

# PILOT STUDY ON ASSESSMENT OF TRACE METALS IN PM<sub>10</sub> AT ROAD SITES IN BAC GIANG PROVINCE, VIETNAM

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## Abstract

In this study, characterization of PM<sub>10</sub> mass and its trace metals concentration at road sites in Bac Giang province located in the North of Vietnam during two different seasons has been performed. PM<sub>10</sub> samples at 11 road sites using the high volume SIBATA HV-500R (Japan) at the flow rate of 400 L/min were collected during 10 hours (from 7:00 to 17:00) at flat low-lying plain, midland, mountain and highland areas. Data on PM<sub>10</sub> mass and metals were used to estimate the pollution level of PM<sub>10</sub> and trace elements bounded with PM<sub>10</sub>. The results showed that 10-hour mean PM<sub>10</sub> concentrations at road sites in Bac Giang ranged from 103.4 µg/m<sup>3</sup> to 577.9 µg/m<sup>3</sup> with the average value of 292.6 µg/m<sup>3</sup>. Generally, the level of trace metals appeared to be higher in autumn than that in summer period. Additionally, concentrations of trace metals were higher at the sites with high traffic density than those with low traffic density. While the concentrations of Cd, Mn and Cr exceeded the guideline values of WHO, arsenic concentration was larger than the concentration limit of EU Directives. The degree of contamination values suggested that level of metal was high contamination at flat low-lying plain, while those values were from low to considerable contamination at the mountain and highland areas. The pollution load index values at most of sampling sites were larger than 1, which indicated the existence of dust pollution at road sides in Bac Giang province.

**Keywords:** PM<sub>10</sub>; trace metals; degree of contamination; pollution load index; roadsides.

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## 1. Introduction

Nowadays, serious environment pollution problems, especially elevated air pollution due to rapid industrialization, population growth, urbanization, dramatic economic rise and increased vehicular emissions have been concerning issues all over the world. Noticeably, air pollution has been the most urgent problem in Vietnam causing more approximately 70,000 deaths per year [1]. According to the environment report of Vietnam Ministry of Natural Resources and Environment (MONRE), in 2016, the pollution level of particulate matter (PM) in urban areas, traffic routes have exceeded the

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Vietnam national ambient air quality standard (NAAQS). A number of studies have demonstrated that traffic, industry emissions, combustion processes and other anthropogenic activities were the main sources of fine particles pollution [2, 3]. In addition, re-suspended road dust, biomass burning, concrete production were primary sources causing coarse particles pollution [2, 4, 5]. Furthermore, many researches figured out that high concentration of particulate matter PM<sub>10</sub> not only caused great impact on the agriculture, ecosystem and economic and global climate [6, 7], but also resulted in adverse human effects such as respiratory problems [8, 9].

Ambient particulate matter is a complex, heterogeneous mixture of harmful metalloid, heavy metals, organic carbon, ammonium, nitrate, sulfate, mineral dust, trace elements [10]. These different constituents are formed by various mechanism associated with both natural and anthropogenic sources. Airborne PM<sub>10</sub> is a carrier of several toxic constituents into the human body via different pathways i.e. inhalation, ingestion and dermal contact. Therefore, metal elements could adversely affect human health such as increasing the incidence of respiratory and cardiovascular diseases.

There are number of scientific researches on the chemical characterization and source apportionment of PM<sub>10</sub> and health risk assessment in industrial, urban area and traffic sites worldwide [5, 11–14]. Rössli (2001) suggested that the sources of particles, the season of the year, the prevailing weather conditions and the chance for dispersion had great impact on the chemical composition of atmospheric particulate matter. These studies concluded that re-suspended, motor vehicle and traffic, coal combustion and industrial activities were the main sources of the measured elements. Recently, the emissions of trace metals from various sources in the atmosphere have been reported in a number of studies worldwide. The results showed that the concentrations of the trace elements in particulate matter were high in Asia, particularly in developing countries and industrial areas [5, 14–16]. Moreover, some studies found higher concentrations of metals and PM<sub>10</sub> mass in industrial, urban areas than the background areas [10, 11, 17, 18]. Concentrations of metals and PM<sub>10</sub> mass during the dry season were higher than those in the rainy season [19].

