**RESEARCH ARTICLE** 



# Physico-chemical properties and transboundary transport of PM<sub>2.5</sub> in Bien Hoa City, Dong Nai Province, Southeastern Vietnam

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### Abstract

Air pollution is one of the most concerning environmental issues, wherein  $PM_{2.5}$  concentration plays an important role. This study monitored and evaluated the  $PM_{2.5}$  concentration trends in Bien Hoa City, Dong Nai Province, Vietnam. Twenty 24-h  $PM_{2.5}$  samples were continuously collected during the rainy (15 Oct. to 25 Oct. 2021) and dry (19 Mar. to 29 Mar. 2022) seasons. The  $PM_{2.5}$  samples were analyzed by scanning electron microscopy to determine the surface pattern and size distributions were analyzed using ImageJ software. The water-soluble fractions of 15 trace metal(oid)s concentrations (Al, Cu, Ni, K, Ca, Co, Mn, Cr, As, Zn, Pb, Cd, Na, Fe, and Mg) bound to  $PM_{2.5}$  were analyzed by inductively coupled plasma-mass spectrometry. The results showed that the 24-h fine fraction  $PM_{2.5}$  concentrations were  $24.1 \pm 12.2 \ \mu g/m^3$  and  $63.0 \pm 18.7 \ \mu g/m^3$  in the rainy and dry seasons, respectively. The results indicate that the size distributions of the particles of  $2.0-2.5 \ \mu m$  are minor, and the majority are ultrafine particles with aerodynamic diameter  $\leq 1.0 \ \mu m$ . Overall, the mass concentration level of the water-soluble fraction of trace metal(oid)s in  $PM_{2.5}$  in the rainy season was higher than that in the dry season. Among these, Ca, K, and Na were the most abundant earth crustal elements in  $PM_{2.5}$  in the rainy and dry seasons, accounting for 85% and 41.2% of the total trace element concentrations, respectively. The major sources of  $PM_{2.5}$  are local and regional sources of thermal power plants, industrial parks, and waterborne transportation (domestic rivers and marine). The activities undertaken to remove Agent Orange (e.g., soil excavation, transportation, and rotary kiln incinerators) at the Bien Hoa airbase area also cause increases in the  $PM_{2.5}$  level in the atmosphere of Bien Hoa City.

Keywords Air pollution  $\cdot$  Particulate matter  $\cdot$  PM<sub>2.5</sub>  $\cdot$  Soluble trace metals  $\cdot$  Vietnam

#### Highlights

This is the first study on physico-chemical properties of  $PM_{2.5}$  in Bien Hoa city.

The 24-h  $PM_{2.5}$  level in the dry season was higher than that in the rainy season.

Ultrafine particles are the dominant fraction of PM<sub>2.5</sub>.

Ca, K, and Na were the most abundant earth crustal elements in  $PM_{2.5}$ . Major sources of  $PM_{2.5}$ : industrial parks and waterbound transportation activities.

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# Introduction

Ambient air quality (AAQ) is assessed using the six most common air pollutants, including particulate matter ( $PM_{10}$ ) and PM<sub>2.5</sub>), carbon monoxide (CO), sulfur dioxide (SO<sub>2</sub>), nitrogen oxides (NOx), ozone (O<sub>3</sub>), and lead (Pb) (MONRE 2021; USEPA 2020). Among those, fine particulate matter  $(PM_{2.5}, with diameters \le 2.5 \ \mu m)$  is a significant criteria pollutant. PM<sub>25</sub> has been identified as one of the key global air pollutants in air quality monitoring with significant impacts on human health (Engel-Cox et al. 2013). These effects are attributed to the long-range transportation of PM<sub>2.5</sub> from the sources, their complex trajectory (Heinrich et al. 1998; Hien et al. 2022b), physico-chemical properties of particle compounds (Bashiri et al. 2017; Hien et al. 2022a; Le et al. 2013; Leal et al. 2007; Nan et al. 2018), and the meteorological and topographical conditions (Bashiri et al. 2017; Le et al. 2013, 2015; Thuy et al. 2021). PM<sub>2.5</sub> particles are formed from ordinary particulate matter or aerosol particles such as

carbon, sulfide, nitrogen, and other metal compounds suspended in the atmosphere (Hajime 2003; Thuy et al. 2021). In addition, organic compounds account for a major fraction (> 30%) of atmospheric particulate matter in urban areas (Jacobson et al. 2000).

