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Epidemiological Study on Health Risk Assessment of Exposure to PM_{2.5}-Bound Toxic Metals in the Industrial Metropolitan of Rayong, Thailand

Sawaeng Kawichai ^{1,2} , Susira Bootdee ^{3,*} , Sopittaporn Sillapapiromsuk ⁴ and Radshadaporn Janta ⁵ 

- ¹ Research Institute for Health Sciences (RIHES), Chiang Mai University, Chiang Mai 50200, Thailand
 - ² Environmental-Occupational Health Sciences and Non Communicable Diseases Research Group (EOHS and NCD Research Group), Research Institute for Health Sciences (RIHES), Chiang Mai University, Chiang Mai 50200, Thailand
 - ³ Chemical Industrial Process and Environment Program, Faculty of Science, Energy and Environment, King Mongkut's University of Technology North Bangkok (Rayong Campus), Rayong 21120, Thailand
 - ⁴ Department of Environmental Science and Technology, Faculty of Science, Lampang Rajabhat University, Lampang 52100, Thailand
 - ⁵ Atmospheric Research Unit, National Astronomical Research Institute of Thailand, Chiang Mai 53000, Thailand
- * Correspondence: susira.b@sciee.kmutnb.ac.th; Tel.: +66-38627000 (ext. 5704)

Abstract: The concentrations of PM_{2.5} and metallic elements were measured in Rayong during the dry season (November 2021 to April 2022). The mean PM_{2.5} concentration was $20.1 \pm 10.9 \mu\text{g}/\text{m}^3$ (4.9–52.3 $\mu\text{g}/\text{m}^3$). Moreover, the percentages of days when those PM_{2.5} concentrations exceeded the daily WHO and US-EPA NAAQS limit were 56.8% and 10.2%, respectively. However, the levels did not exceed 50 $\mu\text{g}/\text{m}^3$, which is the limit of the 24 h standard defined by the PCD in Thailand. The dominant heavy metals and elements in PM_{2.5} samples were Cr, Cu, Fe, Mn, Pb, V, and Zn, which constituted 70%. In Rayong, the PCA results showed that industrial emissions (Cd, Cu, Fe, Mn, Pb, and Zn) and traffic emissions (As, Cd, Cr, K, and Ni) were the major sources of PM_{2.5}-bound heavy metals. Exposure to toxic metals in PM_{2.5} through the inhalation pathway in Rayong obviously entails a high potential risk of cancer ($>10^{-4}$) based on the total lung cancer risk (TCR_{inh}). It was found that the TCR_{inh} values of Cr for combined age groups were higher than 10^{-6} , which implies a high cancer risk in Rayong.

Keywords: fine particulate matter (PM_{2.5}); health risk assessment; hazard quotient (HQ); individual lifetime cancer risk (CR_{inh})



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1. Introduction

The epidemiological research on the increasing mortality and morbidity risks connected to the inhalation of fine particulate matter (PM_{2.5}) has a wide range of results regarding impacts on people's health. PM_{2.5} is smaller in diameter than 2.5 μm , which allows it to penetrate into the cardiovascular system and respiratory system. The World Health Organization (WHO) reported that long-term exposure to PM_{2.5} has been linked to increased mortality from cardiovascular disease, respiratory disease, and lung cancer [1]. The International Agency for Research on Cancer (IARC) has classified particulate matter in group 1 of human carcinogens [2]. PM_{2.5} is released during the combustion of fossil fuels and biomass, and their size is affected by their organic and inorganic composition, which then affects their potential for toxicity [3–6]. According to Sakunkoo et al. [7], exposure to PM_{2.5}-bound heavy metals in residential and industrial zones of Khon Kean, Thailand, has been linked to an increased risk of cancer in children. An investigation of the non-carcinogenic risks of exposure to PM_{2.5}-bound elements and polycyclic aromatic hydrocarbons (PAHs) in Beijing between 2013 and 2015 was performed in China. The target

organs were those of the respiratory system, which showed that about 82% of the non-carcinogenic hazard values were greater than the United States Environmental Protection Agency's (US-EPA) allowable limits. Although people spend most of their time indoors, ambient PM_{2.5} sources can penetrate into residential buildings. Sublett [8] revealed that an air cleaner without a filter for heating, ventilation, and air conditioning (HVAC) removes less than 10% of smaller aerosols, which cause allergic respiratory diseases. Additionally, the effectiveness of air filtration systems for reducing personal exposure to indoor PM_{2.5} is limited, and this exposure has been linked to blood pressure. It was found that PM_{2.5} was reduced by 31–53% [9]. Currently, the duct-type electrostatic precipitators (ESPs), which remove more PM_{2.5} than air filtration systems, are used to improve indoor air quality [10]. On a side note, it was found that the arsenic (As) and hexavalent chromium (Cr(VI)) were present in PM_{2.5}, which are significant risk factors for cancer generated from the burning of fossil fuels [4]. Chang et al. [11] estimated PM_{2.5} emissions using an activity-based model. The Taiwanese Industrial Port of Kaohsiung's PM_{2.5} emissions resulted in an annual environmental cost of USD 2176.04 million and an external health cost of 3238.30 disability-adjusted life years (DALY), or 8.53% of the index of health impact (IHI) value in 2005–2017. As a result, it is clear that PM_{2.5} has an effect on both property and human health.

Long-term exposure to PM_{2.5} is attributed to morbidity and early death. Some PM_{2.5} components, especially heavy metals that penetrate the human body and accumulate in the organs and blood, can be seriously destructive at low concentrations [12]. Hsu et al. [13] reported that concentrations of manganese (Mn), chromium (Cr), and lead (Pb) in ambient PM_{2.5} pose potential carcinogenic risks in urban, sub-urban, rural, and industrial areas of Taiwan. Chromium and cadmium (Cd) are causes of cancer in both children and adults, and they are present in PM_{2.5} according to a health risk assessment in an industrial area of Chelyabinsk, Russia's South Ural Region [14]. Furthermore, Han et al. [15] found that industrial emissions associated with Cr posed risks that were 25% carcinogen-related and 36% non-carcinogen-related. Compare this to coal combustion, which was linked to Cr, As, and Mn and indicated 15.46% and 20.64%, respectively. These heavy metals are a result of the production of reactive oxygen and nitrogen species, which include free radicals such as the hydroxyl ($\bullet\text{HO}$) and superoxide ($\bullet\text{O}_2^-$) radicals, hydrogen peroxide (H_2O_2), nitric oxide (NO), and other endogenous oxidants. Similar DNA damage is caused by free radicals of iron (Fe), copper (Cu), nickel (Ni), Cr, and Cd, especially Ni, Cr, and Cd, which are linked to cancer [16].

In numerous studies of exposure to PM_{2.5}-bound elements, the lifetime cancer risk (CR) and hazard quotient (HQ) models showed increased morbidity and mortality from lung cancer and non-carcinogenic risks over time. They noted that a CR value for the inhalation pathway of more than 10^{-4} implied a high cancer risk, whereas an HQ value of more than 1.0 indicated a significant non-carcinogenic risk. Sakunkoo et al. [7] reported the estimation of lung cancer risk due to exposure to heavy metals in PM_{2.5} that was collected from Khon Kean province, Thailand. The mean CR values due to Cd in children and adults in a university area (3.34×10^{-2} and 8.10×10^{-8}), a residential area (0.23×10^1 and 4.40×10^{-1}), an industrial zone (0.12×10^1 and 2.28×10^{-1}), and an agricultural zone (4.11×10^{-2} and 7.70×10^{-3}) were found to be above 10^{-4} , indicating a high potential risk of lung cancer. Furthermore, they reported that children were particularly aware of the health risks caused by Cd in PM_{2.5} particles. The inhalation of PM_{2.5}-bound heavy metals in an industrial city of Iran was studied. They found the HQ values and non-carcinogenic risk levels for children and adults were associated with Cr. The CR values of Cd for children (1.19×10^{-4}) and adults (4.81×10^{-4}) indicate a high risk of developing cancer [17]. According to Hsu et al. [13], Mn, Cr, and Pb were the main heavy metals in PM_{2.5} released in Taiwan's urban, rural, sub-urban, and industrial areas, raising concerns about the risk to human health. Moreover, the high concentrations of Cr, Ni, and Pb in PM_{2.5} and the risk of cancer were linked with the CR values ($>10^{-6}$) of industrial areas. They observed that industrial zones release high concentrations of iron (Fe), zinc (Zn),

copper (Cu), and manganese (Mn), and traffic-related metals in PM_{2.5} included barium (Ba), chromium (Cr), nickel (Ni), molybdenum (Mo), and cobalt (Co). In the haze season in northern Thailand, Fe, potassium (K), Cr, Pb, and Zn were the most predominant PM_{2.5}-bound elements released from biomass burning and forest fires [18].