There have been number of studies analyzing the particulate matter pollution in Vietnam [20–22]. PM<sub>10</sub> and the trace metals bounded with PM<sub>10</sub> have been previously researched in Hanoi [19, 23] and Ho Chi Minh [24]. However, there is no investigation on characterization of PM<sub>10</sub> and its chemical compositions at road sites in Bac Giang province. According to our best knowledge, only [13, 24] reported the level of metal contamination in street dust in Hanoi and roadside dust along the highways in the North of Vietnam [20, 21]. These studies showed that the high concentration of metals in dust observed from downtown area and the high traffic density. Therefore, the purpose of this study is to investigate the pollution level of PM<sub>10</sub> and PM<sub>10</sub> bounded trace elements at the road sites in Bac Giang province which has been experienced a time of urbanization and industrialization with fast speed of road expansion. According to a report on the implementation of socio-economic development plan of Bac Giang province in 2019, the economic growth of Bac Giang was estimated at 16.2 percent, ranking the second in Vietnam.

## 2. Material and methods

### 2.1. Study area

Bac Giang is a developing area with high urbanization speed. According to the general statistics office of Vietnam [25], the total population is about 210,000 and total area is 667,736 km<sup>2</sup>. In recent year, the substantial expansion of infrastructure including national highways, rural roads and industrial parks would cause the degradation of air quality in Bac Giang. Key industries in Bac Giang are

fertilizer, high technology, engineering, building materials, assembling automobiles, agricultural processing, forestry products, textiles and electric power generation. According Bac Giang portal, there are five industrial parks including Dinh Tram at Viet Yen with area of 100 ha, Song Khe - Noi Hoang at Yen Dung with area of 180 ha, Quang Chau at Viet Yen with area of 426 ha, Van Trung at Hiep Hoa with area of 442 ha, Hoa Phu at Hiep Hoa with area of 207 ha. The climate of Bac Giang province has the dominant features of tropical, temperate climate zone of the Northern Vietnam. There are two distinct seasons: rainy season (May to September) and dry season (November to March). The annual rainfall in Bac Giang is 1584 mm with large amount of rainfall (more than 240 mm) happening in June, July, August. Precipitation amount is lesser in transition months (April and October) and winter months than that in summer period [26].

Fig. 1 shows the map of traffic sampling sites in Bac Giang province with topographic information obtained from Aster DEM 30 m spatial resolution [27]. Roadside PM<sub>10</sub> samples were collected at 10 road sites in Bac Giang province. These sampling sites represented the impact from traffic emissions as the major pollution sources. The sampling sites were divided into two groups: traffic 1 and traffic 2 (Fig. 1). The traffic 1 contained Hiep Hoa (HH) and Viet Yen (VY), which located in the flat low-lying plain and midland region. Son Dong (SD), Luc Nam (LN), Luc Ngan (LNg), Yen The (YT), Tan Yen (TY), Yen Dung (YD) and Lang Giang (LG) were traffic 2 sites, which located in the mountain and highland areas. The vehicle density of traffic 1 group is higher than that of the traffic 2 group.