Owing to their small diameters, particulate matter can penetrate deep into the lungs through the respiratory system, reach the heart, enter the blood stream, and cause serious effects on human health due to the physico-chemical characteristics of fine particles (Bashiri et al. 2017; Hien et al. 2022a; Huiming et al. 2013; Le et al. 2013, 2015; Thuy et al. 2020; USEPA 2020; WHO 2021; Xiangdong et al. 2019). The source and physical properties (size and shape) of PM25 have been studied to understand their impacts on the environment, climate, and human health. Recently, scientists have also discovered that the effects of aerosol particles are highly dependent on their particle size, and that the smaller particle sizes lead to larger impacts (Nan et al. 2018; USEPA 2020; WHO 2021). Additionally, particle size plays an important role in the deposition rate of particles and aerosols when inhaled into different regions of the respiratory system (Arhami et al. 2010). The World Health Organization (WHO) reported that approximately 4.2 million people worldwide die each year from exposure to polluted air (Eun et al. 2018).

To identify AAQ levels, a fixed automatic AAQ monitoring station (AAQMS) is required in the government's environmental management system. In Vietnam, AAQMSs are mostly located in major cities (e.g., Hanoi and Ho Chi Minh City, HCMC) with high installation, operation, and maintenance costs (Le et al. 2021; Thanh et al. 2018). However, to date, only Hanoi has had an AAQMS managed by the Vietnamese government, directly under the Ministry of Natural Resources and Environment (MONRE 2021). The AAQ level is typically indicated by the air quality index (AQI) value for the air pollution level. Recently, the AQI of Vietnam has been approximately 50–100 (moderate level, indicating that the air quality is acceptable; however, for some pollutants, a moderate health concern may exist), and it is sometimes higher than 150 (unhealthy for sensitive groups level, that is, members of sensitive groups may experience health effects) (VEA 2022). Research on AAQ has mostly been conducted in Hanoi and HCMC, which are the most populous areas in Vietnam (Hien et al. 2022b; Le et al. 2013, 2015; MONRE 2021; Thanh et al. 2018). Bien Hoa City (BHC), in Dong Nai Province, Southeastern Vietnam, has a high density of industrial and manufacturing firms. It is also near HCMC and Binh Duong, which are known to be the most polluted cities in southern Vietnam (Thao and Hung 2016). Thus, the AAQ level of BHC may be affected by the emission sources of those cities (Hien and Huy 2011; Hien et al. 2022b; Thao and Hung 2016). Presently, many projects have been undertaken to remove Agent Orange from the Bien Hoa airbase area, which also partly disperses dust and aerosols into the environment with large and small

particle sizes (USAID 2016). However, USAID (2016) also reported that no AAQ data, particularly at the fine particle level, were reported for BHC. Although increasing attention has recently been paid to the chemical composition of  $PM_{25}$ . comprehensive studies on the presence of trace metal(oid) s in urban atmospheric aerosols remain relatively scarce. PM<sub>2.5</sub> has been investigated because of its potentially harmful effects, increase in the AQI, and increase in serious haze events in Vietnam. Therefore, we investigated the seasonal variation of PM25 mass concentration during both the rainy season (RS) and dry season (DS) at BHC. The mass concentration, morphology, and trace metal(oid) concentration levels of PM25 were analyzed to understand their physico-chemical properties. The hybrid single-particle Lagrangian integrated trajectory (HYSPLIT) model was used to identify the main source of trace metals in BHC.

# **Materials and methods**

### Sampling site and description

BHC, a grade I urban center, is the political, economic, cultural, scientific, and technical center of Dong Nai Province. Samples of 24-h PM25 were collected at the Bien Hoa airbase (an area of approximately 1000 ha, located 10° 57' 22.6" N-106° 49' 06.2" E, approximately 30 km northeast of HCMC), in BHC, Dong Nai Province, Southeastern Vietnam (Fig. 1). In 2020, the population of BHC was approximately 1.1 million in an area of 26,362 ha (DNS 2021). The Bien Hoa airbase is situated on low-lying land, located 2.25 m above sea level. It is the main military airport in the southern region, acting as a base for training flights, combat readiness, and military transport (USAID 2016). However, the number of flights is small (10 flights/month), operating mainly in the east apron area, and no tall buildings are present (20 units of single flat-roof army guards for the offices and residing facilities for the soldiers, as reported by USAID (2016)), which is satisfactory for the location of air quality monitoring stations. The sampler equipment was fixed at the roof of the army guard with a height of approximately 10 m from the ground surface to the hood of the PM<sub>2.5</sub> sampler.