The air quality of industrial metropolitan regions is regarded as a major problem that poses a threat to human health. As a result, the primary goal of this research was to properly determine the concentrations of PM_{2.5} and heavy metals in PM_{2.5} samples in the industrial metropolitan of Rayong during the dry season. Secondly, we studied the non-carcinogenic and lung cancer risk levels posed by the determined PM_{2.5}-bound toxic metals.

2. Materials and Methods

2.1. Sampling Site

Rayong province is located in eastern Thailand. It is a coastal town and the principal producer of various fruits and seafood. Additionally, Rayong province is well known for having an industrial estate that has attracted significant investment, produced a large number of jobs, and stimulated significant immigration to the area. The Eastern Economic Corridor (EEC) innovation platform has chosen Rayong province to plan industrial zones in the future. Next-generation automotive technology, intelligent electronics, advanced agriculture and biotechnology, food for the future, high-value and medical tourism, automation and robotics, aviation, logistics, and digital technology are all developed in industrial estates. The EEC is a strategic plan derived from the royal Thai government's 20-year strategy to achieve high-income status by 2036, named "Thailand 4.0". The goals are to encourage investment in Thailand's industrial sector, increase the country's competitiveness, and allow for long-term growth. Under the management system of the initiative of the EEC area for the years 2017–2021, this will strengthen Thailand's position as the ASEAN economic hub [19]. As a consequence, sustainable natural resource management and a well-conserved environmental system are critical in Rayong province.

2.2. PM_{2.5} Sampling

PM_{2.5} samples were collected on the rooftop of the nine-story office of the President Building (KMUTNB, Rayong campus; 12°49'26.4792" N, 101°12'59.2416" E, 340 m MSL) at King Mongkut's University of Technology North Bangkok, Rayong campus, Rayong Province, from November 2021 to April 2022 (Figure 1). PM_{2.5} samples were collected from ambient air using a PQ 200 particulate matter sampler (Mesa Labs BGI Inc., Butler, NJ, USA) at 16.7 L/min on Teflon (PTFE) filters (2 µm pore size, Ø 47 mm; Measurement Technology Laboratories, Bloomington, MN, USA). The PM_{2.5} was collected on alternate days for 24 h. Then, the PTFE filters were stored in vacuum desiccators for 48 h before and after the sampling for 24 h; they were weighed using a microbalance (Mettler Toledo, Switzerland). Each filter was weighed three times in a controlled room (temperature, 25 ± 2 °C; relative humidity, 50 ± 5%). After that, all the filters were placed in the freezer (−20 °C) until elemental analysis. Filter blanks were collected in the same manner as samples using an unopened air sampler, which were used to correct subsequent analysis deviation and ensure its accuracy.

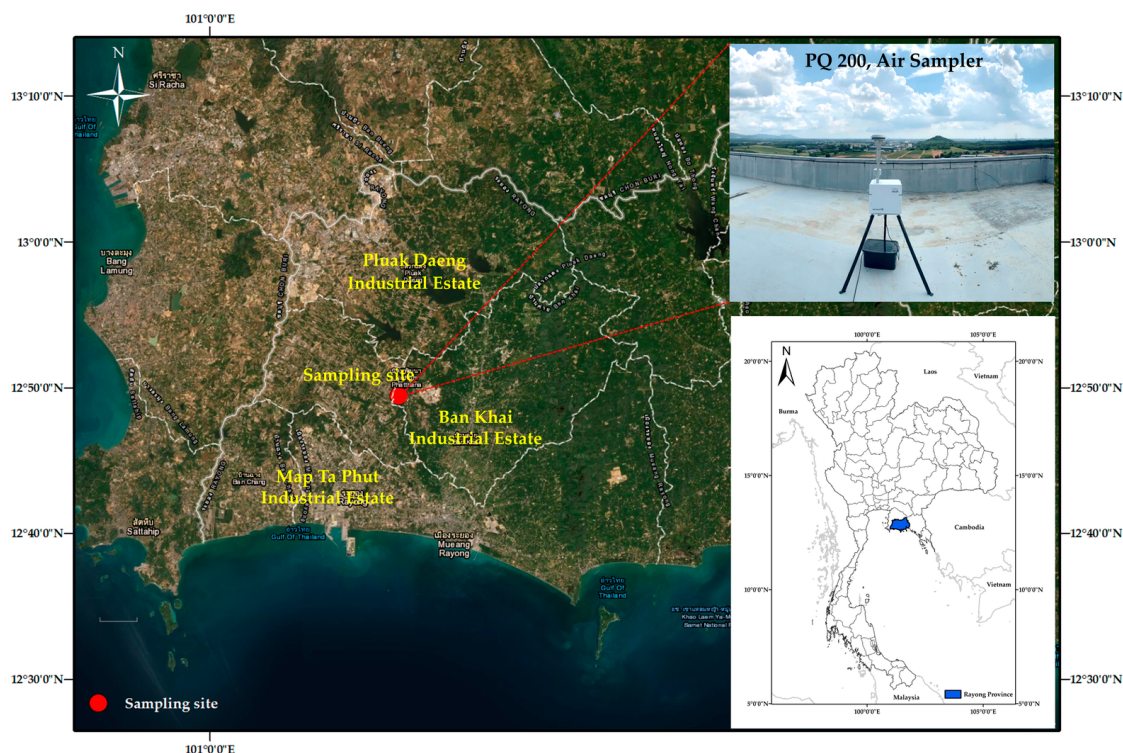


Figure 1. The study sites in Rayong, Thailand.

2.3. Extraction and Analysis of PM_{2.5}-Bound Elements

Following sampling, the filter samples were cut into small pieces and placed in Teflon vessels before being dissolved in mixtures of 9 mL of 65% nitric acid (HNO₃) and 1 mL of 30% hydrogen peroxide (H₂O₂) in a microwave oven (Milestone Ethos Up, Sorisole, Italy). All the digestion was performed in two steps over 35 min, with the temperature being raised to 190 °C at a rate of 9.5 °C/min (25 min) and held there for 20 min (Methods library for SK-15 ET high pressure rotor). Finally, the solution was filtered through a nylon syringe filter (0.45 µm, 13 mm) and diluted with 25 mL of deionized water. Additionally, after digestion preparation, eleven elements, including arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), potassium (K), manganese (Mn), nickel (Ni), lead (Pb), vanadium (V), and zinc (Zn), were analyzed using an inductively coupled plasma–optical-emission spectrometer (ICP-OES, Agilent 5800, Santa Clara, CA, USA).

The accuracy of the analysis and extraction conditions was verified using 50 mg of the National Institute for Environmental Studies (NIES) Certified Reference Material (CRM) Number 28 Urban Aerosols (Ibaraki, Japan) and the spiking method. Table S1 shows the element recoveries from seven replications with CRM and seven replications with the spiking methods. CRM element recoveries ranged from 67.1% (K) to 97.4% (Pb), and spiking method recoveries ranged from 79.3% (As) to 98.6% (V). The limit of detection for elements ranged from 0.005 to 0.141 µg/m³, and the limit of quantitation was 0.017–0.470 µg/m³, as shown in Table S2.