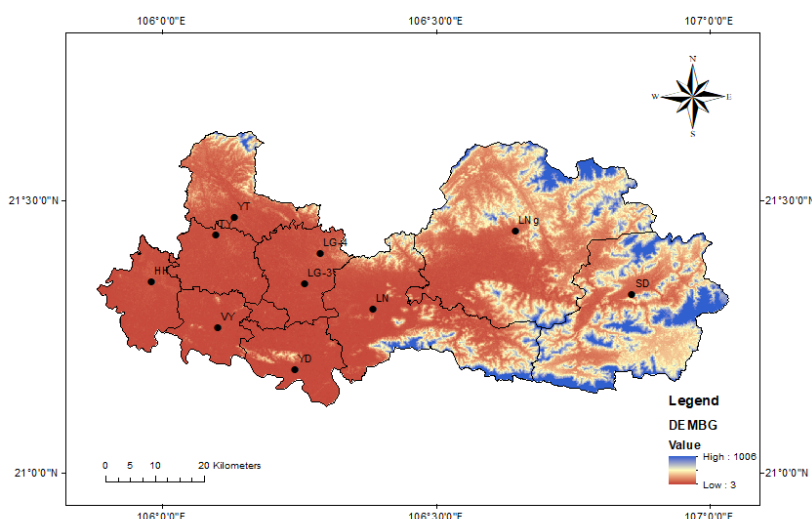


Figure 1. Map of traffic sampling sites in Bac Giang province

## 2.2. Sample collection

Samples were collected during summer (June - July) and autumn (October) periods in 2018. The samplers were placed approximately 1.5 m above the ground according to the Directive European Council 2008, 2010 (Annex III, Microscale sitting of sampling points). A ten-hour sampling was performed from 7:00 to 17:00 at each sampling site using the high volume SIBATA HV-500R (Japan) at the flow rate of 400 L/min. Quartz fiber filter papers (Advance, QR-100, size 110 nm, Japan), which have been dried for 6 hours at 400 °C in an oven, were used for collecting PM<sub>10</sub> samples. Filter papers were preconditioned at least 48 hours in desiccators under the conditions of temperature of  $25 \pm 2$  °C and relative humidity  $50 \pm 5\%$  before and after sampling. After collection, these samples were

sealed in clean plastic bag, then were transported to the laboratory and stored in a desiccator with silica gel particles.

### 2.3. $PM_{10}$ mass concentration and trace metals analysis

The  $PM_{10}$  mass of each sample was determined gravimetrically by subtracting the initial sample filter weight from the final sampled filter weight. The quartz fiber filters samples were repeatedly weighed by an analytical balance (Adam AEA-160DG,  $d = 0.01/0.1$  mg) until three reproducible values were obtained.

All chemical used in this study were of analytical grade and purchased from Merck. The detail of procedure was performed according to our previous report [28]. Briefly, a quarter of the samples filter was digested in 5 mL of mixed acid solution  $HNO_3/HCl$  (1:3, v/v) and kept on a hot plate at high temperature until transparent solution was boiled. After complete digestion, the digested sample was heated at low temperature until nearly dry to remove excess acid. Heavy metals (for As, Cd, Co, Cr, Cu, Ni, Mn, V, Pb and Zn) were determined by ICP-MS (ELAN 900, Perkin Elmer). A blank sample, a duplicated sample and a spike sample were analyzed with real samples of each bath for the quality control. The relative standard deviation of each element is within 10%, the analytical errors were less than 10%. The detection limit for most of the trace elements was  $0.01 \text{ ng/m}^3$ .

### 2.4. Assessment of trace metal contamination level

Contamination levels of trace metals in  $PM_{10}$  were assessed using contamination indices including contamination factor ( $C_F$ ), the degree of contamination ( $C_d$ ) and the pollution load index ( $P_{Li}$ ).  $C_F$  and  $C_d$  were calculated using the method described in Hakanson (1980) [29] as following equations:

$$C_F = C_{metal}/C_{background} \quad (1)$$

$$C_d = \sum_{i=1}^n C_f \quad (2)$$

where  $C_{metal}$  is the sample metal concentration,  $C_{background}$  is the background value of that metal. The background value of elemental concentration was taken from typical U.S. National Ambient (Urban) Air Concentrations from EPA (majority of the values are based on  $PM_{10}$  measurements) [10, 30]. According to Hakanson (1980) [29],  $C_d$  has four categories as follows:  $C_d < 8$  is low contamination;  $8 \leq C_d < 16$  is moderate degree of contamination;  $16 \leq C_d < 32$  is considerable degree of contamination;  $C_d \geq 32$  is very high degree of contamination.

