hence, both wet removal and better dispersion explained for the lower levels of $PM_{2.5}$ and BC measured at the sites. Daily pollution levels between 24 and 27 Jan were the lowest in the winter period with 24 h $PM_{2.5}$ in fact mostly meeting the NAAQS (50 μg/m^3). The rainfall on these days was too small (< 0.6 mm) to induce any effective wet removal of the pollutants but the relatively strong wind (> 1.5 m/s) would enhance the dispersion. After heavy rain events, such as those occurring on Jan 9–12, Oct 9–10, Oct 29, and Nov 7, the pollution levels were consistently low for 2–3 following days. Apart from the strong wind conditions, e.g., those observed around Oct 31 (> 2 m/s), the heavy rain events before the monitoring days effectively scavenged the air pollution hence reduced the background ambient pollution in the city. In addition, the wet conditions after rain would also substantially reduce the open burning activities, i.e., rice straw and solid waste open burning for a few following days.

Furthermore, it is worth emphasizing that meteorology represents a totality with all the local weather conditions are interlinked, which in turn are related to the synoptic weather patterns governing the region (Hai and Kim Oanh 2013). Accordingly, in winters, the PM levels in Hanoi are normally higher than other seasons due to more stable winter atmosphere, less wet removal, and high potential of long range pollution transport following the Northeast monsoon (Cohen et al. 2010). The effects of large scale meteorology on the pollution levels in Hanoi are further discussed in the airmass types associated with the HYSPLIT trajectory analysis in the "Potential long-range transport contribution to pollution levels PM and BC at ambient site" section.

Spatial distributions of pollutants

The average levels of PMx (both filter-based and GRIMM data), BC, and BTEX over the monitoring period at the 3 sites are presented in Fig. 5. The larger road TC had the highest levels of all pollutants. The small road NN had higher levels than AA only for BTEX, PM_{10}, and BC, while the opposite was seen for fine PMx ($PM_{2.5}$ and PM_1) with AA having higher levels than NN. The largest difference between the roadsides and AA is shown for BTEX, i.e., on average, the levels at TC were 5 times higher than AA and that at NN were 4 times higher than AA. For the small distance between the sites of 150 m, the corresponding horizontal gradients of BTEX were substantial. The large spatial variations of the BTEX within this relatively small congested area suggested that these pollutants were mostly released from the on-road traffic exhaust emissions. The second largest difference in the levels between the sites was found for PM_{10}, with TC and NN levels were about 2.0 and 1.3 times higher than AA, respectively, which in turn indicated a substantial contribution of the coarse particles from road dust resuspension caused by vehicle movements in the roads. These coarse particles ($PM_{10\text{-}2.5}$) are mainly of geological origin and because of their relatively large size they can quickly