2.4. Morphology of PM_{2.5}

The morphology and elemental composition of ambient aerosols are important indicators for determining the sources of PM. To identify potential sources, scanning electron microscopy combined with an energy dispersive spectrometer (SEM-EDS) provides detailed information on aerosol particle size, chemical (elemental) composition, density, origin of the particles, and surface morphology [3,20–22]. The PM_{2.5} samples were randomly collected on quartz filters (47 mm, Whatman, Hangzhou, China) in a PQ200 air sampler and analyzed for elemental ingredients using SEM-EDS (JEOL JSM-5910LV scanning electron microscope, Tokyo, Japan). A 1 mm² section of each selected filter was cut with cutters and

raised to an aluminum SEM stub for probing. Each sample was coated with a very thin layer of gold using a vacuum coating unit, also known as a gold sputter coater.

2.5. Source Identification of PM_{2.5} Using Principal Component Analysis (PCA)

Principal component analysis (PCA), a multivariate statistical technique, has been widely used to find possible association sources of variance in the atmospheric sciences [15,17]. PCA is a type of multivariate statistical analysis that groups data into comparable groups if there is a relationship among them. Therefore, PCA was used to determine the possible sources of ambient PM_{2.5} and heavy metals in PM_{2.5} collected during the dry season in Rayong. By clearly explaining the source characteristics of each heavy metal in PM_{2.5}, this study improved the analysis and monitoring of heavy metal pollution emissions in the study area, and the health of the local population.

2.6. Statistical Analysis

The data were statistically analyzed using a one-way ANOVA to determine the mean difference in PM_{2.5} between the months. To achieve a normal distribution, the log-transformed PM_{2.5} concentrations were used. Spearman's rho correlation coefficient analysis was used to assess the relations between the meteorological parameter and the PM_{2.5} concentrations. The meteorological parameter was received from the Thai Meteorological Department (TMD), as shown in Table S3. The meteorological factors were total precipitation (Prec.), relative humidity in the atmosphere (RH), wind speed (WS), and ambient temperature (Temp.).

2.7. Health Risk Assessment for Heavy Metals

2.7.1. Non-Carcinogenic Risk Assessment

The hazard quotient (HQ) was used in this study to calculate a health risk assessment for non-carcinogenic heavy metals in PM_{2.5} exposure. The hazard quotient (HQ) is a numerical ratio of a single substance's exposure level during a particular period to a reference dose for that substance derived from a similar exposure period (WHO, 2005). Non-carcinogenic long-term effects in organisms are caused by issues other than cancer. The HQ was computed from the ratio of the exposure concentration through inhalation (EC_{inh}) to the reference concentration (RfC) of each pollutant using Equation (1). RfC is the reference concentration (mg/m^3) of a heavy metal in ambient air from the Regional Screening Level (RSL) Summary Table in 2022 [23,24], as shown in Table 4.

$$HQ = \frac{EC_{inh}}{RfC \times 1000 \mu g/mg} \quad (1)$$

When the $HQ > 1.0$, the occurrence of chronic effects is considered probable, and $HQ < 1.0$ indicates no significant risk of chronic effects [7,15,25]. Equation (2) can be used to compute the EC_{inh} through inhalation.

$$EC_{inh} = \frac{C \times E_i \times ET \times EF \times ED}{AT_n} \quad (2)$$

where EC_{inh} is the exposure concentration through inhalation of a given heavy metal [15]; C is the concentration of the heavy metal ($\mu g/m^3$); E_i is the deposition fraction of PM_{2.5} that can pass through the lungs ($E_i = 0.48$); ET is the exposure time ($ET = 24$ h/day); EF is the exposure frequency ($EF = 365$ days/year); ED is the exposure duration ($ED = 2$ years for baby (0–2 years), 14 years for children (2–15 years), and 55 years for adults (16–70 years)—refer to Greene and Morris et al. [26]); and AT_n is the average time ($AT_n = ED \times 365$ days/year $\times 24$ h/day for non-carcinogenic risk and $AT_n = 70$ years $\times 365$ days/year $\times 24$ h/day for carcinogenic risk).

The hazard index (HI) was also used to determine the total non-carcinogenic risk from exposure to many contaminants at the same time. The HI value is the sum of the HQ of the heavy metals in a mixture. It is calculated by Equation (3).

$$HI = \sum HQ = HQ_1 + HQ_2 + \dots + HQ_n \quad (3)$$

HI values less than 1.0 suggest that the pollutant under evaluation is unlikely to have a health impact, and HI values greater than 1.0 indicate that the pollutant will probably affect people's health [27,28].

2.7.2. Lung Cancer Risks

The individual lifetime cancer risk (CR_{inh}) is the potential for an individual to develop lung cancer as a result of lifetime exposure to carcinogenic metals, and it was estimated with Equation (4) [15,26]. According to the CR_{inh} categories, an CR_{inh} value $<10^{-6}$ is considered negligible, whereas a CR_{inh} level $>10^{-4}$ is high risk. An acceptable or tolerable risk is represented by CR_{inh} 10^{-6} to 10^{-4} [5,24].

$$CR_{inh} = EC_{inh} \times IUR \quad (4)$$

where IUR is the inhalation unit risk (per $\mu\text{g}/\text{m}^3$), which was obtained from the RSL Summary Table [23], as shown in Table 5.

However, this is not the actual lifetime risk. Intake (inhalation rate), metabolism, and absorption rates cause major variations in dose and exposure of chemical concentration in the air. The US-EPA [29] recommended that carcinogens that have a mutagenic mode of action should be the focus, using age-dependent potency adjustment factors (ADAFs). Before 2 years of age, there is a 10-fold ADAF, whereas an age of exposure between 2 and 15 years means a 3-fold ADAF. For ages above 15 years, there is no adjustment (1-fold ADAF). As shown in Equation (5), the ADAFs must be used to accurately assess the lifetime risks for age groups.

$$CR_{age\ groups} = CR_{inh} \times \left(ADAFs \times \frac{ED}{70\ years} \right) \quad (5)$$

The total lung cancer risk (TCR_{ic}) (for a population with an average life expectancy of 70 years) was combined across all the age groups (Equation (6)).

$$TCR_{inh} = \sum CR_{inh} = CR_{baby} + CR_{children} + CR_{adults} \quad (6)$$

3. Results and Discussion

3.1. PM_{2.5} Concentrations

The average concentration of PM_{2.5} in the dry season was $20.1 \pm 10.9 \mu\text{g}/\text{m}^3$ ($4.9\text{--}52.3 \mu\text{g}/\text{m}^3$) (Table 1). The levels did not exceed $50 \mu\text{g}/\text{m}^3$ for the 24 h standard defined by the Ambient Air Quality Standard of the Pollution Control Department (PCD) in Thailand. Furthermore, the percentages of days exceeding the daily WHO limit of $15 \mu\text{g}/\text{m}^3$ and the US-EPA National Air Quality Standards (NAAQs) of $35 \mu\text{g}/\text{m}^3$ were 56.8% (50 days) and 10.2% (9 days), respectively. However, the daily mean PM_{2.5} in Rayong, Thailand ($20.1 \pm 10.9 \mu\text{g}/\text{m}^3$) was determined to not exceed the standards of Malaysia ($35 \mu\text{g}/\text{m}^3$), Singapore ($37.5 \mu\text{g}/\text{m}^3$), Vietnam, the Philippines, Sri Lanka ($50 \mu\text{g}/\text{m}^3$), Indonesia ($65 \mu\text{g}/\text{m}^3$), and China ($75 \mu\text{g}/\text{m}^3$) [30,31]. Aside from that, the daily PM_{2.5} concentrations ($4.9\text{--}52.3 \mu\text{g}/\text{m}^3$) found in this study were lower than those found in ambient air in the industrial zone in Khon Kaen province, Thailand ($35.1\text{--}56.4 \mu\text{g}/\text{m}^3$) [7]. Previous studies of ambient PM_{2.5} concentrations in urban industrial areas [5,15,32,33] revealed that the ambient PM_{2.5} concentrations of urban industrial areas in China were 2–7 times higher than the daily values in this study. These areas were Taiyuan ($147 \mu\text{g}/\text{m}^3$), Jiangjin ($97.1 \mu\text{g}/\text{m}^3$), Xi'an ($50.6 \mu\text{g}/\text{m}^3$), and near steel and glass industries at Kunming ($130 \mu\text{g}/\text{m}^3$). The concentrations were also about the same