The pollution load index ( $P_{Li}$ ) was used to evaluate the elemental contamination of the ambient environment. The  $P_{Li}$  was obtained as a concentration factor of each heavy metal with respect to the background value.  $P_{Li}$  has four categories including:  $P_{Li} < 1$  signifies no pollution;  $P_{Li} = 1$  shows baseline levels of pollution and  $P_{Li} > 1$  signifies existence of pollution.  $P_{Li}$  can be calculated as follows, as suggested by [31].

$$P_{Li} = \sqrt[n]{C_{F1} \times C_{F2} \times C_{F3} \times \dots \times C_{Fn}} \quad (3)$$

where  $n$  is the number of metals studied and  $C_F$  is the contamination factor.

### 3. Results and discussion

#### 3.1. $PM_{10}$ concentrations

During the study period, at 10 road sites, the average  $PM_{10}$  concentration ranged from  $103.4 \mu\text{g}/\text{m}^3$  to  $577.8 \mu\text{g}/\text{m}^3$ , the mean concentration of  $PM_{10}$  in LG-3 was 1.5 times more than the  $PM_{10}$  concentration in other sites. As shown in Table 1, the average  $PM_{10}$  concentration at LG-3 was the highest, which was followed by  $TY > LNg > VY > LG > HH > YD > SD > YT$ . The average concentration of  $PM_{10}$  at all road sites except for SD and YT was much higher than the national ambient air quality standard ( $150 \mu\text{g}/\text{m}^3$ , QCVN 05:2013/BTNMT), suggesting very serious  $PM_{10}$  pollution at the traffic sites in Bac Giang province. The mean of  $PM_{10}$  at traffic sites ( $103.4 - 577.8 \mu\text{g}/\text{m}^3$ ) in this study was much higher than the daily average concentrations found in Navarra, Spain ( $17 - 54 \mu\text{g}/\text{m}^3$ ) [18], Altamira, Mexico ( $52.2 \mu\text{g}/\text{m}^3$ ) [17], Thessaloniki, Greece ( $42 - 97 \mu\text{g}/\text{m}^3$ ) [2], in Hanoi ( $87.1 - 108.5 \mu\text{g}/\text{m}^3$ ) [19, 32], in Bangkok ( $56.3 \mu\text{g}/\text{m}^3$ ) [19], Osaka, Japan ( $44.0 \mu\text{g}/\text{m}^3$ ) [33] and Ho Chi Minh city ( $31.77 \mu\text{g}/\text{m}^3$ ) [24]. However, the average of  $PM_{10}$  in the present study was lower than the 24-hour average concentration of  $529 \mu\text{g}/\text{m}^3$  in Pindingshan, China reported by Song [15].