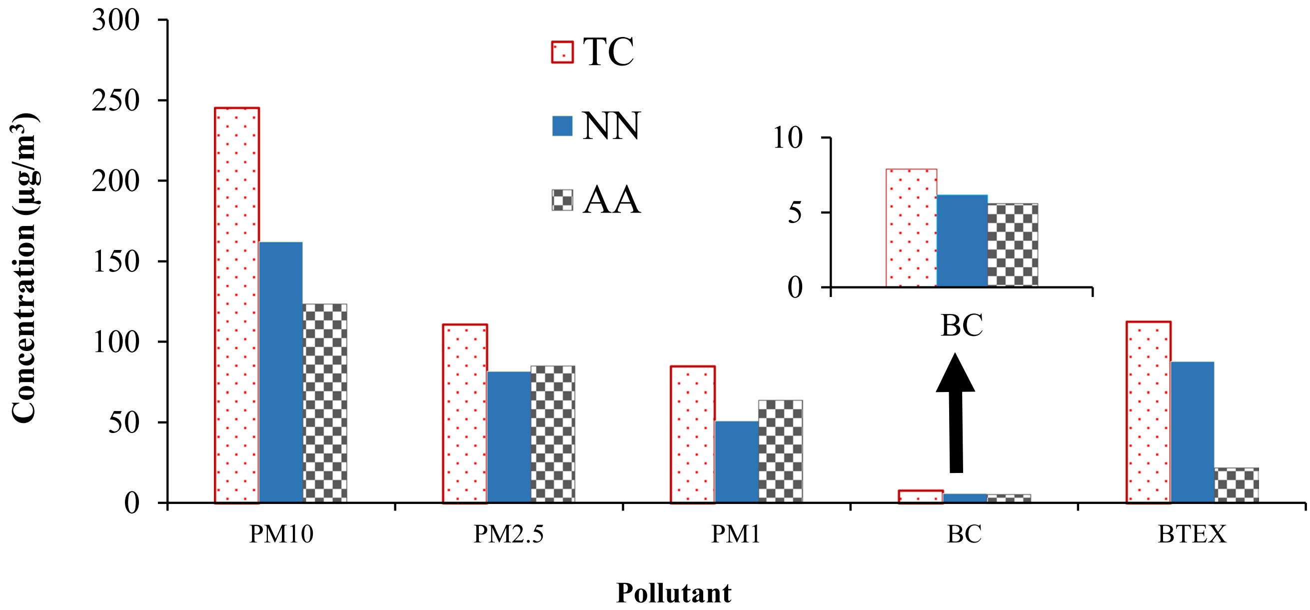

Fig. 5 Spatial variation of pollutants levels in the study area

deposit back to the earth. BC had less variations between the sites than PM_{10}, i.e., the levels at TC and NN were about 1.4 and 1.1 above AA, respectively. Although BC was primarily released from the diesel vehicle exhaust, there may be other sources of BC in the area such as residential combustion and solid waste open burning. Besides, BC particles are of submicron size, which was measured as a component of $PM_{2.5}$ (using the $PM_{2.5}$ filter samples) in this study; hence, they can disperse better and are more uniformly distributed than the coarse particles.

The spatial distribution of both fine fractions ($PM_{2.5}$ and PM_1), however, was different from the other pollutants. TC still had the largest levels of but the levels at AA were actually higher than NN, more significant for PM_1 than $PM_{2.5}$. The higher levels at TC, i.e., about 1.3 times higher than AA, again indicated the contribution from the large fleet of vehicles on this road. However, the higher levels at AA than that at the small road NN, especially for PM_1 (1.25 times higher), suggested a complex interaction between emissions, secondary formation, and dispersion of these fine particles. Beside the fact that there are many emission sources in the study area, these fine particles are also formed in the atmosphere from the precursors that are released from multiple sources, both local and regional/distant. These fine particles present in the atmosphere for a longer time and their behavior is more influenced by meteorological conditions which induce the dispersion (mixing, regional/long range transport) of particles and precursors, as well as the secondary formation process. Therefore, their levels, as well as diurnal variations, were more uniformly distributed among the sites and the ambient site may have the levels above that at the small road.

These large differences in BTEX and PMx levels, as well as in the diurnal variations between the monitored sites located at 150 m away from each other in this small urban area suggested that to adequately characterize the exposure risks of population, the monitoring should generate data with sufficient fine spatial resolutions. Due to the high resources required to generate such monitoring data, a properly evaluated low cost sensor network or a refined modeling study may be used as the supplement.

Multivariate relationship analysis

The results of PCA provided further insights into the multivariate relationship between pollutant levels, traffic flows, and meteorology. The pollutants included bi-hourly levels of $PM_{2.5}$ and coarse particles $PM_{10-2.5}$ (the difference between simultaneously measured PM_{10} and $PM_{2.5}$ by GRIMM), as well as BTEX levels (Phuc and Kim Oanh 2018). Note that PM_1 levels were automatically rolled out due to the very high correlations with $PM_{2.5}$ at every site (Table S3, SI). The meteorological variables included temperature (Temp), relative humidity (RH), barometric pressure (P), and wind speed (WS). Due to the street canyon configuration of the roads, the wind direction was not included in the roadside datasets for PCA, but it was included in the PCA analysis for AA.

