Trace elemental composition and source identification of $PM_{2.5}$ in the metropolitan area of Bangkok, Thailand

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Abstract. The trace elements in $PM_{2.5}$ samples collected in Pathum Thani province, the metropolitan area of Bangkok were determined and their possible sources were analyzed. $PM_{2.5}$ samples were monitored from January to December in 2022. The mass concentration of $PM_{2.5}$ ranged from 4.33 to 66.02 μg/m^3 with and an average (± std.) value of 24.64 ± 13.97 μg/m^3. The concentrations of trace elements (Na, Mg, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Zn, Ni and Cu) were measured using accelerated based ion beam analysis (IBA) methods, particle-induced X-ray emission (PIXE). Elements such as K, Ca, Fe, S, Si and Cr were found to be the dominant elements in $PM_{2.5}$ across all seasons. The Pearson correlation coefficients between Fe-Cr, Ni-Cr, Fe-Ni, Mn-Cr, Fe-Mn and Ni-Mn were all above 0.893 with $p < 0.01$. The enrichment factor indicated that S, Cl, V, Cr, Mn, Fe, Zn, Ni and Cu came mainly from anthropogenic emissions; Ca, Ti and Fe were generated from natural and anthropogenic sources; Na, Mg, Al and Si originated from natural sources. These findings suggested that biomass burning, industrial and soil dust emissions were the major sources of $PM_{2.5}$ in the study area.

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1 Introduction

Haze pollution with high loading of fine particulate matter ($PM_{2.5}$) has been recognized as the largest environmental problem in Thailand and has received extensive attention from the government. Bangkok is not the only city in Thailand that annually faces serious $PM_{2.5}$ air pollution problems; there are many other cities that are recently encounter similar problems. Pathum Thani is one of the provinces surrounding the Bangkok Metropolitan Area (BMA) that is confronted with the problem of $PM_{2.5}$ pollution during winter (November – February). Traffic (44%), biomass burning (24%), sea spay (11%), power plants (7%) and industrials (4%) have been identified as sources of fine particles emitted in BMA [1]. There were approximately 10,395,256.917 acres of rice field, cultivated by more than 2,450 rice farmers in this Province [2] and normally, farmers plant 2-3 crops per year. During winter season, the rice paddies are burned. Intensive rice filed and crop residues burning resulted in the $PM_{2.5}$ emissions in the area and transported towards Bangkok by the northeast monsoon winds [3]. In addition, this province has several industrial parks including metal processing, aluminum die casting, steel wires, electronic power equipment etc.

Trace elements are persistent and widespread in the environment, and can interact with natural components to form hazardous $PM_{2.5}$. They can be emitted from natural and human activities. For example, Al, Ca, and Mg are normally associated with natural soil dust emissions, whereas Ni, Cu, Mn, Cr and Zn are associated with anthropogenic sources such as combustion of fossil fuels, industrial activities, vehicle exhaust and waste incinerations [4]. Trace elements could remain in the air with relatively long residence time and enter to human tissues and internal organs. They can induce cancer in humans in the form of lung, liver, kidney, and nose cancer via inhalation and ingestion [5].

This study aimed to investigate the concentration of trace elements in $PM_{2.5}$ samples in Pathum Thani Province, the Bangkok metropolitan area, Thailand and their seasonal variations and possible sources. The output can provide better understanding of $PM_{2.5}$ sources in Bangkok and its metropolitan area. This can support stakeholders and policymakers to design an action plan for reducing high $PM_{2.5}$ loadings and improving public health and the environment.

2 Experimental design

2.1 Sampling site and sample collection

$PM_{2.5}$ samples were collected at Thailand Institute of Nuclear Technology, Khlong Luang district, Pathum Thani Province, Thailand (14.24°N, 100.43°E) at ground level (2 m above ground) in the year 2022. The map of the sampling site is shown in Fig. 1. Pathum Thani is north of Bangkok and is part of the Bangkok metropolitan area. This area possesses 67% rice fields of the total land of the province and normally, farmers in this site cultivate 3-4 crops of rice per year. There is agricultural waste produced in this area includes: rice straw, sugarcane leaves, bamboo trees and corn cobs, coconut husks, and other pelleted tree debris [6]. In addition, this site is surrounded by more than 3,400 industrial factories, including electronics, metal processing, automotive parts, plastics and textiles. One of the major industrial parks is 20 km away from the sampling site. A main road is also located about 5 km south.