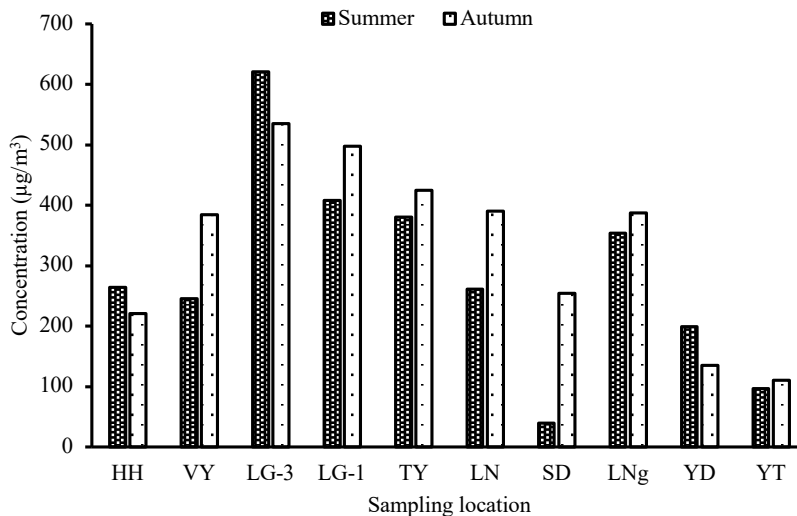


Figure 2. Seasonal variation of  $PM_{10}$  at the road sites

In this study, most of  $PM_{10}$  levels during autumn were higher than those in summer at the different sampling sites (Fig. 2). These results were similar to those observed at road sites in Spain [18, 34], Pakistan [35] and Argentina [36]. The variation of  $PM_{10}$  levels was affected by source emission, meteorological condition and condition of the monitoring sites [10, 23, 37]. In addition, Hien et al. (2002) [32] also reported the higher concentration of daily average  $PM_{10}$  during dry season ( $122 \mu\text{g}/\text{m}^3$ ) than that during wet season ( $47 \mu\text{g}/\text{m}^3$ ). The low  $PM_{10}$  levels observed in summer were possibly attributed to frequent wet precipitation affected by Asian monsoon. In addition, more stable atmosphere, higher probability of long range transport to Northern Vietnam by the North-East monsoon could lead to this result [19, 32].

#### 3.2. Levels and characteristics of trace elements associated with $PM_{10}$

10 metals (As, Cd, Co, Cr, Cu, Ni, Mn, V, Pb, Zn) in  $PM_{10}$  were analyzed in this study. As shown in Table 1, Ni, Pb and Zn presented the highest concentrations, accounting for 80.8% of total

obtained elements. The Cd and Co expressed lowest concentration which occupied 0.7% (Table 1). The level of detected elements was compared to the European Air Quality Directives for As, Cd, Pb and Ni (EU (DIRECTIVE 2004/107/EC)) [38]. The WHO standard was utilized to compare the levels of Cd, Cr (VI), Mn, Pb, V and Ni. The compared results indicated that the mean concentrations of Cd, Pb, V and Ni at road sites were closed to or lower than the EU guideline (5 ng/m<sup>3</sup> for Cd, 500 ng/m<sup>3</sup> for Pb and 20 ng/m<sup>3</sup> for Ni) and WHO standard (25 ng/m<sup>3</sup> for Ni, 1000 ng/m<sup>3</sup> for V and 500 ng/m<sup>3</sup> for Pb). However, the concentrations of Cd (from 0.4 ng/m<sup>3</sup> to 5.29 ng/m<sup>3</sup>) were from 4 to 53 times higher than the concentration limit for Cd (0.1 ng/m<sup>3</sup>) according to WHO standard. The average concentration of As (8.8 ng/m<sup>3</sup>) was 1.5 times higher than the concentration limit for As as stated by EU Directives (6 ng/m<sup>3</sup>). The mean concentration of Cr (31.80 ng/m<sup>3</sup>) was much higher than the concentration limit for Cr (VI) according to WHO standard (0.25 ng/m<sup>3</sup>). The average concentration of Mn (168.33 ng/m<sup>3</sup>) was 1.1 times higher than the target values of WHO (150 ng/m<sup>3</sup> for Mn). These results showed that the mean concentrations of Cd, Co, Cr, Cu, Mn, Ni, Pb, V and Zn at traffic 1 were from 1.2 to 2.7 times higher than those at traffic 2. Previous studies have showed that Pb and Zn in particulate matter originated from various sources such as exhaust emission from vehicles, wearing of auto brake pads, industrial processes and natural soil [5, 17, 20]. In this study, higher concentrations of Cu, Pb, Cr and Co in PM<sub>10</sub> at traffic 1 were probably attributed to high traffic density causing higher emission of Cu, Pb, Cr and Co due to processes of tires, brake wearing and diesel combustion [18, 39]. Moreover, the high level of Zn analyzed at traffic site, implying that the source of Zn might be related to the corrosion of galvanized vehicle parts [40]. The mean concentrations of As, Co, Cd, Cr, Mn, Pb, Ni at traffic sites were higher than the values obtained in Navarra, Spain [18], Altamira Mexico [17] and Ho Chi Minh city, Vietnam [24]. In contrast, the mean concentrations of Zn, Cu and Ni were lower than those at the other cities such as Thessaloniki, Greece [2], Osaka, Japan [33]. The average concentration of V, Cu and Cr in Naples, Italy [41] were higher than the measured values in our study.