At TC, the first four principal components were retained which collectively explained 78% of the total variance (Table 2) of the original data set of 1,134 data points (14 variables × 81 measurements). PC1 explained 33% of the total variance and showed high loadings (above 0.4) of the pollutants, ranked in descending order of T, X, B, E, $PM_{2.5-10}$, the traffic flow of MC&DTC, and meteorological variables of Temp (−0.51, negative) and P (0.57). The dominance of the gasoline powered MC&DTC fleet in the urban road in Hanoi (89% in TC) explained the significant association with BTEX which was also previously confirmed (Phuc and Kim Oanh 2018). The strong association between $PM_{2.5-10}$ with MC&DTC activities should be linked to the mechanical turbulence induced by the on-road traffic fleet (dominated by MC&DTC) that enhanced road dust resuspension. PC2 (19%) showed high positive loadings of $PM_{2.5}$, $PM_{2.5-10}$ and traffic flows of diesel vehicles and PC&Taxi, and a negative loading of WS. Large amount of particles (mainly fine fractions) emitted from diesel vehicle exhausts (Kim Oanh et al. 2009) explained their association with $PM_{2.5}$. The high loading of PC&Taxi appeared in this PC may be largely due to the fact that they commonly co-operated in this road with the diesel vehicles, e.g., during late evening (Fig. 2). A negative loading of WS showed the wind effects on the dispersion of the pollutants. The last two components, each explained around 13–14% of the total variance, had high loadings of meteorological variables and traffic flows, and this still needs further insights into the relationship between the traffic modal patterns in different weather conditions to explain. For example, the association between car (negative), Temp (negative), and RH (positive) may suggest that people traveled more by car and taxi (than by open MC) in the weather conditions with low temperature and high humidity (rainy/foggy). However, further investigation is required to confirm such relationship and this is beyond the scope of the current study. The positive loadings of two vehicle groups of MC&DTC and Van&Pick, and negative leadings of Diesel shown in PC3 suggested that the first 2 groups normally co-operated in this street during daytime, opposite to that of diesel vehicles which operated at late evening hours (Fig. 2).

For NN, the first three components were retained that together explained 77% of the total variance (Table 2) in the original dataset of 1,106 data points (14 variables × 79 measurements). The loading pattern on PC at NN was quite similar with those observed in TC. PC1 (35%) shows significant positive associations between BTEX (> 0.90 for every species) with traffic flows of MC&DTC (0.87) and PC&Taxi (0.51) and this in turn indicates a strong influence of these gasoline powered vehicles on the BTEX levels. PC2 (28%) showed strong associations between $PM_{2.5}$ and $PM_{2.5-10}$ with diesel vehicles. High loadings of the meteorological variables of Temp (negative), P

Table 2 Loadings of variables on selected PC for TC and NN roadsides (SPSS results for normalized variables with Varimax)

Variables	TC road					NN road			
	Principal component				Communality	Principal component			Communality
	1	2	3	4		1	2	3	
Temp	**−0.51**	−0.02	**0.51**	**−0.65**	0.94	−0.22	**−0.88**	−0.32	0.92
RH	−0.04	−0.14	−0.12	**0.90**	0.84	0.07	0.24	**0.90**	0.87
P	**0.57**	0.09	**−0.51**	0.38	0.74	0.12	**0.91**	0.17	0.87
WS	−0.03	**−0.84**	0.00	0.27	0.78	−0.33	**−0.63**	−0.30	0.60
$PM_{2.5-10}$	**0.51**	**0.56**	−0.24	−0.12	0.64	0.29	**0.59**	**−0.56**	0.75
$PM_{2.5}$	0.19	**0.73**	−0.28	0.24	0.70	0.19	**0.87**	0.12	0.81
Benzene	**0.95**	−0.18	0.05	0.05	0.93	**0.97**	0.09	0.08	0.95
Toluene	**0.98**	−0.13	−0.02	0.02	0.98	**0.92**	0.36	0.06	0.97
Ethylbenzene	**0.66**	0.02	−0.02	0.01	0.44	**0.91**	0.29	−0.05	0.91
Xylenes	**0.97**	−0.10	−0.07	0.05	0.97	**0.92**	0.33	0.10	0.96
MC&DTC	**0.67**	0.15	**0.59**	0.04	0.82	**0.87**	−0.13	0.25	0.84
PC&Taxi	0.14	**0.67**	−0.08	**−0.40**	0.63	**0.51**	0.08	**0.68**	0.72
Van&Pick	−0.01	0.05	**0.79**	−0.19	0.66	−0.21	−0.39	0.09	0.20
Diesel	−0.08	**0.73**	**−0.52**	−0.25	0.88	−0.22	**0.58**	−0.23	0.44
Eigenvalues	*5.0*	*2.5*	*2.3*	*1.1*		*6.4*	*2.7*	*1.7*	
% Variance	*32.7*	*19.0*	*13.7*	*12.7*		*34.7*	*28.5*	*14.1*	
% Cumulative	*32.7*	*51.7*	*65.4*	*78.2*		*34.7*	*63.1*	*77.2*	