as in Kaohsiung's industrial estate, Taiwan ($25.3 \mu\text{g}/\text{m}^3$), where there is steel making, oil refining, freight transport, and shipbuilding [13]. In this study, PM_{2.5} levels were lower than those found in an industrial city ($40.6 \mu\text{g}/\text{m}^3$): Karaj, Iran [17]. However, a one-way ANOVA was used to differentiate the average PM_{2.5} concentrations during the dry season. There was no significant difference in the PM_{2.5} concentrations between the months ($p > 0.05$). According to the findings, the urban industrial area and transportation may not significantly affect PM_{2.5} concentrations in the dry season.

The correlations between the PM_{2.5} concentrations and meteorological conditions, i.e., total precipitation (Prec.), relative humidity in the atmosphere (RH), wind speed (WS), and ambient temperature (Temp.), are shown in Table 2 and Figure 2. The results showed that the PM_{2.5} concentrations measured in Rayong were negatively correlated ($r = -0.258$) with temperature ($p < 0.05$), indicating the possibility of the mix height increasing when the temperature is high; hence, PM_{2.5} levels were found to be decreased. Furthermore, the negative correlations of wind speed with ambient temperature and relative humidity were found to be significant ($r = -0.416$ and -0.630 , $p < 0.01$), implying that high wind speeds lowered the ambient temperature, relative humidity, and PM_{2.5}. In general, increased rainfall and wind speed cause the dilution of air pollutants at the ground level, resulting in a reduction in ambient PM_{2.5} as well [34].

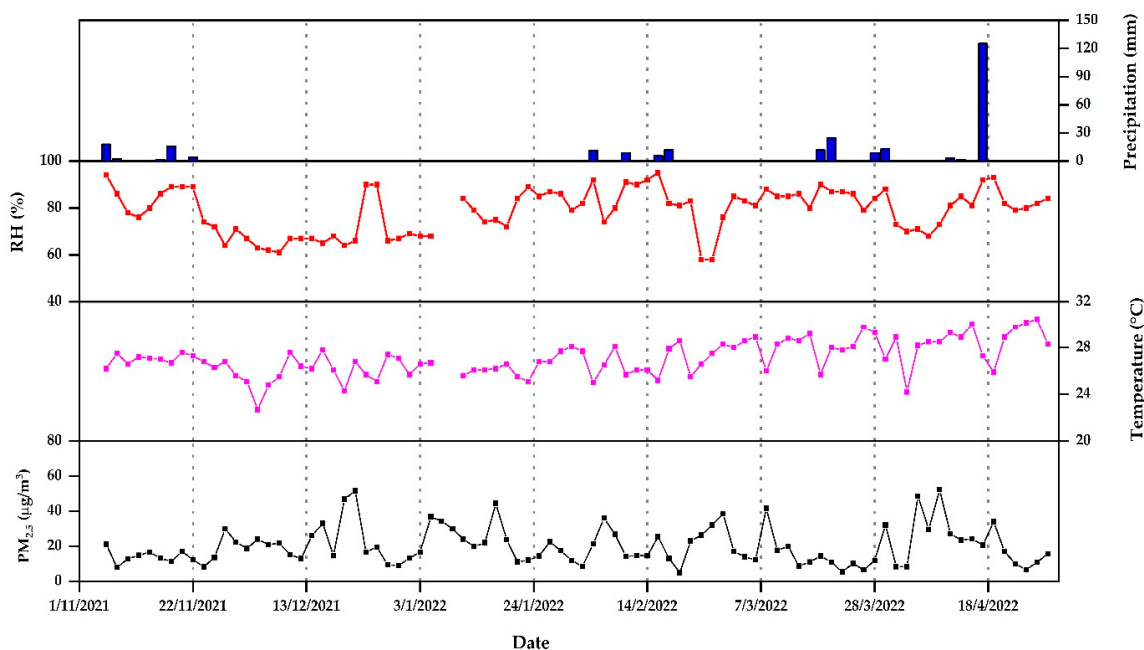


Figure 2. Daily PM_{2.5} concentrations and meteorological parameters during the sampling period in Rayong.

Figure 3 illustrates the wind direction and wind speed during the sampling period in the dry season from north (N) to south (S), which is influenced by the northeast monsoon (NE monsoon) in Thailand. There is then less PM_{2.5} accumulation on land, as it will be blown into the sea. Furthermore, the World Meteorological Organization (WMO) Global Producing Centers of Long-Range Forecasts estimated that the La Niña conditions at the time of writing would persist during March–May 2022, reducing the PM_{2.5} by 65%. The reason is that rain has diluted the PM_{2.5} in Rayong [35]. Meteorological factors may have influenced PM_{2.5} concentrations, though most of the correlations of PM_{2.5} with meteorological factors revealed no significance. However, the PM_{2.5} values in Rayong during the dry season were influenced by air temperature. Reduced ambient temperature is accompanied by a lower mixing height, which prevents pollutant dispersion [34].

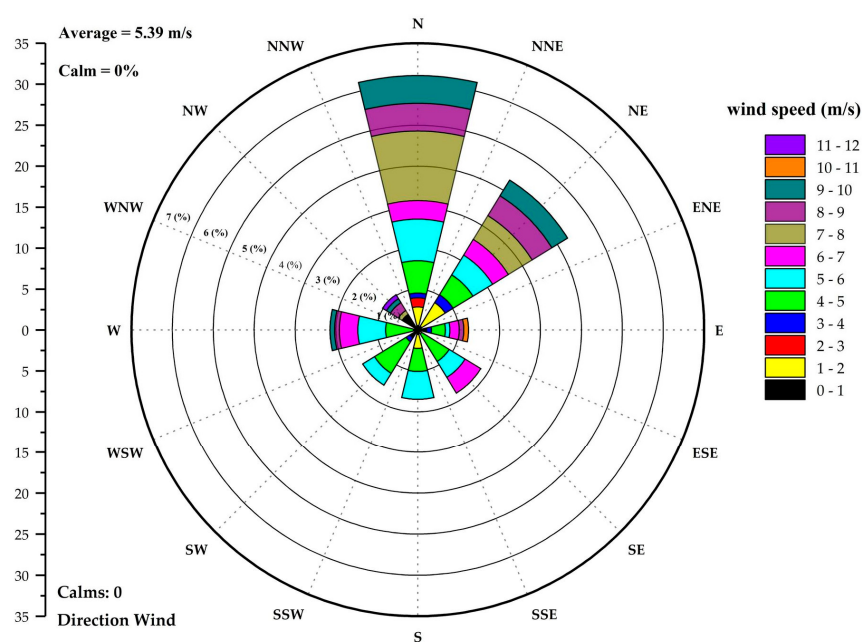


Figure 3. Distribution of wind speed and wind direction measured at Rayong during the sampling period.

Table 1. The concentrations of heavy metals and elements in PM_{2.5} in the urban and industrial areas of Rayong; Khon Kaen, Thailand; and other cities.