Table 1. Mean concentrations of PM<sub>10</sub> trace metals at road sites  
(unit: ng/m<sup>3</sup>; except for the units of PM<sub>10</sub> with µg/m<sup>3</sup>)

Sampling point	PM <sub>10</sub>	As	Cd	Co	Cr	Cu	Mn	N	Pb	V	Zn
LG-3	577.8	17.91	2.06	2.88	78.51	79.83	119.31	12.60	112.50	19.76	142.00
LG-4	253.3	10.56	0.56	0.53	9.36	15.27	17.97	9.00	93.33	1.00	79.67
TY	402.7	3.06	0.63	0.56	21.10	12.10	27.33	8.09	14.00	7.41	91.67
LN	325.6	7.96	1.02	2.32	44.53	47.72	173.71	15.09	96.36	17.96	291.65
SD	146.9	5.65	0.94	1.62	16.92	15.38	47.04	10.78	133.96	4.30	53.00
LN <sub>g</sub>	370.8	13.39	4.99	2.54	26.72	24.15	275.31	8.12	153.83	15.53	187.83
VY	314.8	4.69	5.29	2.91	29.31	32.03	233.20	11.15	179.50	21.20	385.67
YD	167.2	4.66	0.40	3.58	17.77	30.29	186.27	9.82	138.35	20.17	150.10
YT	103.4	9.98	1.05	2.29	29.23	50.08	263.87	20.19	137.67	21.30	241.42
HH	242.1	10.13	2.60	2.54	44.55	102.14	339.31	17.51	150.64	13.93	231.44
VY	314.8	4.69	5.29	2.91	29.31	32.03	233.20	11.15	179.50	21.20	385.67
Average	292.6	8.79	1.95	2.18	31.80	40.90	110.82	12.23	121.01	8.55	185.44

As shown in Fig. 3, the concentrations of trace elements observed in autumn season were higher than those in summer season due to influence of meteorological condition in Vietnam. The higher levels in dry season were associated with the less precipitation, more stable atmosphere under the