# Determination of PM<sub>2.5</sub> mass concentration

 $PM_{2.5}$  samples were collected using a high-volume air sampler (HVAS; Tisch Environmental Inc., USA) TE-1000 DBLX PUF polyurethane foam ambient air sampler (shelter box:  $48'' \times 20'' \times 20''$ , 74 lbs), which consists of a TE-1004 PUF blower motor that plugs into the TE-5010 motor voltage control. The HVAS was operated at a constant flow rate of 200 L/min. PM<sub>2.5</sub> was collected on a quartz fiber filter and substrates with a diameter of 102 mm to facilitate a more



Fig. 1 Map of the study area and location of sampling site: Vietnam (top-left), Dong Nai Province (bottom left), Bien Hoa City with the sampling site (right). This figure was generated using ArcGIS version 10.2 (https://desktop.arcgis.com/en/arcmap/)

complete physico-chemical characterization of PM<sub>2.5</sub>. The filter was pre-baked at 550 °C for 5-6 h to remove carbonaceous/organic contaminants before sample collection. It was conditioned at a temperature of  $20 \pm 5$  °C and relative humidity of  $40 \pm 10\%$  for approximately 24 h for gravimetric mass determination. After being pre-weighed, the filter was kept in a Petri dish and stored in an airtight bag for transfer from the laboratory to the sampling site to collect the sample. Each sample was collected for 24 h, starting at approximately 6:00 am to 6:00 am on the next day. After sampling, the sampled filter was placed back in the Petri dish and stored in an airtight bag and refrigerated. All samples were transported to the Department of Chemistry and Environment Laboratory (Vietnam-Russian Tropical Center, headquartered in Hanoi) for physico-chemical analyses. Each filter or mass concentration of PM<sub>2.5</sub> sample (including the filter) was determined by an analytical balance (XS205, Mettler Toledo, USA). The PM<sub>2.5</sub> mass concentration was calculated as Eq. 1:

$$C = \frac{M_2 - M_1}{V} \tag{1}$$

where C: daily (24-h)  $PM_{2.5}$  mass concentration ( $\mu g/m^3$ ),  $M_1$ : initial mass of conditioned filter before sample collection ( $\mu g$ ),  $M_2$ : final mass of the conditioned filter [filter +  $PM_{2.5}$ ] after sample collection ( $\mu g$ ), and V: total volume of air sampled taken directly from sampler ( $m^3$ ).

### Microscopy and chemical analyses

The morphology of PM<sub>2.5</sub> samples were analyzed using a field emission-scanning electron microscope (S-4800, Hitachi, Tokyo, Japan) at the Vietnam Academy of Science and Technology (VAST). Scanning electron microscopy (SEM) combined with ImageJ software provided information on the surface morphology and size distribution of ambient PM<sub>2.5</sub>.

In this study, we analyzed the water-soluble fraction of 15 trace metal(oid)s concentrations associated with PM2 5, including Al, Cu, Ni, K, Ca, Co, Mn, Cr, As, Zn, Pb, Cd, Na, Fe, and Mg. A detailed description of the experimental analysis has been provided in our previous study by Hien et al. (2022a). One-fourth of the filter samples were cut with ceramic-bladed scissors into small pieces for chemical extraction. The samples were mechanically shaken for 30 min with 25 mL of 0.01 M nitric acid (TraceSELECT<sup>™</sup>, for trace analysis, ≥69.0%, Honeywell Fluka<sup>TM</sup>) at pH 2, after which the extracted solutions were passed through 0.2 µm cellulose acetate filters (Minisart® Sartorius, Germany). Lab blank samples were run in parallel with each batch of sample extractions. Calibration solutions were prepared by diluting single-element laboratory fortifying stock solutions (SPEX CertiPrep, USA) in 0.01 M nitric acid. Iridium was chosen as an internal standard for trace metal analysis because it was not detected in the preliminary scans of the samples. One milliliter of iridium (100 ppb) was added to 9 mL of the samples, calibration standards, and certified reference materials before analysis by inductively coupled plasma-mass spectrometry (ICP-MS; iCAP-RQ, Thermo Scientific, USA). Polypropylene centrifuge tubes used for the extraction and storage of filtered extracts were pre-treated by soaking in 1.54 M HNO<sub>3</sub> for at least 48 h, rinsed with ultrapure water, and dried.

# Source and trajectory cluster analysis by HYSPLIT model

To identify the main sources of fine particle in BHC, backward trajectory analysis was conducted at the central site using the NOAA-HYSPLT model in an R environment (SplitR package). Accordingly, 96-h backward trajectory arrivals were calculated every 3 h at BHC at an arrival height of 100 m. Concentration-weighted trajectory (CWT) analysis was used to understand the potential sources of individual fine particle in this study. The logarithmic mean concentration of a pollutant species (or CWT) was calculated as follows (Eq. 2):

$$\ln(C_{ij}) = \frac{1}{\sum_{k=1}^{n} \tau_{ijk}} \ln(C_k) \tau_{ij}$$
<sup>(2)</sup>

where *i* and *j* are the parameters of a grid cell, *k* is the index of the trajectory, *n* is the total number of trajectories,  $C_k$  is the pollutant (PM<sub>2.5</sub>) concentration measured upon arrival of trajectory *k*, and  $\tau_{ijk}$  is the residence time of trajectory *k* in grid cell (*i*, *j*). The higher the value of  $C_{ij}$ , the higher the concentration at the receptor site when air parcels pass over grid cell (*i*, *j*) (Carslaw 2015). CWT analysis was conducted using the Openair package in the R software.