$PM_{2.5}$ samples were conducted during January to mid-February 2022, mid-February to April 2022 and October to December 2022, representing winter at the beginning of the year, summer and winter at the end of the year, respectively. Samples were collected on a 46.2 mm in diameter Teflon® filter using low-volume air sampler (Tisch TE-Wilbur) according to the

Federal Reference Method (40 CFR, Part 53, 50 Appendix L). Sampling was performed over 24 h intervals every second day at a flow rate of 16.67 L/min. The collection duration of each sample started at 9.00 am local time each day and ended at 9:00 am the following day. The total number of samples was 86.

Fig. 1. Map of Thailand (left) and expanded view of sample area (right) showing the site of Khlong Luang, Pathum Thani [7,8].

2.2 Gravimetric analysis

The $PM_{2.5}$ Teflon filters were conditioned in a desiccator for 24 h under a constant temperature (25 ± 5 °C), and humidity (50 ± 5%) to maintain an equilibrium condition before and after sampling and were weighed using a 5-digit analytical balance (Ohaus Adventure). The mass concentrations of $PM_{2.5}$ (μg/m^3) were calculated by subtracting the pre-collection weight post-collection weight and dividing by the volume of total air sampling (m^3).

2.3 Elemental analysis

The collected $PM_{2.5}$ samples were analyzed for their elemental composition using PIXE. These PIXE measurements were performed using a 2 MeV proton beam generated by a 1.7 MeV tandem accelerator at the Department of Physics and Materials Science, Faculty of Science, Chiang Mai University, Thailand. A lithium-drifted silicon detector, [Si(Li)] was used to detect X-ray emission. Parameters for PIXE analysis used in this study have been given elsewhere [9]. The resulting PIXE spectra were analyzed with the GUPIX software. The calibration of the PIXE system was performed using Micro Matter thin film standards. The uncertainty of the PIXE measurement was between 5% to 15%.

2.4 Data processing

Pearson correlation analysis was performed using the ORIGIN 8 software for analyzing the relationship between $PM_{2.5}$ and trace elements, as well as individual trace elements in $PM_{2.5}$. To identify the origin of the trace elements presented in the $PM_{2.5}$ samples, either from natural (crustal) sources or anthropogenic sources, the enrichment factor (EF) was employed using

the Microsoft Excel 2021 software. The EF was used to determine the enrichment or depletion of a specific element in a sample compared to its natural abundance in the crust [10] and can be calculated using Eq. (1):

$$EF = \frac{(X_{PM2.5}/Al_{PM25})}{(X_{crust}/Al_{crust})} \quad (1)$$

where $X_{PM2.5}/Al_{PM2.5}$ and X_{crust}/Al_{crust} are the concentration ratios between the target element and the reference element in $PM_{2.5}$ and in the Earth's crust, respectively. In this study, the element Al was used as the reference element [11] and the elemental composition of the upper continental crust was taken from [11]. Three contamination categories of EF were recognized as follows [12]: EF less than 10 indicated the element mainly originates from natural sources (minimally enriched), while EF between 10 and 100 suggested mixed origin of the elements with contribution of both anthropogenic and natural sources (moderately enriched), and EF more than 100 was considered that the element was greatly enriched with major anthropogenic sources.

3 Results and discussion

The time series of $PM_{2.5}$ in the study area during January to December, 2022 are shown in Fig.2. The average mass concentrations of $PM_{2.5}$ in winter at the beginning of the year (January to mid-February), summer (mid-February to April) and winter at the end of the year (October to December) were 33.96 ± 13.43 μg/m^3, 23.56 ± 14.80 μg/m^3 and 20.74 ± 11.48 μg/m^3, respectively, which were lower than the national ambient air quality standard the (Thailand NAAQS) of 37.5 μg/m^3, the United States Environmental Protection Agency (US-EPA) of 35 μg/m^3 on approximately 92% of the days, but exceeded the World Health Organization (WHO) guideline of 15 μg/m^3. The maximum $PM_{2.5}$ concentrations were recorded 66.02 μg/m^3, 65.95 μg/m^3, 51.81μg/m^3 and 52.23 μg/m^3 on 5 February, 17 February, 9 April and 3 December, respectively, which may be the effects of open burning of rice straw in the field around the site. The intensive open field burning of rice straw peaks during two periods: January to April and November to December [13].