Sampling Sites	Duration	PM _{2.5} ($\mu\text{g}/\text{m}^3$)	Elements Concentrations (ng/m ³)											Ref.
			As	Cd	Cr	Cu	Fe	K	Mn	Ni	Pb	V	Zn	
Urban–Industrial area in Rayong, Thailand (n = 88)	Mean $\pm$ SD	20.1 $\pm$ 10.9	0.84 $\pm$ 0.65	1.24 $\pm$ 2.32	18.9 $\pm$ 8.2	6.63 $\pm$ 6.58	248 $\pm$ 381	438 $\pm$ 301	20.1 $\pm$ 29.9	3.13 $\pm$ 5.49	28.5 $\pm$ 47.8	0.65 $\pm$ 0.55	553 $\pm$ 1040	This work
	Range	4.9–52.3	n.d.–2.91	n.d.–12.9	n.d.–67.7	n.d.–39.4	n.d.–2811	7.1–1398	n.d.–196	n.d.–27.4	n.d.–350	n.d.–3.53	n.d.–7223	
	Median	17.0	0.88	0.42	18.5	4.78	140	350	11.5	1.14	13.6	0.52	254	
Urban–Industrial area in Kunming, China	2013 to 2014	130	26.8	6.00	29.1	78.8	-	-	156	20.5	282	40.3	327	Han et al. [5]
Industrial area in Khon Kaen, Thailand	December 2020 to February 2021	44.5	-	101	-	186	4.7	-	2296	-	273	-	4.5	Sakunkoo et al. [7]
Industrial area in Kaohsiung, Taiwan	May 2015 to April 2018	25.3	1.07	0.679	6.59	8.31	216	198	15.4	9.43	18.9	18.0	125	Hsu et al. [13]
Industrial area in Jiangjin, China	6 to 28 January 2019	97.1	7.56	-	4.29	15.8	-	-	58.6	1.39	37.9	0.60	94.2	Han et al. [15]
Industry city in Karaj, Iran	2018 to 2019	40.6	32.1	84.0	49.5	203	-	-	84.7	60.8	133	-	242	Kermani et al. [17]
Urban–Industrial area in Xi'an, China	2015 to 2016	50.6	116	16.5	-	41.3	-	-	33.7	10.6	34.1	6.00	200	Liu et al. [32]
Urban–Industrial area in Taiyuan, China	7 to 22 November 2016	147	7.07	7.60	175	161	-	-	218	96.2	498	-	513	Liu et al. [33]
WHO guideline		15	6.6	5	0.25	70 *	-	-	150	25	500	1000	-	WHO [36]

n.d.—not detected; * WHO (2014) cited in Niampradit et al. [18].

Table 2. Spearman's rho correlation coefficients between the meteorological data and PM_{2.5} concentrations in Rayong.

Meteorological Factors	Temp	RH	Prec.	WS	PM _{2.5}
Temperature (°C)	1				
Relative Humidity (%)	0.024	1			
Precipitation (mm)	−0.100	0.528 **	1		
Wind Speed (m/s)	−0.416 **	−0.630 **	−0.123	1	
PM _{2.5} ($\mu\text{g}/\text{m}^3$)	−0.253 *	−0.202	−0.112	0.046	1

* Correlation is significant at the 0.05 level (two tailed). ** Correlation is significant at the 0.01 level (two tailed).

3.2. Heavy Metals and Elements Concentrations in PM2.5

The average concentrations of heavy metals and elements analyzed from the PM2.5 samples are presented in Table 1. Concentrations of heavy metals and elements in the dry season at Rayong in descending order were Zn ($553 \pm 1040 \text{ ng/m}^3$) > K ($438 \pm 301 \text{ ng/m}^3$) > Fe ($248 \pm 381 \text{ ng/m}^3$) >> Pb ($28.5 \pm 47.8 \text{ ng/m}^3$) > Mn ($20.1 \pm 29.9 \text{ ng/m}^3$) > Cr ($18.9 \pm 8.2 \text{ ng/m}^3$) > Cu ($6.63 \pm 6.58 \text{ ng/m}^3$) > Ni ($3.13 \pm 5.49 \text{ ng/m}^3$) > Cd ($1.24 \pm 2.32 \text{ ng/m}^3$) > As ($0.84 \pm 0.65 \text{ ng/m}^3$) > V ($0.65 \pm 0.55 \text{ ng/m}^3$). In the PM2.5 samples, the dominant percentages of heavy metals and elements were 41.95% Zn, 33.20% K, 18.80% Fe, and 2.16% Pb (Figures 4 and 5). Some of the heavy metals (As, Cd, Cu, Ni, and V) were very rare ($\leq 2\%$), and the total amount of heavy metals, including Cr, Cu, Fe, Mn, Pb, V, and Zn, constituted 70% of the PM2.5 samples. This finding is consistent with the findings of Hsu et al. [13], who found high concentrations of K, Fe, Zn, and Pb in industrial estates in Taiwan that included steel making, oil refining, freight transport, and shipbuilding, the same types of industry found in Rayong. Zn, Cu, Ni, Mn, and Pb are the most common metals found in industrial zones [5,15,17,32,33]. To preserve the ambient air quality, the WHO has set limits on toxic metal concentrations (Table 1). The guideline values for Cd, As, Cr, Ni, Cu, Mn, Pb, and V values are 5, 6.6, 20, 25, 70, 150, 500, and 1000 ng/m^3 , respectively [36]. When heavy-metal levels were compared to WHO guidelines, it was discovered that the Cr concentration in Rayong was 75 times higher than the WHO limit. Kermani et al. [17] reported that Al, Zn, Cu, Fe, Mg, and Ni emissions from the manufacturing of products such as scrap containers, fashion accessories, stainless steel, ceramics, and electronic parts, were consistent, along with emissions from the mechanical wear of electrical components, automotive parts, and metal parts in machinery. Furthermore, Fe, Zn, V, Cu, and Mn were found at the industrial site, whereas Ba, Cr, Ni, Mo, and Co were found to be traffic related in Taiwan's urban area [13]. Mn, Al, Pb, and Cu were the most prevalent PM2.5-bound heavy metals emitted from the industrial zone, residential area, university area, and agricultural area in Khon Kaen, Thailand [7]. For the dominant heavy metals and elements in PM2.5, we identified the sources by SEM-EDS and PCA.

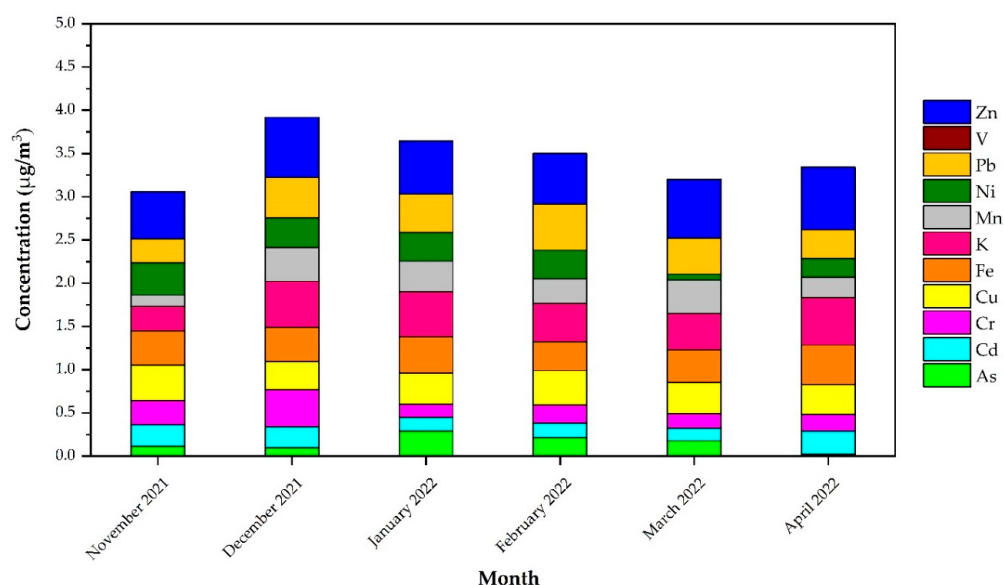


Figure 4. Monthly average concentrations of heavy metals in PM2.5 during the sampling period in Rayong.