Trajectory analysis is commonly used to illustrate the transboundary transport routes of particulate matters. However, due to the uncertainties created by simplifying assumptions utilized in the single trajectory model, cluster analysis is introduced (Qor-el-aine et al. 2022). In this method, the number of clusters is determined by the user and the individual trajectories are ensembled to average out the errors. Based on that, the connections to the receptor may be recognized statistically. Thus, HYSPLIT trajectory cluster analysis is employed in this study to aid in establishing the knowledge of the effects of the sources to the study location.

# **Results and discussion**

## Seasonal mass concentration of PM<sub>2.5</sub>

Table 1 provides the ID samples, sampling dates, and the 24-h PM<sub>2.5</sub> mass concentration measured at BHC. In total, 20 of 24-h PM<sub>2.5</sub> samples were collected in two campaigns, including 10 continuous samples for each season (RS: 15 Oct. to 25 Oct. 2021; and DS: 19 Mar. to 29 Mar. 2022). For convenience in data storage and reporting, the collected samples' ID were denoted in turn with the letters representing the sampling site (Bien Hoa airbase, BHC) as BH; the third letter represented the RS and DS as R and D, respectively; the last numeric notation n (with n = 1-10) represented the order of the observed sample. For example, BHR8: sample observed (at BHC, during the RS, and sample number 8; additional information is presented in Table 1; this sample was collected during 22–23 Oct. 2021 (Friday)).

The 24-h fine fraction PM<sub>2.5</sub> concentration ranged from 15.2  $\mu$ g/m<sup>3</sup> to 50.4  $\mu$ g/m<sup>3</sup> with a mean value of 24.1  $\mu$ g/m<sup>3</sup>, and from 35.8  $\mu$ g/m<sup>3</sup> to 98.7  $\mu$ g/m<sup>3</sup> with a mean value of  $63.0 \,\mu\text{g/m}^3$  in the RS and DS, respectively (Table 1). The highest and lowest fine particle concentrations were found on the third day of the DS campaign (BHD3 = 98.7  $\mu$ g/m<sup>3</sup>) and on the last day of the RS campaign (BHR10 =  $15.2 \mu g/$ m<sup>3</sup>), respectively. As mentioned in "Introduction", no data on PM25 in BHC or Dong Nai Province have been reported. The PM<sub>2.5</sub> concentration level in HCMC, a neighboring city, was collected for comparison. During the RS, the PM<sub>25</sub> concentration level at BHC is lower than that at HCMC, as reported by Huy et al. (2018) (data from 2013 to 2017:  $28.0 \pm 18.1 \, \mu \text{g/m}^3$ ), Hien et al. (2019) (data from Mar. 2017 to Mar. 2018: 24-h mean, 33.6 µg/  $m^{3}$ ), and significantly lower than the value reported by Giang and Oanh (2014) (data from Dec. 2007 to Jan. 2008:  $97 \pm 31 \ \mu g/m^3$ ). However, during the DS, the PM<sub>2.5</sub> concentration level in BHC is higher than its level in HCMC  $(39.2 \ \mu g/m^3, 24-h \ mean)$  (Hien et al. 2019). In long-term (2013–2017) statistics and analysis, Huy et al. (2018) showed that the 24-h PM2.5 concentration level in HCMC reached 100  $\mu$ g/m<sup>3</sup> on some days.

In the RS, the lowest PM2 5 concentration value (BHR-10 sample,  $15.2 \,\mu\text{g/m}^3$ ) is similar to that in the WHO guidelines (24-h mean, 15  $\mu$ g/m<sup>3</sup>), but does not exceed (24-h mean,  $50 \ \mu\text{g/m}^3$ ) of the QCVN 05:2013/BTNMT in the Vietnam National Ambient Air Quality Standard (NAAQS) (Fig. 2a). However, the 24-h PM<sub>2.5</sub> concentration values during the DS campaign were higher than the WHO guideline value, and a remarkable 80% daily PM2.5 level was higher than the NAAQS level (Fig. 2b). Notably, during the RS campaign (Mar. 2021), the restrictions with the COVID-19 pandemic were largely in effect with the 5 K message: face masks, disinfection, distance, no gathering, and health declaration (respectively, in Vietnamese: Khâu trang, Khử khuẩn, Khoảng cách, Không tụ tập, and Khai báo y tế); discos, bars, karaoke, pubs, beer clubs, and sports facilities were to be continued in enclosed spaces (such as gym, fitness, billiards, and yoga); and the religious activities, ceremonies at religious establishments may be held but not gather more than 50 people at a time. This may have led to limited activities for the emission of air pollutants. However, during the second campaign (in the DS), the "new normal conditions" is an