Temp, air temperature; *RH*, relative humidity; *P*, barometric pressure; *WS*, wind speed; *WDI*, wind direction index; *MC&DTC*, Motorcycle and Delivery Three-wheel; *PC&Taxi*, Sedan car, Taxi, and multi-purpose/sport-utility vehicle (MPV/SUV); *Van&Pick*, MiniVan and Pick-up vehicles, *Diesel*, all types of diesel-fueled vehicles

[a]**Bold v**alues show significant loadings (> 0.4) of the variable on the PC

(positive), and WS (negative) in PC2 implied their significant roles in the PMx variation. PC3 explained 14% of the total variance and showed a strong negative relationship between coarse PM with RH that linked to the fact that humid weather induced by rain would suppress the road dust resuspension. This PC also showed a positive loading of PC&Taxi that again may link to the reality that in rainy/foggy weather people prefers these close vehicles than MC.

The PCA results for AA (Table S6, SI) retained four PCs which explained 78% of the total variance of the original dataset of 1,260 points (15 variables × 84 measurements). The patterns of the loadings on PCs at AA were different from those at the roadsides. PC1 (28%) showed strong associations between $PM_{2.5-10}$ (loading of 0.82) with all meteorological variables, including a positive loading of Temp (0.94) and negative loadings of above 0.6 for RH, P, and WS. The interrelation suggested that the dry weather conditions (low RH) and high Temp likely to enhance resuspension of coarse particles of soil/road dust. Significant loadings of diesel (negative) may be due to the fact that more diesel vehicles were observed at late evening when low Temp and high RH prevailed. Likewise, the opposite may be for the Van&Pick which generally had higher shares around noon, 10:00–14:00, when Temp was high hence showed a positive loading. PC2 (24%) showed high positive loadings of BTEX species and negative loadings of RH and WS. It is interesting to note that PC3 (14%) had high positive loadings for both *WDI* and $PM_{2.5}$ and this association of $PM_{2.5}$ with the wind direction implying the role of wind transport to the AA site. Likewise, the negative loading of ethylbenzene on this PC also indicated its dependency on the wind direction (Phuc and Kim Oanh 2018). In fact, higher *WDI* values are associated with the wind blowing from the north-northeast directions, i.e., locally from TC to AA, whereas regionally, these may indicate the long-range transport associated with the northeast monsoon (see detail in the next section). On the other side, low *WDI* values are associated with the southwest monsoon, and locally, it is from the small road NN to AA. Again, the opposite signs of loadings by Diesel and Van&Pick largely indicated that they did not appear together in the roads. PC4 (11%) had high loadings for MC&DTC and PC&Taxi as the two types of vehicles had similar diurnal variations (Fig. 2). The PCA results thus indicated that the pollution levels at AA were affected more by meteorological conditions than the vehicular emissions which is opposite to the cases of TC and NN. Overall, the pollution levels measured at AA reflected a complex interaction between meteorology, chemistry, and emissions of local and regional/distant origins.

It is interesting to note that in all PCA cases, the Temp and P, if both appeared significantly in a PC, always had opposite loading signs and this largely due to the situation that a cold weather spell is observed in Hanoi when a high pressure ridge extending from the central China reaches the area (Hai and Kim Oanh 2013). Overall, the PCA results in this study agreed well with the PCA results reported in the previous study (Phuc and Kim Oanh 2018) which analyzed the datasets with similar meteorology and traffic flows but only with BTEX as the pollutants (not PMx). The results of both studies thus showed a strong influence of different on-road traffic categories to the considered pollutants levels at roadsides, whereas at the ambient site, the influences of meteorological variables were more important.