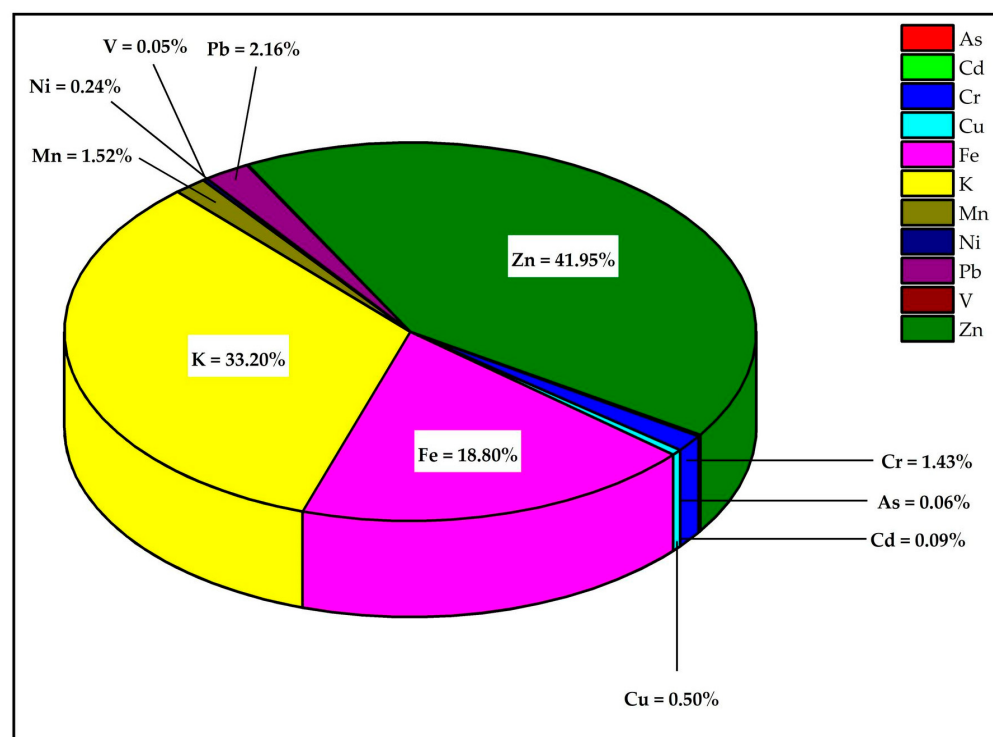


Figure 5. Species distribution of heavy metals in PM_{2.5} during the sampling period in Rayong.

3.3. Investigation of the Morphological and Elemental Characterization of PM_{2.5}

PM_{2.5} samples were collected on a quartz filter at Rayong during the dry season for SEM-EDS analysis. According to the SEM-EDS analysis of characteristics, ambient PM_{2.5} collected from Rayong contained the elements listed in Table S4. EDS analyses revealed C (26.6 to 68.1%), O (22.4 to 49.6%), and Si (4.70 to 39.6%) to be major components in the samples, which was expected, given the presence of soot particles formed as a result of burning or combustion [3]. The most common particle type in Figure 6e was soot aggregate in the form of chains and large aggregated clusters. Figure 6a–d,h illustrates particles with irregular shapes and abundant phases of O, Al, and Si. These particles could have originated from construction activities and fragmentation/melting within engines, boilers, and burners [3]. According to previous research by Li et al. [3] and Labrada-Delgado et al. [37], the major components of O and Fe (0.18 to 3.26%) with sizes ranging from 0.1 to 5 µm and spherical and irregular shapes were obtained from extractive metallurgy activities such as mining, smelting, calcining, and refining, as illustrated in Figure 6d–f. The PM_{2.5} samples in this study from Rayong revealed the presence of Fe-rich particles, which originated from metallurgical activities. Furthermore, these samples contained Zn (0.87 to 4.60%) and Cu (1.02 to 2.09%), which are products of metallic industrial activity [14,37], and one of our testing sites was close to an industrial smelting plant. Then, regarding diesel combustion and traffic emissions, V (0.08 to 0.33%) and Cr (0.02 to 0.77%) were found [38]. A significant amount of K (0.01 to 3.12%) was obtained from biomass burning in agricultural activities [3,39,40].

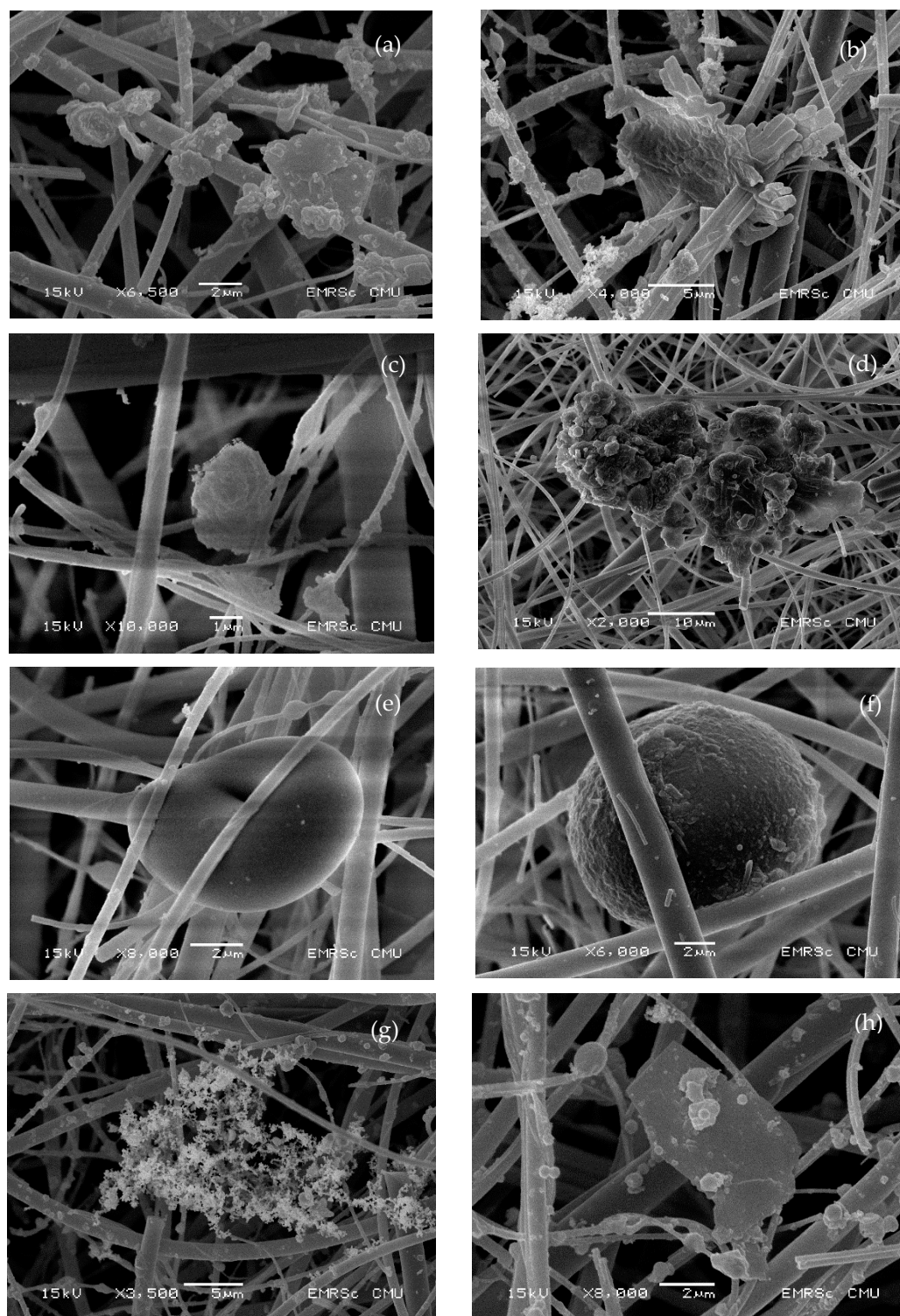


Figure 6. Morphologies of PM_{2.5} samples in Rayong; (a–d,h) irregular shapes, (e,f) aggregate shapes, (g) soot aggregate in the form of chains.