ID sample	Date	PM <sub>2.5</sub> concentra- tion	Statistical values	
Rain season				
BHR1	15-16 Oct. 2021 (Friday)	32.0	Range (15.2—50.4) Mean±SD (24.1±12.2)	
BHR2	16-17 Oct. 2021 (Saturday)	26.7		
BHR3	17-18 Oct. 2021 (Sunday)	21.5		
BHR4	18-19 Oct. 2021 (Monday)	19.1		
BHR5	19-20 Oct. 2021 (Tuesday)	21.1		
BHR6	20-21 Oct. 2021 (Wednesday)	18.4		
BHR7	21-22 Oct. 2021 (Thursday)	47.6		
BHR8	22-23 Oct. 2021 (Friday)	50.4		
BHR9	23-24 Oct. 2021 (Saturday)	30.4		
BHR10	24-25 Oct. 2021 (Sunday)	15.2		
Dry season				
BHD1	19-20 Mar. 2022 (Saturday)	57.4	Range (35.8—98.7) Mean±SD (63.0±18.7)	
BHD2	20-21 Mar. 2022 (Sunday)	35.8		
BHD3	21-22 Mar. 2022 (Monday)	98.7		
BHD4	22-23 Mar. 2022 (Tuesday)	85.2		
BHD5	23–24 Mar. 2022 (Wednesday)	70.6		
BHD6	24-25 Mar. 2022 (Thursday)	77.7		
BHD7	25-26 Mar. 2022 (Friday)	63.7		
BHD8	26–27 Mar. 2022 (Saturday)	56.4		
BHD9	27-28 Mar. 2022 (Sunday)	44.2		
BHD10	28-29 Mar. 2022 (Monday)	62.3		

important concept of normal life without coronavirus, that is, in dealing with the COVID-19 pandemic. Most business and technology activities are reoperations; hence, all sources of air pollutants are reemitted, which would increase the contribution of  $PM_{2.5}$  to the AAQ level.

As shown in Table 1, the trend of the PM<sub>25</sub> concentration level on weekends was similarly lower than that on weekdays in both the RS (BHR2, BHR3, BHR9, and BHR10) and DS (BHD1, BHD2, BHD8, and BHD9). Statistically, the mass concentrations (mean ± standard deviation) of PM2.5 at weekends and weekdays during the RS were  $(24.1 \pm 6.6 \,\mu\text{g/m}^3)$  and  $(26.5 \pm 14.5 \,\mu\text{g/m}^3)$ m<sup>3</sup>) during the RS, respectively, and  $(50.3 \pm 10.4 \ \mu g/m^3)$  and  $(74.2 \pm 13.9 \ \mu\text{g/m}^3)$  during the DS, respectively. Herein, the variation in the mean PM25 concentration level on weekdays and weekends in the RS (2.4  $\mu$ g/m<sup>3</sup>) was tenfold lower than that in the DS (23.9  $\mu$ g/m<sup>3</sup>). The correlation between the 24-h PM<sub>25</sub> mass concentration collected during the RS and DS was  $R^2 = 0.15$ (Fig. S1). This low value indicates that PM<sub>2.5</sub> is not highly related in either season. This is attributed to direct factors such as the COVID-19 pandemic, weather conditions, and indirect factors such as the reoperation of business and industrial sectors. In addition, they were separated into three groups (G1, G2, and G3), with most of them distributed in group G1 (Fig. S1). In the G1 cluster, the mass concentration in the DS was high, whereas that during, the RS was low. This is explained by the seasonal variation of the 24-h PM<sub>2.5</sub> mass concentration.

Weather conditions, such as temperature, precipitation (Figs. 2c and d), wind speed, and wind direction (Figs. 2e and f), are one of the main factors that may affect the AAQ level. The sampling data from the meteorological station (located at Bien Hoa airport, see Fig. 1) recorded that the temperature during the RS ranged from 22.9 °C to 35.2 °C (Fig. 2c). In this sampling campaign, the weather was normally sunny in the daytime, but light rain occurred at night. The prevailing wind directions were north, northeast, north-northeast, and westsouthwest (Fig. 2e). During the DS, the temperature variation level (23.3–38 °C) was not significantly higher than the range compared with the temperature range that was monitored during the RS (Fig. 2c). However, the main wind directions were stable at an angle of north to north-northeast and northwest to north-northwest (Fig. 2f). Therefore, AAQ is affected by many local emission sources from industrial parks in BHC and Dong Nai Province (Fig. S2), such as AMATA Industrial Park, AGTEX Long Binh Industrial Park, Bien Hoa I Industrial Park, Bien Hoa II Industrial Park, and Tam Phuoc Industrial Park. These are large industrial parks and may emit high concentrations of particulate matter to the atmosphere under the "new normal conditions." The meteorological condition database also shows that the incidence of air mass may transport particles from emission sources of neighboring provinces which are located in the Mekong River Delta (MRD) area (Fig. S3) to be loaded at the local AAQ level. Particulate matter sources