Fig. 3 shows the Box-whisker plots of elemental concentrations of PM2.5 in the different seasons. The results showed that $PM_{2.5}$ samples possessed high concentrations of K, Ca, Fe, S, Si and Cr for all three study seasons. The average concentrations of elements were found in the descending order of K > Ca > Fe > S > Si > Cr for the winter at the beginning of the year and their average concentrations were 177.19 ± 148.03 ng/m^3, 94.25 ± 95.57 ng/m^3, 82.01 ± 95.24 ng/m^3, 42.15 ± 27.36 ng/m^3, 31.74 ± 20.11 ng/m^3 and 28.90 ± 42.12 ng/m^3, respectively. For the summer, the descending order of K > Fe > Ca > S > Cr > Si were obtained with the concentrations of 71.81 ± 62.25 ng/m^3, 31.62 ± 53.33 ng/m^3, 28.52 ± 26.10 ng/m^3, 24.17 ± 17.66 ng/m^3, 10.48 ± 19.26 ng/m^3 and 8.24 ± 8.20 ng/m^3, respectively. While in winter at the end of the year, the descending of Fe > K > Ca > Cr > S > Si were observed and their concentrations were 93.30 ± 239.84 ng/m^3, 53.83 ± 41.06 ng/m^3, 48.67 ± 68.63 ng/m^3, 39.26 ± 103.96 ng/m^3, 27.76 ± 25.22 ng/m^3 and 18.53 ± 12.79 ng/m^3, respectively. It can be noted that the concentrations of all dominant elements in the winter at the beginning of the year were significantly higher than those found in both summer and winter at the end of the year. Other trace elements, namely Cl, Na, Mg, Al, Zn, Ni, Cu, Mn, Ti and V were also observed in the $PM_{2.5}$ samples. The high concentration of K can be attributed to the emissions from local biomass burning; Ca, Si, Mg, Al, Na and Ti were assigned to crustal sources; S, Zn, V, Mn, Fe and Cu may originate from industrial emissions (S from coal

combustion; Zn, V, Mn, Fe, and Cu from metallurgical production); Ni, Mn and Cr were linked to brake wear and tires, and road dust resuspension.

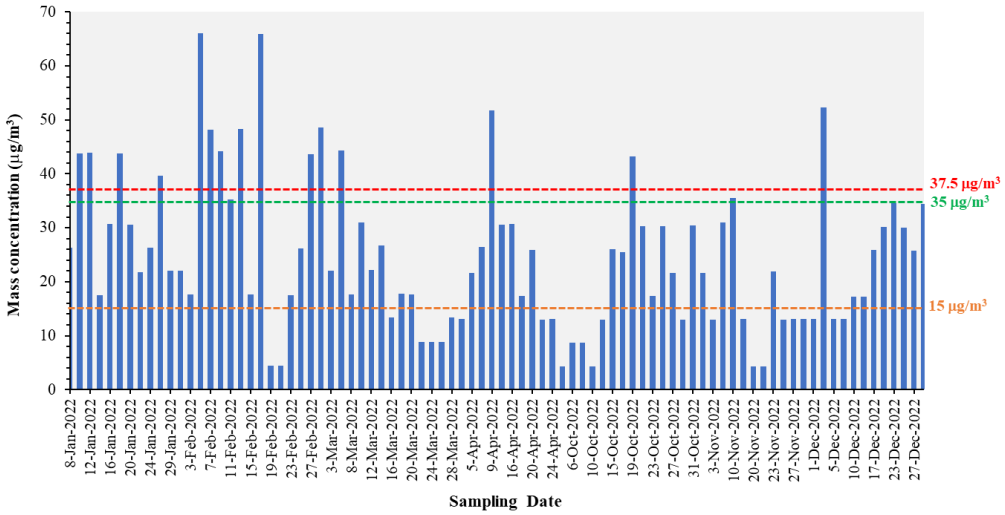

Fig. 2. Time series of $PM_{2.5}$ in Pathum Thani during 2022. The horizontal dashed lines represent the National Ambient Quality Standards (Thailand NAAQS) limit at 37.5 µg/m³, the United States Environmental Protection Agency (US-EPA) limit at 35 µg/m³ and the World Health Organization (WHO) limit at 15 µg/m³.