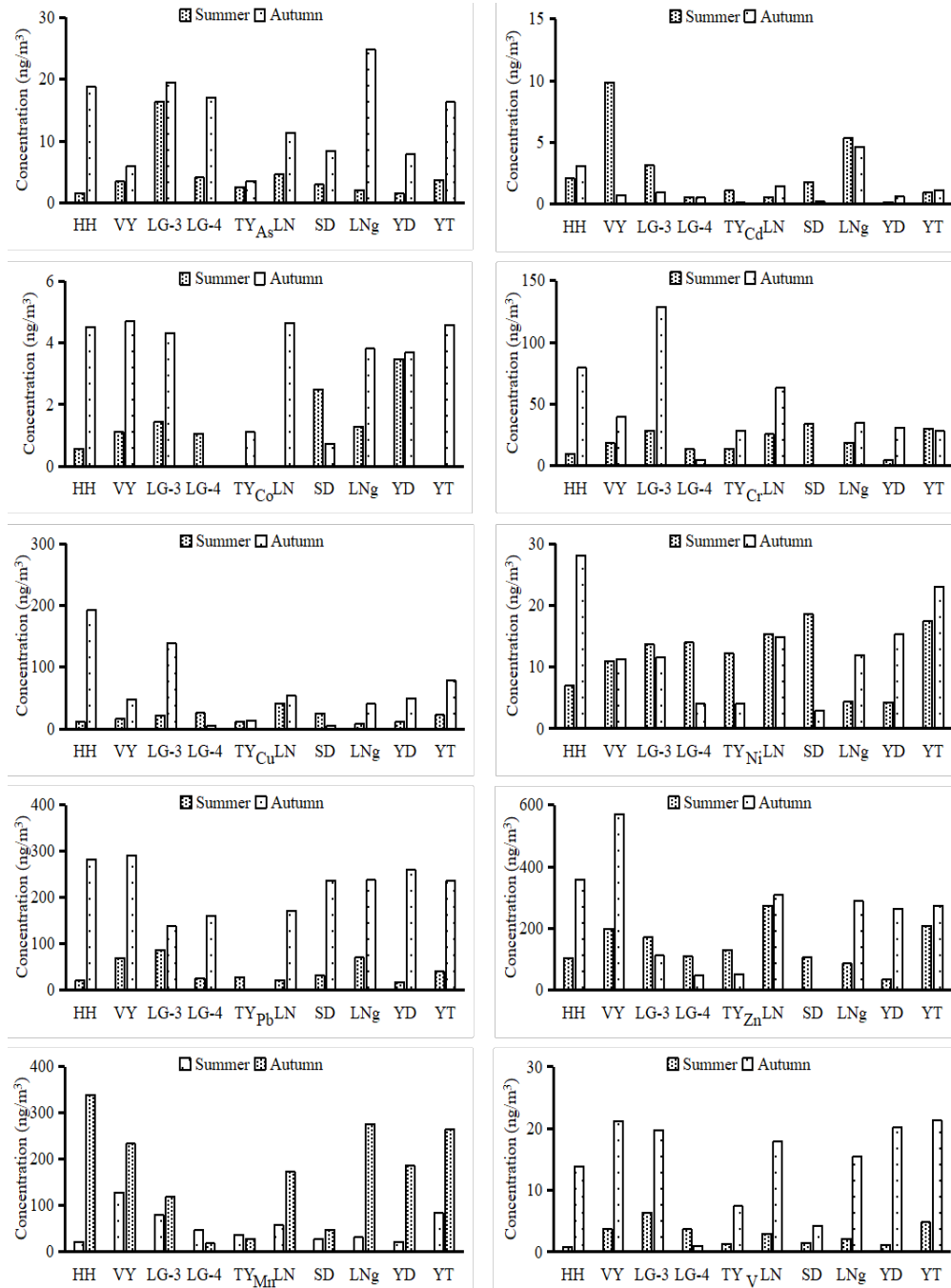


Figure 3. Seasonal variation of trace metals at the road sites

influence of a high-pressure ridge, higher probability of long range transport to Northern Vietnam by the North-East monsoon [19, 32]. In general, most of the metal concentrations observed in autumn were 1.2 to 3 times higher than those in summer. However, concentrations of As in autumn were 12 times and 13 times higher than those in summer at LN<sub>g</sub> and HH sites, respectively. Whereas, the concentrations of As at VY, LG-3, TY and LN sites in autumn were about twice than those in summer. The concentrations of Pb and V in autumn were observed to be from 14 times to 17 times higher than



those in summer at HH and YD. The concentrations of Pb at LN and SD in autumn were 8 times higher than those in summer. While, the concentrations of Cu, Co and Cr were 16 times and 8 times higher than those observed in summer at HH.