**Fig. 2** Level of  $PM_{2.5}(\mathbf{a}, \mathbf{b})$  and weather conditions (temperature and precipitation ( $\mathbf{c}, \mathbf{d}$ ); wind rose ( $\mathbf{e}, \mathbf{f}$ )) in rainy season and dry season at Bien Hoa City, respectively. This figure was generated using OriginPro 2022 (https://www.originlab.com)

from MRD areas with a high density of industrial zones and transportation activities, such as HCMC and Binh Duong, have been reported by Thao and Hung (2016). In 2021, HCMC had 19 manufacturing and industrial zones, 30 industrial clusters, 3 thermal power plants, numerous other factories, a large volume of household cooking using deference fuels (coal and biomass), and a high density of traffic vehicles (Bang et al. 2021; Hien et al. 2022a, 2022b). Emissions from inland waterborne transportation activities in the Mekong River and domestic ports in the MRD area are considered important sources of  $PM_{2.5}$ , affecting the AAQ of surrounding areas (Khue et al. 2019; Thanh et al. 2018). Thus, the high  $PM_{2.5}$  levels during the DS at BHC may be due to the mixing of higher local and regional traffic flow and the emissions from the industrial and

commercial areas after the COVID-19 lockdown in terms of "new normal conditions" in Vietnam.

#### Morphology of PM<sub>2.5</sub> and its chemical composition

# Morphology characteristics of PM<sub>2.5</sub>

Figure 3(a-f) show the SEM micrographs of PM<sub>2.5</sub> at BHC during the RS and DS, respectively. The particulate matter had a high density, was evenly distributed on the surface of the filter paper, and interspersed with the fibrous structures of the quartz filter paper (Fig. 3a, b, d, and e). The surface structure when photographing samples at higher magnifications

(Fig. 3c and f) showed that the predominance of ultrafine particles (with a kinetic diameter  $\leq 1.0 \,\mu$ m) may be predicted with a significantly high percentage. This characteristic was evident, as shown in Figs. 3c and f, for the samples obtained after SEM images were taken and processed using ImageJ software. During the RS, the particle size distribution results (Fig. 3g) showed that although PM<sub>2.5</sub> samples were collected, the particle size with kinetic diameter ranging 2.0–2.5 µm accounted for an extremely small proportion. The proportion was not high even for a particle size of 1.0–2.0 µm. However, the distribution of ultrafine particles was dominant in the samples. Initially, the PM<sub>2.5</sub> content in this area was found to be mainly contributed by the sources dispersed far from the sampling site. Furthermore, ultrafine particles were dominant during the DS and mostly distributed in the particles with diameters of  $0.2-1.0 \ \mu m$  (Fig. 3h). Ultra-fine dust particles can disperse far from the emission source and move to the study area, affecting its AAQ (Hien et al. 2022b).

#### **Elemental concentrations**

Figure 4 presents the heat maps showing the concentrations of elements in  $PM_{2.5}$  for the RS (Fig. 4a) and DS (Fig. 4b). The summary statistics were listed in Table 2. These elements can be divided into two major groups: anthropogenic tracers (Cr, Cd, Ni, Cu, Pb, Zn, As, and Co) and earth crust elements (or soil tracers: Na, Al, K, Mg, Ca, Fe, Ti, and Mn). For the seasonal mean concentrations, the sequence of the water-soluble fraction of 15 trace metal(oid)s concentration levels in  $PM_{2.5}$  during the RS was Ca>K>Na>Al>Zn>Mg>Fe>Cu>C r>Pb>Mn>Ni>As>Cd>Co and that for the DS was Na >Ca>Zn>Al>Mg>Cu>Fe>K>Cr>Pb>Mn>Ni>Cd >As>Co. Overall, the trend of mass concentration of trace

metal(oid)s in  $PM_{2.5}$  was similar in both the RS and DS. However, the mass concentration of the water-soluble fraction of trace metal(oid)s in  $PM_{2.5}$  in the RS was higher than that in the DS (Fig. 4). Ca, K, and Na were the most abundant earth crustal elements in  $PM_{2.5}$ , accounting for 85% and 41.2% of the total trace element concentrations in  $PM_{2.5}$  for the RS and DS, respectively. This group of elements may originate from regional sources (e.g., wind erosion and wind-blowing soil dust) and local sources (e.g., transportation as well as activities to remove Agent Orange such as soil excavation, transportation, and rotary kiln incinerators) at the Bien Hoa airbase area (USAID (2016)). Thus, fine particle loading increased in the atmosphere at BHC. The mass concentration of the five metals (Mn, Ni, As, Cd, and Co) in the RS and DS was the lowest at 0.28% and 1.4% for the RS and DS, respectively.