Potential long-range transport contribution to pollution levels at ambient site

To examine the potential contribution of the regional/long-range transport pollution to the site, we used 3-day HYSPLIT backward trajectories of airmass arriving at AA in every monitoring day (Fig. S4, SI). The AA site was selected for the analysis because it had less direct impacts of the local traffic emissions. During the winter period, majority of the airmass arriving at AA were originated from the west territories, e.g., India, and transported east-ward passing Northern Thailand, Myanmar, and Laos. Only on 13, 14, 21, and 22 Jan, the trajectories were more northerly and passing the Southwestern part of China. During the transitional period, two airmass types were mainly observed. A higher frequency was seen for the airmass type which originated from the east and had long marine pathways before arriving to the site. A lower frequency was shown for the airmass type that originated from the Southeastern part of China and had more continental pathways before arriving to the site. The airmass arriving at the site were grouped into 4 types according to the origins and pathways: North (N), East (E), South (S), and West (W). The W category had the highest frequency of occurrence with 40%, followed closely by N with 38%, E with 17%, and finally S with 5%.

Table 3 presents the statistics of the $PM_{2.5}$, BC, and UVPM in different airmass types, together with the previously reported BTEX results (Phuc and Kim Oanh 2018). The East airmass type (E) was characterized by the lowest daily levels of all pollutants among the four types. The E type occurred mainly in the transitional period and was originated from the Pacific Ocean with long marine pathways before arriving at the site (Fig. S4, SI), which implied less potential of the regional pollution transport. This airmass type also induced higher rainfall (observed during the transitional period) hence a more intensive wet removal of pollution (and less biomass open burning). The North airmass type (N), originated from the north and mainly had a long continental pathway over China, was characterized by the highest levels of $PM_{2.5}$, BC, UVPM, and most BTEX species. Locally, the meteorological conditions associated

Table 3 Statistics of 24 h average levels of pollutants (μg/m^3) measured at the ambient site associated with airmass types

Airmass	Statistics[a]	$PM_{2.5}$	BC	UVPM	Benzene	Toluene	Ethylbenzene	mp-Xylenes	o-Xylene
North (38%), $n=16$	Mean ± SD	116 ± 35	6.6 ± 1.8	13.0 ± 3.4	9.0 ± 4.1	10.3 ± 6	3.5 ± 2.8	6.3 ± 2.9	4.6 ± 2.2
	Median	114	6.6	12.7	8	7.7	2.4	5.9	3.1
East (17%), $n=10$	Mean ± SD	48 ± 27	3.4 ± 1.3	7.0 ± 2.4	5.5 ± 1.3	6.2 ± 2.7	3.0 ± 1.3	4.1 ± 1.5	2.5
	Median	48	3.0	6.3	5.9	5.3	2.5	3.7	2.5
South (5%), $n=2$	Mean ± SD	113 ± 26	6.4 ± 1.6	12.4 ± 0.6	7.9 ± 3.1	10.5 ± 2.8	4.1 ± 2.2	4.8 ± 1.8	n/a
	Median	113	6.4	12.4	7.9	10.5	4.1	4.8	n/a
West (40%), $n=17$	Mean ± SD	105 ± 47	6.0 ± 2.4	11.6 ± 4.5	8.3 ± 2.6	10.7 ± 4.2	3.8 ± 0.8	5.9 ± 2	3.5 ± 0.6
	Median	113	6.0	12.4	7.9	10.7	3.5	5.8	3.4
Comparison, ***p*** value[b]									
N-E		**0.001**	**0.01**	**0.01**	**0.03**	0.11	0.92	0.31	n/a
N-W		0.89	0.357	0.316	0.95	0.99	0.99	1.0	0.19
E-W		**0.01**	**0.009**	**0.009**	**0.01**	**0.01**	0.58	0.69	n/a

n, number of sampling days belonging to airmass pathway type; *p* values are the significance of the difference in mean levels of pollutants between North and East (N-E), North and West (N-W), and East and West (E-W) types; southern airmass type had a small sample size (i.e., 2) was not included in the test; [a]based on 1,000 bootstrapped samples; [b]post hoc analysis results following univariate ANOVA test with significant level of 0.05; *n/a*, not analyzed due to small number of samples with detectable levels; bold values are statistically significant