### 3.3. Pollution assessment of elemental metal

The contamination factors, degree of contamination and pollution load index were used to monitor the level of trace metals in the ambient air. Table 2 presented the contamination factors, degree of contamination and pollution load index for each element at road sites. The average  $C_F$  values of trace metals at traffic sites were low ( $C_F < 1$ ), moderate ( $1 \leq C_F < 3$ ) or considerable degree ( $3 \leq C_F < 6$ ), except for Cr at LG-3, Mn at LN, LN<sub>g</sub>, YT, VT, HH, VY and Pb at YD, HH, VY with contamination factor values larger than 6. The average  $C_d$  values at all traffic sites for the two seasons were classified into low contamination group (LG and TY), moderate contamination (SD), considerable contamination (LG, LN, LN<sub>g</sub>, VD, VT and VY), very high contamination (HH). The  $P_{Li}$  values ranged from 0.256 to 0.259 at LG, TY and SD were lower ( $C_d < 8$ ) than those at other sites, except for HH experiencing the  $P_{Li}$  values ranged from 1.105 to 1.490. The highest  $P_{Li}$  values at HH was 1.25, which was consistent with  $C_d$  values. These  $P_{Li}$  values suggested that air environment at most of sampling points in this study has been polluted, except for some points located at high land (LG, TY and SD).

Table 2. Contamination factor, degree of contamination and pollution load index obtained at traffic sites

Sampling point	Contamination factor (CF)										Degree ( $C_d$ )	$P_{Li}$
	As	Cd	Co	Cr	Cu	Mn	Ni	Pb	V	Zn		
LG-3	0.97	0.49	2.69	6.44	0.48	5.97	0.58	3.46	0.30	1.09	22.47	1.32
LG-4	0.85	0.27	0.00	0.25	0.02	0.90	0.20	4.03	0.02	0.48	7.00	0.26
TY	0.18	0.09	0.70	1.43	0.05	1.37	0.20	0.00	0.11	0.52	4.64	0.28
LN	0.57	0.72	2.90	3.17	0.19	8.69	0.74	4.30	0.28	3.00	24.56	1.32
SD	0.42	0.04	0.46	0.00	0.02	2.35	0.15	5.93	0.07	0.00	9.43	0.26
LN <sub>g</sub>	1.24	2.32	2.38	1.74	0.14	13.77	0.59	5.94	0.24	2.81	31.17	1.49
YD	0.39	0.31	2.30	1.55	0.17	9.31	0.77	6.49	0.31	2.57	24.18	1.11
VT	0.82	0.58	2.87	1.40	0.27	13.19	1.15	5.88	0.33	2.67	29.15	1.44
HH	0.94	1.56	2.82	3.97	0.66	16.97	1.40	7.03	0.21	3.49	39.05	2.06
VY	0.30	0.38	2.95	1.99	0.17	11.66	0.57	7.26	0.33	5.55	31.13	1.25

## 4. Conclusions

In this study, the PM<sub>10</sub> mass and trace metals bounded with PM<sub>10</sub> at the road sites in Bac Giang province were analyzed. The results indicated that the mean concentrations of PM<sub>10</sub> at most of road sites exceeded the allowable values of the national ambient air quality standard, except for two sampling sites (SD and YT) located in the mountainous area. Most of trace metals showed the higher levels during autumn than those during summer period. Ni, Pb and Zn presented the highest concentrations, accounting for 80.8% of total obtained elements, while Cd and Co expressed the lowest, occupying 0.7%. The average concentrations of Cd, Mn and Cr were higher than the target values of WHO. Meanwhile, the average concentration of As was higher than the concentration limit for As



from EU Directives ( $6 \text{ ng/m}^3$ ). These results showed that the mean concentrations of Cd, Co, Cr, Cu, Mn, Ni, Pb, V and Zn at traffic 1 were from 1.2 to 2.7 times higher than that at traffic 2. Higher Zn, Cu, Pb, Cr and Co concentrations in road sites could be resulted from processes of tires, brake wearing, corrosion of galvanized vehicle parts and diesel combustion. The assessment of pollution level indicated that most of  $P_{Li}$  values at road sites were higher than the benchmark level, implying that the ambient air around the studying site was being polluted. This finding pointed out the need to take the mitigation measures to reduce the air pollution in the study areas.

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