Fly ash particles, soot particles, and some mineral particles were detected using a SEM-EDS analysis of individual particles. Based on their morphologies and elemental composition, industrial activities, traffic emissions, and biomass burning activities were identified. However, we could not obtain an accurate source apportionment result solely through SEM-EDS analysis. As a result, the sample filters were extracted and analyzed using ICP-OES to confirm the sources of particles.

3.4. Source Identification by Principal Component Analysis (PCA)

The results of the PCA classifying the sources of heavy metals in PM_{2.5} during the dry season in Rayong are shown in Table 3. Only factor loadings higher than 0.5 were deemed to be statistically significant. The loadings having a greater value than 0.50 are marked bold in the table [41]. There were two components contributing to Rayong's PM_{2.5} during the dry season of 2022. This suggests that two factors producing heavy metals that end up as PM_{2.5} account for 72.8% of the cumulative variance and that the eigenvalues from the varimax-rotated factor analysis were more than one. The cumulative variance of the first component at the receptor site was 46.3%, which demonstrates the high proportions of Zn, Fe, Pb, Mn, Cu, and Cd. These parameters suggest contamination from industrial sources [42,43]. Tokaloglu et al. [44] found a significant positive relationship between each pair among Cu, Ni, Cd, Co, Cr, and Zn, which could be derived from industrial activities. Moreover, Bhuiyan et al. [45] showed that the highest loadings of total variance in the first factor are mostly contributed by Cu, Ni, Cd, Co, Cr, and Zn. These metal compounds were identified as coming from various metal processing industries. These heavy metal compounds could indicate that the major sources of pollutants in Rayong are industrial emissions. The second component showed high loadings of Ni, Cr, As, Cd, V, and K, which were contributed from traffic emissions [46,47]. Road traffic in the urban areas is one of the major sources of Cd and Cr [48]. Furthermore, Cr and Ni elements are suggested to be emitted from mechanical friction and motor vehicle exhausts [49,50]. As a result, particle emissions were linked to traffic emissions such as fuel combustion, vehicle component wear, road abrasion, and roadway maintenance [51,52].

Table 3. Rotated principal component loadings of heavy metals in PM_{2.5} fractions in Rayong.

Heavy Metals	Component	
	1	2
As	0.033	0.642
Cd	0.504	0.586
Cr	0.255	0.762
Cu	0.883	0.339
Fe	0.977	0.135
K	0.401	0.507
Mn	0.934	0.320
Ni	0.018	0.760
Pb	0.971	0.162
V	0.303	0.687
Zn	0.984	0.122
Eigenvalues	6.23	1.78
% of Variance	46.3	26.5
Cumulative %	46.3	72.8
Estimate sources	Industrial sources	Traffic sources

Factor loading values of >0.50 are shown in bold.

3.5. Health Risk Assessment of Heavy Metals

3.5.1. Non-Carcinogenic Risk Assessment for Heavy Metals

All adverse health effects in an organism brought on by environmental exposures other than cancer are referred to as non-carcinogenic risks. The epidemiological study of the non-carcinogenic risk of heavy metals in PM_{2.5} via inhalation used the hazard quotient (HQ); see Table 4. Along with Cd, Ni, Pb, V, Cu, Fe, Mn, and Zn, the concentrations of heavy metals constituted a major element in the analysis of non-carcinogenic risk. The HQ of every heavy metal in ambient air was lower than 1.0, indicating no significant risk of chronic non-carcinogenic effects from the PM_{2.5}-bound heavy metals in Rayong during the dry season. For babies, children, and adults, the results of non-carcinogenic risks from the inhalation of PM_{2.5}-bound heavy metals were similar. The non-carcinogenic risks for

Rayong in descending order were $Mn > Ni > Cr > Cd > As > Pb > V > Zn > Fe > Cu$. In addition, the hazard index (HI) of Rayong for all age groups during the dry season was 0.39, which is less than 1.0 and denotes a low likelihood of impacting health. The result of this study was lower than that of Sakunkoo et al. [7], which revealed that the HQ values of heavy metals in PM_{2.5} in an industrial area in Khon Kaen province, Thailand, were greater than 1.0 for all age groups; children appeared to be more likely to be affected than adults. Han et al. [13] reported that HQ values for the inhalation of PM_{2.5}-bound toxic metals emitted from coal combustion, biomass burning, industrial emission, ship emission, and vehicle emission were 6.52×10^{-2} , 3.33×10^{-2} , 1.16×10^{-1} , 3.48×10^{-2} , and 2.59×10^{-2} , respectively. However, these are less than 1.0, indicating safety. Additionally, Krupnava et al. [14] found that the non-cancer risk of exposure to heavy metals in PM_{2.5} in Chelyabinsk's industrial districts was lower than 1.0 for both children and adults, excepted for Mn, whose HQ values were higher than those of all other heavy metals in PM_{2.5} and higher than 1.0 in children (1.02 and 1.19). The HQ of Mn was related to metallurgical plants. Taiwan's Kaohsiung is located by an industrial complex, where industries including shipbuilding, freight transportation, and oil refining are located. They observed that there was concern about the non-carcinogenic risk posed by Mn [13]. The HQ of Mn (0.19) in our study was related to the iron ore and steel mill industries.

Table 4. Non-carcinogenic risks from PM_{2.5}-bound heavy metals calculated by HQ and HI.

Elements	RfC * (mg/m ³)	HQ		
		Baby (≥2 y)	Children (2–15 y)	Adults (16–70 y)
As	1.5×10^{-5}	2.69×10^{-2}	2.69×10^{-2}	2.69×10^{-2}
Cd	1.0×10^{-5}	5.73×10^{-2}	5.73×10^{-2}	5.73×10^{-2}
Cr	1.0×10^{-4}	9.07×10^{-2}	9.07×10^{-2}	9.07×10^{-2}
Ni	1.4×10^{-5}	1.06×10^{-1}	1.06×10^{-1}	1.06×10^{-1}
Pb	3.5×10^{-3} **	3.91×10^{-3}	3.91×10^{-3}	3.91×10^{-3}
V	1.0×10^{-4}	3.12×10^{-3}	3.12×10^{-3}	3.12×10^{-3}
Cu	4.0×10^{-2} **	7.95×10^{-5}	7.95×10^{-5}	7.95×10^{-5}
Fe	7.0×10^{-1} **	1.70×10^{-4}	1.70×10^{-4}	1.70×10^{-4}
Mn	5.0×10^{-5}	1.90×10^{-1}	1.90×10^{-1}	1.90×10^{-1}
Zn	3.0×10^{-1} **	8.85×10^{-4}	8.85×10^{-4}	8.85×10^{-4}
HI		3.88×10^{-1}	3.88×10^{-1}	3.88×10^{-1}

* US-EPA [23]; ** Yang et al. [24].