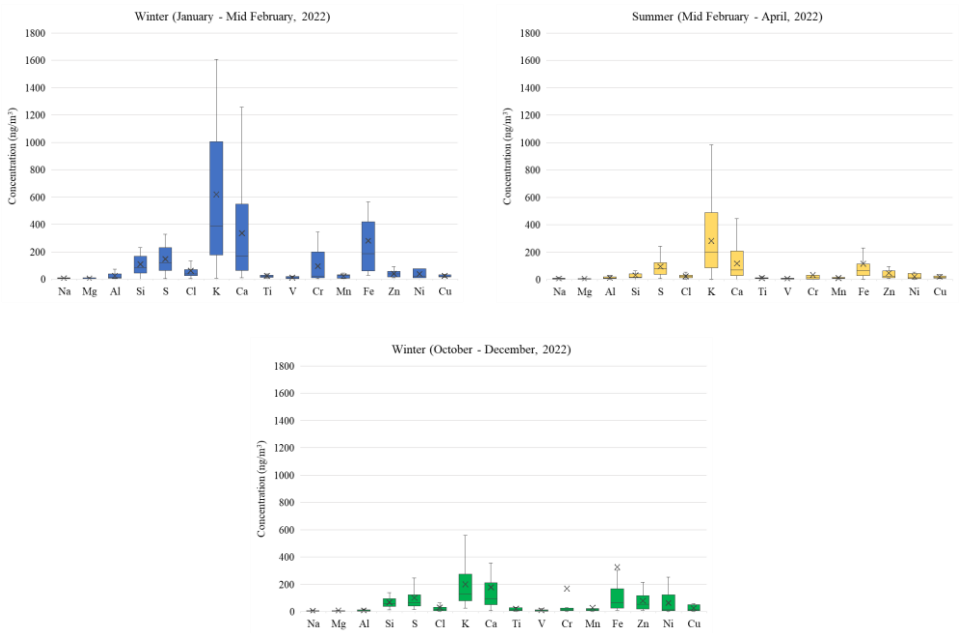

Fig. 3. Box-whisker plots of elemental concentrations of $PM_{2.5}$ in the different seasons of the year 2022.

As can be seen in Table 1, the strongest correlations were between Fe-Cr (r = 0.981), Ni-Cr (r = 0.972), Fe-Ni (r = 0.969), Mn-Cr (r = 0.949) and Fe-Mn (r = 0.919), which indicated that these elemental associations might derive from industrial and vehicle emission sources. A good correlation involving industrial emission sources such as S-Si (r = 0.789), Mg-V (r = 0.706), and Al-V (r = 0.700) was also observed. Ni showed a strong correlation with Mn (r = 0.893), implying that they might come from vehicle emission sources. There were significant correlations between Mg-Na (r = 0.855), Mg-Ti (r = 0.692), Si-Ca (0.644) and Mn-Ti (r = 0.711), which could be a contribution from crustal sources. In addition, a correlation between K-Si (r = 0.760), K-S (r = 0.690), K-Ca (r = 0.704), Cl-Ca (r= 0.704) and Cl-Mg (r = 0.854) was detected, suggesting a possible contribution from the biomass burning sources.

Table 1. Pearson's correlation coefficients between mass and trace elemental concentrations in $PM_{2.5}$.

	$PM_{2.5}$	Na	Mg	Al	Si	S	Cl	K	Ca	Ti	V	Cr	Mn	Fe	Zn	Ni	Cu
$PM_{2.5}$	1																
Na	-0.134	1															
Mg	-0.231	0.855	1														
Al	0.080	0.332	0.568	1													
Si	0.167	0.174	0.374	0.530	1												
S	0.224	0.109	0.275	0.338	0.789	1											
Cl	0.166	0.146	0.854	0.540	0.538	0.474	1										
K	0.339	0.323	0.105	0.438	0.760	0.690	0.467	1									
Ca	0.204	0.220	0.131	0.480	0.644	0.499	0.319	0.704	1								
Ti	-0.039	0.177	0.692	0.504	0.439	0.280	0.432	0.314	0.537	1							
V	0.281	0.214	0.706	0.700	0.459	0.089	0.139	0.360	0.483	0.652	1						
Cr	0.097	0.207	0.242	0.268	0.108	-0.046	-0.005	0.031	0.177	0.636	0.591	1					
Mn	0.095	0.312	0.434	0.566	0.147	0.050	0.094	0.121	0.184	0.711	0.637	0.949	1				
Fe	0.082	0.317	0.014	0.296	0.174	0.070	0.015	0.166	0.339	0.567	0.626	0.981	0.919	1			
Zn	-0.003	0.277	0.075	0.063	0.109	0.185	0.106	0.262	0.536	0.341	0.279	0.049	0.103	0.241	1		
Ni	0.185	0.392	-0.097	0.042	-0.053	-0.143	-0.185	-0.112	0.181	0.397	0.512	0.972	0.893	0.969	0.074	1	
Cu	0.472	0.666	-0.082	-0.098	-0.059	0.056	-0.042	0.125	0.099	-0.033	-0.666	0.354	0.636	0.324	0.562	0.490	1

Shaded correlation coefficients are significant at the 0.01 level.