The water-soluble fractions of metals (Cu, Al, Zn, Mg, and Fe) are found in  $PM_{2.5}$ , which suggests that coal burning (for thermal power plants), brake/tire wear, and steel smelting are significant pollution sources (Hien et al. 2022a). Al, Fe, and Zn, which are major components of the upper continental crust (Rudnick and Gao 2003), are also found in high concentrations in the atmosphere of populous Hanoi and HCMC (Cohen et al. 2010; Gataria et al. 2005; Giang and Oanh 2014; McNeill et al. 2020). The presence of these elements imply that soil dust is the dominant source of  $PM_{2.5}$  affecting the AAQ at BHC. In the industrial sector, high-temperature processes, such as metal smelting and fuel combustion, are usually the source of non-crustal volatile metals (e.g., Cu, Cd, Zn, and Fe) in the atmosphere.

Figure 5 illustrates the daily concentrations of selected trace metals (Al, Cu, Ni, Cr, As, Zn, Pb, Cd, and Fe) in  $PM_{2.5}$  during the two sampling periods of the RS and DS. The concentrations of Cu, Cr, As, and Pb during the DS



**Fig. 3** SEM micrographs of  $PM_{2.5}$  at different magnifications (*a*,*d*×10.000, *b*,*e*×20.000, and *c*,*f*×50.000): (**a**, **b**, **c**) in the rainy season and (**d**, **e**, **f**) in the dry season; and particle size distribution (**g**) in the rainy season and (**h**) in the dry season

Fig. 4 Heat maps showing the concentration of elements in  $PM_{2.5}$  during the (a) rainy season and (b) dry season. This figure was generated using OriginPro 2022 (https://www. originlab.com)



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Element	Rainy season (15–25 Oct. 2021, $n = 10$ )		Dry season (18–29 Mar. 2022, $n = 10$ )	
	Range	Mean $\pm$ st. dev	Range	Mean $\pm$ st. dev
Al	0.19-0.08	$0.46 \pm 0.22$	0.41-0.82	$0.55 \pm 0.14$
Cu	0.02-0.23	$0.11 \pm 0.07$	0.12-0.55	$0.39 \pm 0.17$
Ni	0.001-0.05	$0.007 \pm 0.01$	0.01-0.32	$0.02 \pm 0.09$
Κ	1.19-4.99	$3.05 \pm 1.26$	0.12-0.63	$0.37 \pm 0.13$
Ca	2.23-7.41	$5.25 \pm 1.83$	0.17-1.07	$0.77 \pm 0.25$
Co	0.0001-0.0008	$0.0003 \pm 0.0002$	0.0001-0.0028	$0.0004 \pm 0.0009$
Mn	0.01-0.03	$0.03 \pm 0.07$	0.02-0.06	$0.03 \pm 0.01$
Cr	0.03-0.13	$0.06 \pm 0.03$	0.16-0.6	$0.30 \pm 0.14$
As	0.0003-0.0015	$0.001 \pm 0.0003$	0.0002-0.0082	$0.005 \pm 0.003$
Zn	0.06-0.64	$0.45 \pm 0.21$	0.5-0.79	$0.67 \pm 0.09$
Pb	0.004-0.06	$0.04 \pm 0.02$	0.01-0.18	$0.07 \pm 0.05$
Cd	0.0003-0.0015	$0.0009 \pm 0.0004$	0.001-0.3	$0.006 \pm 0.09$
Na	1.1-5.56	$2.73 \pm 1.42$	0.37-1.38	$0.85 \pm 0.28$
Fe	0.25-0.66	$0.37 \pm 0.13$	0.13-0.69	$0.37 \pm 0.16$
Mg	0.02-0.48	$0.38 \pm 0.15$	0.13-0.89	$0.44 \pm 0.25$

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**Table 2** Summary statistics (mean  $\pm$  standard deviation and range concentrations; unit:  $\mu g/m^3$ ) of the water-soluble trace metal(oid)s concentrations in PM<sub>2.5</sub> at Bien Hoa City during the dry and rainy season sampling campaigns

(Mar. 2022) were significantly higher than those during the RS (Oct. 2021). The activities of the business and industrial sectors are resurrecting after the COVID-19 lockdown. The high concentration levels of selected trace metals suggested that both regional and local sources are major contributors to the  $PM_{2.5}$  distribution (Hien et al. 2022a) in the AAQ of BHC. Many of the selected trace metals (e.g., Fe, Mn, Pb, and Cr) are associated with waterborne transportation, mostly from the long coastal regions to the northeast of the city (Fig. S3). The dominant sources of Cd and As at BHC are evidently associated with transport from the northeast. The potential sources of trace metals are heavy industrial

activities such as mining, non-ferrous metal refining, thermal power plants (coal-fired), and maritime activities in the surrounding region. This is described in detail in "Influence of transboundary transportation of  $PM_{2.5}$ ".