with the high-pressure ridge over the study area, i.e., stagnant air with limited mixing height, calm wind, and low rain, typically observed when the N airmass type prevails would enhance the build-up of high levels of air pollution (Hai and Kim Oanh 2013). There was no significant difference between the levels of any pollutants measured in the two continental airmass types of N and W, meaning statistically both had equally high pollutants levels. The origin and the continental pathways of these airmass types show the potential contribution of the regional pollution transport to the site. The difference in the mean pollutants levels between E and N airmass types was statistically significant at $p \leq 0.05$ for $PM_{2.5}$, BC, UVPM, and benzene, while that between E and W has one more pollutant (toluene) also significantly different. The South type was observed only in 2 monitoring days hence was not included in this statistical test (post hoc univariate ANOVA test). It is worth mentioning that species with a relative long atmospheric life of $PM_{2.5}$ and BC (a few weeks) and benzene (about 9 days) are more reliable tracers of the regional transport, and in fact, their levels exhibited the statistically significant difference between E and N, as well as between E and W. Other species (TEX) have a shorter atmospheric life, from a few hours to less than 2 days, hence may not serve as suitable tracers for long range transport studies. Probably due to this reason, their mean levels were not statistically different between the airmass types.

Conclusions

High levels of PM and BC were observed during the monitoring periods with 24 h $PM_{2.5}$ exceeded the NAAQS (50 μg/m^3) by 77–92% of the measurements at all the sites. The 24 h PM_{10} levels exceeded NAAQS (150 μg/m^3) by 100% at the big road TC and by 40–46% at the small road NN and the ambient site AA. At the roadsides, coarse particles ($PM_{10\text{-}2.5}$) contributed more significantly to 24 h PM_{10} mass (50–54%) than at AA (< 40%). Hourly PM_1 and $PM_{2.5}$ were strongly correlated, with PM_1 sharing the majority of $PM_{2.5}$ mass, more at AA with 82–87% than the roadsides with 71–75%. Within this small area of 300 m wide with heavy traffic, the levels of the pollutants were found to be highly fluctuating in space, more for PM_{10} and BTEX than for fine particles ($PM_{2.5}$ and PM_1). The finding indicates the need for pollution data with sufficiently refined spatial resolution for health effect studies.

The traffic activities strongly influenced both levels and diurnal variations of the pollutants, not only at roadsides but also at AA located 150 m away from the roads. Diurnal variation patterns of fine PMx ($PM_{2.5}$ and PM_1) at all sites reflected the morning rush hours, but the evening levels remained high until late night owning to several factors, including the traffic compositions change (more diesel-powered trucks), meteorological conditions, other potential emission sources, and secondary formation process. The diurnal variations of PM_{10} at 3 sites were more consistent with the traffic flows showing more clearly both morning and afternoon peaks, which indicated the influence of road dust resuspension induced by vehicle activities.

Multivariate analysis (PCA) results showed that the ambient levels were complexly influenced by local emissions, meteorological conditions, and regional/long-range transport that calls for comprehensive management strategies. The roadside pollution levels showed strong influences of traffic total flows and traffic compositions on the BC levels (diesel vehicles), BTEX levels (MC fleet), and $PM_{10\text{-}2.5}$ originated from the soil/road dust resuspension. In particular, actions to reduce the road dust resuspension, such as water spraying and ban of dusty vehicles, should be effective to reduce the roadside PM_{10} pollution.

Local weather conditions strongly influenced the pollutant levels, with lower 24 h $PM_{2.5}$ and BC observed during the rainy days and/or higher wind speed. Regionally, the airmass types had continental origin and long continental pathways were shown to associate with the highest levels of $PM_{2.5}$, BC, and other pollutants at the AA site. Airmass with marine origin and pathways (E) prevalent during transitional period were associated with lower pollution levels.

Further studies should produce continuous monitoring data over longer period to better reveal the levels and influencing factors of PM pollution in this congested area. Especially, detailed PM composition data are necessary for source apportionment studies by receptor modeling which would comprehensively quantify the contributions from emission sources. Data with sufficiently fine resolution are required to assess health effects resulted from exposure to air pollution in populated urban areas. A longer simulation period for HYSPLIT backward trajectories, e.g., 5-day or 10-day, should be considered to get a better coverage of the origins and pathways of the airmass influencing the PMx levels.

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Author contribution Nguyen Hong Phuc: data collection, data analysis and visualization, writing: draft preparation, revision. Nguyen Thi Kim Oanh: conceptualization, study design, data review and analysis, writing: draft, revision and finalization of the paper content.

Declarations

Ethics approval The authors confirm that the manuscript has been read and approved by all authors.

Consent to participate All authors have been personally and actively involved in the research and will hold themselves jointly and individually responsible for the content of the manuscript.

Consent for publication All authors consent to publish this research.

Competing interests The authors declare no competing interests.

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