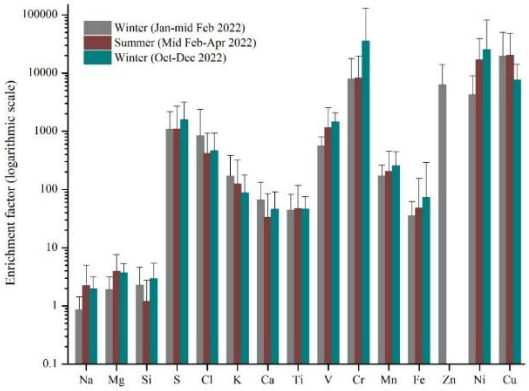

Fig. 4. Enrichment factor of trace elements in $PM_{2.5}$.

In this study, EF was used to assess the degree of anthropogenic contribution of elements in $PM_{2.5}$ and the results are shown in Fig.4. The uncertainty of the EF for each element in each season showed the variability of EF values across different $PM_{2.5}$ samples collected during that season. As shown in Fig. 4, the elements with mean EF > 100 were S, Cl, K, V, Cr, Mn, Zn, Ni and Cu, indicating their strong anthropogenic origin for almost all study seasons, except for the EF value of K in winter at the end of the year. S, Cl, V, Zn, and Cu can be attributed to industrial emission sources (e.g. coal combustion, ferrous and non-ferrous, metal pressing, steel production), which might originate from the large industrial zone situated approximately 20 km northwest of the monitoring site. Ni, Mn and Cr came from vehicle emissions, which probably were associated with two main roads situated approximately 5 km north and south of the sampling site. K is usually considered as a marker of biomass burning, which might link to the open burning of agricultural waste surrounding the sampling sites, especially rice stubble and straw. Ca, Ti and Fe were intermediately enriched (10 < EF < 100), and K was lower than 100 during winter at the end of the year, suggesting probably attributed to both natural and anthropogenic sources. Ca can be linked with building construction and cement processing, which were attributed to massive housing developments being underway in Pathum Thani province, driven by the overflow of Bangkok's population. Ti, K and Fe are commonly recognized as elements emitted from crustal sources. EF values of Na, Mg, Al and Si were found to be less enriched (EF < 10). These elements are typically crustal species.

4 Conclusions

In this study, the trace elements and their sources of $PM_{2.5}$ were comprehensively investigated at Pathum Thani Province, the metropolitan area of Bangkok during January to December 2022. The results showed that the average concentration of $PM_{2.5}$ in winter at the beginning of the year (33.96 ± 13.43 μg/m^3) was higher than that in summer (23.56 ± 14.80 μg/m^3) and winter at the end of the year (20.74 ± 11.48 μg/m^3). K, Ca, Fe, S, Si and Cr were mainly in $PM_{2.5}$ samples. K was found to be higher in winter at the beginning of the year and summer, whereas Fe was the most abundant element in winter at the end of the year. Pearson correlation coefficient showed a strong association between Fe-Cr, Ni-Cr, Fe-Ni, Mn-Cr, Fe-Mn and Ni-Mn ($r > 0.893$), suggesting possible contribution from industrial sources. EF analysis revealed that S, Cl, K, V, Cr, Mn, Zn, Ni and Cu were highly enriched, indicating the substantial contribution of anthropogenic sources; i.e industrial, vehicle emission sources.

A moderate enrichment of Ca, Ti and Fe was identified, implying a portion of these elements probably comes from crustal and anthropogenic sources. The elements Na, Mg, Al and Si possessed minimal enrichment, revealing originated mainly from crustal sources. In conclusion, biomass burning, industrial and soil dust were probably the most significant sources of $PM_{2.5}$ pollutant in Pathum Thani Province. The local authorities should support formers for recycling and upcycling of agricultural wastes and encourage the industrial sector to use advanced technologies in controlling air pollution.

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