3.5.2. Lung Cancer Risk

Heavy metals such as Cd, Cr, Ni, and Pb are thought to cause cancer (toxic metals) [2,23]. For babies, children, and adults in Rayong during the dry season, the individual lifetime cancer risk (CR_{inh}) is an estimate of the risk of developing lung cancer from the inhalation of PM_{2.5} and toxic metals bound to PM_{2.5}. The results of the estimation of lung cancer risks from PM_{2.5} and toxic metals in PM_{2.5} exposure are shown in Table 5. The CR_{inh} values for babies, children, and adults regarding inhalation of As, Cd, Cr, Ni, Pb, and PM_{2.5} were 4.95×10^{-8} to 1.36×10^{-6} , 3.06×10^{-8} to 8.39×10^{-7} , 2.18×10^{-5} to 5.98×10^{-4} , 9.98×10^{-9} to 2.74×10^{-7} , 4.69×10^{-9} to 1.29×10^{-7} , and 2.20×10^{-3} to 6.06×10^{-2} , respectively. However, the CR_{inh} levels for exposure to all carcinogenic metals in Rayong during the dry season denoted no risk ($<10^{-6}$), with the exception of the Cr-related carcinogenic risk, which was an acceptable or tolerable risk (10^{-6} to 10^{-4}). On the other hand, CR_{inh} values for the inhalation of PM_{2.5} revealed a high lung cancer risk ($>10^{-4}$). The actual lifetime risk was estimated by age-dependent potency adjustment factors (ADAFs). Table 5 shows the TCR_{inh} —the combined carcinogenic risk for babies, children, and adults due to the inhalation of PM_{2.5} and carcinogenic metals in PM_{2.5}. It was found that the TCR_{inh} of Cr was acceptable (10^{-6} to 10^{-4}), but PM_{2.5} inhalation implied a high lung cancer risk ($>10^{-4}$). Hsu et al. [13] reported that 80% of Al and Cr were

related to non-ferrous metallurgy and traffic emissions in an industrial estate of Taiwan. The total Cr was analyzed in this study, and the type of Cr that is most hazardous and able to enter the lungs is Cr(VI). Cr(VI) exposure has strong associations with mortality from lung, nose, and nasal cavity cancers [18].

Table 5. The mean lifetime carcinogenic risk (CR_{inh}) and total lung cancer risk (TCR_{inh}) resulting from PM2.5 and toxic elements in PM2.5.

Elements	IUR * (Per $\mu\text{g}/\text{m}^3$)	CR_{inh}			ADAFs			TCR_{inh}
		Baby (≥ 2 y)	Children (2–15 y)	Adults (16–70 y)	CR_{baby}	$CR_{children}$	CR_{adults}	
As	0.0043	4.95×10^{-8}	3.47×10^{-7}	1.36×10^{-6}	1.38×10^{-8}	2.03×10^{-7}	1.04×10^{-6}	1.29×10^{-6}
Cd	0.0018	3.05×10^{-8}	2.13×10^{-7}	8.39×10^{-7}	8.71×10^{-9}	1.28×10^{-7}	6.59×10^{-7}	7.96×10^{-7}
Cr	0.084 ¹	2.18×10^{-5}	1.52×10^{-4}	5.98×10^{-4}	6.22×10^{-6}	9.14×10^{-5}	4.70×10^{-4}	5.68×10^{-4}
Ni	0.00024	9.98×10^{-9}	6.99×10^{-8}	2.74×10^{-7}	2.94×10^{-9}	4.32×10^{-8}	2.22×10^{-7}	2.68×10^{-7}
Pb	0.000012	4.69×10^{-9}	3.28×10^{-8}	1.29×10^{-7}	1.34×10^{-9}	1.97×10^{-8}	1.01×10^{-7}	1.22×10^{-7}
PM2.5	0.008 **	2.20×10^{-3}	1.54×10^{-2}	6.06×10^{-2}	6.30×10^{-4}	9.26×10^{-3}	4.76×10^{-2}	5.75×10^{-2}

¹ IUR of Cr is represented by hexavalent chromium (Cr(VI)), which has been classified as an inhalation-related human carcinogen; * US-EPA [23]; ** Geene and Morris [26].

The lung cancer risk was estimated using a million people. The cases of lung cancer related to As, Cd, Cr, Ni, and Pb inhalation in Rayong during the dry season were 1.29, 0.80, 568, 0.27, and 0.12 per million people, and those related to PM2.5 exposure were approximately 57,500 per million. Furthermore, among babies, children, and adults, adults had a higher risk of developing lung cancer from exposure to PM2.5 and toxic metals, which may be related to the bioaccumulation of heavy metals in the body [12,13]. According to the findings of the study, inhaling PM2.5 in Rayong was associated with an increased chance of developing lung cancer, although exposure to toxic metals may not have an effect on this risk. However, the human health risk assessment has proved that areas with the greatest levels of heavy metals and PM2.5 exposure are urban and industrial areas. The risk assessment of PM2.5 toxicity demonstrated the significance of applying a risk-oriented approach to any possible risks related to metal exposure.

4. Conclusions

The concentrations of PM2.5 and heavy metals in PM2.5 were measured in industrial and metropolitan parts of Rayong during the dry season. The average PM2.5 concentration was $20.1 \pm 10.9 \mu\text{g}/\text{m}^3$. The percentages of days exceeding the daily WHO limit and the US-EPA NAAQS were 56.8% and 10.2%, respectively, but the levels did not exceed $50 \mu\text{g}/\text{m}^3$, which is the limit of the 24 h standard defined by the PCD in Thailand. The concentrations of major components in PM2.5 were Zn ($553 \pm 1040 \text{ ng}/\text{m}^3$), K ($438 \pm 301 \text{ ng}/\text{m}^3$), Fe ($248 \pm 381 \text{ ng}/\text{m}^3$), Pb ($28.5 \pm 47.8 \text{ ng}/\text{m}^3$), Mn ($20.1 \pm 29.9 \text{ ng}/\text{m}^3$), and Cr ($18.9 \pm 8.2 \text{ ng}/\text{m}^3$). Zn (41.7%), K (33.2%), and Fe (18.8%) were found to be the dominant heavy metals and elements. Cr, Cu, Fe, Mn, Pb, V, and Zn constituted 70% of the heavy metals in PM2.5 samples. PM2.5 concentration and air temperature were significantly negatively correlated ($p < 0.01$) with high temperatures. The PCA results showed two factors contributed to Rayong's PM2.5 during the dry season of 2022. The industrial emissions (Cd, Cu, Fe, Mn, Pb, and Zn) and traffic emissions (As, Cd, Cr, K, and Ni) were the major sources of PM2.5-bound heavy metals in Rayong.

The non-carcinogenic risk levels presented by PM2.5-bound heavy metals in Rayong during the dry season were measured with the hazard quotient (HQ). The HQ values of all the heavy metals in ambient air were lower than 1.0, indicating no significant risk of chronic effects, and the hazard index (HI) for each age groups was less than 1.0 (HI explains risk levels that are probable to have an impact on health). The carcinogenic risk assessments of PM2.5 and toxic metals in PM2.5 were based on the lifetime cancer risk (CR_{inh}). The CR_{ic} for babies (6.30×10^{-4}), children (6.30×10^{-4}), and adults (4.76×10^{-2}) in Rayong

during the dry season indicate a high lung cancer risk ($>10^{-4}$). Exposure to toxic metals in PM_{2.5} through the inhalation pathway in Rayong obviously presents a high risk of cancer ($>10^{-4}$) based on the TCR_{inh}. The TCR_{inh} values of Cr for the combined age groups were higher than 10^{-6} , which indicates a high cancer risk in Rayong. Therefore, the results of PM_{2.5} concentrations and Cr in PM_{2.5} are important for environmental management and protection in Rayong. Moreover, the morphology of PM_{2.5} and PCA analysis indicated the sources of PM_{2.5} to be industrial and vehicle emissions.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/su142215368/s1>, Table S1: Recoveries of elements obtained from CRM No. 28 Urban Aerosols and spiking method; Table S2: The limit of detection (LOD) and limit of quantitation (LOQ) of elements; Table S3: Meteorological parameters in Rayong city during dry season (November 2021 to April 2022); Table S4: EDS atomic percentage for individual PM_{2.5} samples in Rayong city.

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