### Influence of transboundary transportation of PM<sub>2.5</sub>

The back-trajectory concentrations of  $PM_{2.5}$  using the CWT approach for the RS and DS are illustrated in Fig. 6a and b, respectively. The regions in red correspond to the main contributing sources associated with the highest mean  $PM_{2.5}$  concentrations, which were 39 µg/m<sup>3</sup> and 57 µg/m<sup>3</sup> during the RS and DS,

**Fig. 5** Daily variation of selected trace metals in PM<sub>2.5</sub> at Bien Hoa City: (**a**) in the rainy season and (**b**) in the dry season. This figure was generated using OriginPro 2022 (https://www.originlab.com)



respectively. For the RS, the highest CWT values covering the map were distributed from southwest Indonesia and affected the AAQ in BHC. In particular, the wind direction from the northeast may transport the  $PM_{2.5}$  emitted by vessels in the Indian Ocean to BHC. In contrast, during the DS, the CWT shows the effects of  $PM_{2.5}$  concentration from the sea located in the southeast area including the Philippines, Brunei, and Malaysia.

Five clusters of  $PM_{2.5}$  concentrations are shown in Fig. S4. Overall, the air mass of the clusters started at a location in the coastal zones of South Asian countries. The air masses of Cluster C4 were the major polluted trajectories, accounting for 34.5% and 37.5% of  $PM_{2.5}$  in BHC during the RS and DS, respectively. In the RS, clusters C1 and C5 (both 20.7%) were from China and the Indian Ocean that contributed to **Fig. 6** Concentration-weighted trajectory (CWT) plots for PM<sub>2.5</sub>, using concentration data and 3-d air mass back trajectories for the Bien Hoa City site (black dot) in (**a**) the rainy season (Oct. 2021) and (**b**) the dry season (Mar. 2022) at Bien Hoa City. This figure was generated using the NOAA-HYSPLIT model (https://www.ready.noaa.gov/HYSPLIT.php)



the PM<sub>2.5</sub> affecting the AAQ in BHC. The air masses associated with cluster C2 (13.8%) started from southern Laos and passed through the center of Cambodia, with average CWT values of approximately 38–40  $\mu$ g/m<sup>3</sup>. During transboundary transportation, the wind may transport PM<sub>2.5</sub> emitted from local sources to the sampling site at BHC.

Several trace metals, such as Mn, Cr, Cd, As, and Pb, are associated with emissions from coal combustion (Jozef and Elisabeth 2001; McNeill et al. 2020), within three thermal power plants in southern Vietnam (Hien et al. 2022a). All the thermal power plants using coal contribute to the main fuel combustion in the southeastern provinces (Guizhou, Guangxi, and Hunan) of China, which is a potential source of PM longrange transportation (Cohen et al. 2010). This is consistent with the results of NOAA-HYSPLIT (Figs. 6 and S4) and accounts for the transport along the main wind direction from the north and northeast (Figs. 2e and f) to the sampling site. Zn concentration levels were high in both RS and DS, which could originate from multiple sources related to vehicle emissions, tire tread (Smolders and Degryse 2002), or from Znenriched emissions during the combustion of lubricating oil, diesel soot, and oil industries (Hien et al. 2022a), as indicated by the analysis of  $PM_{2.5}$ . Herein, emissions from industrial parks and marine transportation are important sources of  $PM_{2.5}$  affecting the AAQ in BHC. This finding is consistent with the suggestion by Hien et al. (2022a) for a neighboring city, HCM, that shipping activities affect the AAQ level.

# Conclusions

This study is the first to evaluate the physico-chemical properties of fine particle matter  $(PM_{2.5})$  in BHC, known as an Agent Orange pollution area and located in Dong Nai Province, Southern Vietnam. Twenty 24-h PM<sub>2.5</sub> samples

were continuously collected during the rainy season (15 Oct. to 25 Oct. 2021) and the dry season (19 Mar. to 29 Mar. 2022) in BHC. The 24-h PM<sub>2.5</sub> mass concentrations were  $24.1 \pm 12.2 \ \mu g/m^3$  and  $63.0 \pm 18.7 \ \mu g/m^3$  during the RS and DS, respectively. The majority were ultrafine particles with an aerodynamic diameter  $\leq 1.0 \ \mu m$  in the cluster of PM<sub>2.5</sub>. The mass concentration of the water-soluble fraction of 15 trace metal(oid)s in PM<sub>2.5</sub> in the RS was higher than that in the DS. Additionally, Ca, K, and Na were the most abundant earth crustal elements in PM2.5 during the rainy and dry seasons, accounting for 85% and 41.2% of the total trace element concentrations, respectively. The water-soluble concentrations of Mn, Ni, As, Cd, and Co in the RS and DS were 0.28% and 1.4%, respectively. The results suggest that the source emissions from local (industrial parks) and a combination of local and regional (thermal power plants and domestic river and marine transportation activities) are the major sources of PM<sub>2.5</sub> that strongly affect the AAQ in BHC.

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### Declarations

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