



Surveillance of long-term environmental elements and PM_{2.5} health risk assessment in Yangtze River Delta, China, from 2016 to 2020

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Abstract

PM_{2.5} metal pollution significantly harms human health. The air quality in Wuxi is poor, especially in winter, and long-term monitoring of PM_{2.5} elements comprising has not been performed previously. In the present study, 420 PM_{2.5} samples were collected from January 2016 to December 2020. Eleven elements, including Al, Mn, Ni, Cr, As, Cd, Sb, Hg, Pb, Se, and Tl, were analyzed by inductively coupled plasma mass spectrometry. The mean PM_{2.5} level was $56.1 \pm 31.0 \mu\text{g}/\text{m}^3$, with a tendency of yearly decreasing and a significant seasonal distribution variation. The concentration of 11 elements in the PM_{2.5} samples was $0.38 \pm 0.33 \mu\text{g}/\text{m}^3$. Al was the highest element with a range of 37.5–2148 ng/m³. Meanwhile, the spatial distribution differences were compared by literatures review. Based on the Crystal Ball model, health risks were assessed dynamically using Monte Carlo uncertainty analysis. After 10,000 simulations, the mean value of the hazard index for nine elements was 0.743, and Mn contributed the most to the hazard index among elements, with a correlation of 0.3464. The average carcinogenic risk was 1.01×10^{-5} , which indicated that the non-carcinogenic and carcinogenic risks were within the acceptable range. However, considerable attention should be paid to the potential health risks associated with long-term Al, Mn, and As exposure. This study provides detailed data on local atmospheric pollution characteristics, helps identify potential risk elements, and contributes to the development of effective regional air quality management.

Keywords PM_{2.5} · Elements · Long-term surveillance · Health risk assessment · Yangtze River Delta

Introduction

Air pollution has become a serious environmental issue and now causes annual several million deaths worldwide (Toriba and Hayakawa 2021). Fine particulate matter (PM_{2.5}) refers to particulate matter suspended in air with an aerodynamic equivalent diameter of 2.5 μm or less. PM_{2.5} has a long atmospheric residence time and transportation distance. Long-term exposure to PM_{2.5} has been associated with increased all-cause and cardiopulmonary mortality

(Mohammed et al. 2016; Barrett 2020; Xu et al. 2020; Akhtar and Palagiano 2018). A cohort study of 18.9 million Medicare beneficiaries showed that a 10-μg/m³ elevation in 12-month moving average PM_{2.5} exposure was associated with 24%, 60%, and 10% greater risks of respiratory disease, pneumonia, and COPD mortality, respectively, in an elderly population of USA (Pun et al. 2017). Also, airway epithelial cell exposure to PM_{2.5} significantly disturbs cell membranes, inducing cell necrosis, necroptosis, or pyroptosis and lung microbiome and its metabolic profile showed considerable alteration in mice (Li et al. 2020). In addition, owing to the large specific surface area of PM_{2.5}, it can easily absorb harmful heavy metals, organic matter, bacteria, and viruses (Teng et al. 2016; Wang et al. 2021a, b; Kang et al. 2020; Badaloni et al. 2017).

Heavy metals and other elements are the main components of ambient PM_{2.5}, and can lead to human functional dysfunction and irreversible damage. For instance, a previous study showed that Cd levels are associated with an increase in cell-free bronchoalveolar lavage fluid in smokers in a cohort of patients with chronic obstructive pulmonary

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disease (Sundblad et al. 2016). Cd and Pb exposure have also been shown to induce peribronchiolar fibrosis and lung remodeling due to stimulation of vimentin phosphorylation (Li et al. 2017; Gogoi et al. 2019). Major toxic metals and their interactions are also understood in *in vitro* studies. The toxicity contribution and combined effects of PM-bound metals in human lung epithelial cells (A549) have been confirmed (Yuan et al. 2019). The nominal exposure concentration of PM_{2.5} suspension was 100 mg/L, and exposure to Zn (480 µg/L), Cr (40.2 µg/L), Mn (79 µg/L), Fe (3920 µg/L), Cu (61 µg/L), or Pb (655 µg/L) significantly decreased the cell viability of A549. The contribution of metals contained was estimated to be approximately $22.9 \pm 11.5\%$ calculated as the decreased mortality after metals removal. Besides, the cell mortality increased more significantly in A549 cells exposed to the binary mixtures than single element. Therefore, although metals account for a small proportion of PM_{2.5} mass, their potential toxicity should not be neglected.

Wuxi, one of the most developed and important areas in China, is located in the middle of the Yangtze River Delta. With rapid industrialization and urbanization, a series of environmental problems has emerged in recent years (Wu et al. 2022; Mao et al. 2020a, b). In 2016, “the outline of the Yangtze River Economic Belt Development Plan” officially became a national development strategy with the objective of promoting a new round of urbanization. It is imperative to obtain a complete picture of the distribution characteristics of air pollutants to implement effective measures to control and avoid anthropogenic pollution. Previous studies have principally focused on PM_{2.5}, PM₁₀ (particulate matter

suspended in air with an aerodynamic equivalent diameter of 10.0 µm or less), O₃, SO₂, NO₂, or have been limited to short durations (Han et al. 2018; Mao et al. 2020a, b; Yu et al. 2020). To the best of the authors’ knowledge, long-term PM_{2.5} and element surveillance in this area are still sparse. Therefore, to fill the research gap, we collected PM_{2.5} samples for five consecutive years and analyzed 11 major elements by inductively coupled plasma mass spectrometry (ICP-MS). The temporal and spatial distribution differences were obtained through literature research and statistical analysis. Our study provides accurate data on element pollution levels and evaluates the possible risks to local human health. It is of great practical significance to improve urban air quality, rationally formulate relevant policies, and protect the health of residents.

Materials and methods

Sample collection

The sampling site was chosen at the center of the main urban area in Wuxi, as shown in Fig. 1. A KC-120H intelligent medium-flow TSP sampler equipped with a quartz filter film was used to collect the air samples. The sampler cutoff was at 2.5 µm, and the sampling flow rate was 100L/min, and the sampling height was 1.5 m. After collection, the quartz filter films were stored in a plastic filter box. The temperature, humidity, and air pressure were recorded to calculate the sampling volume under standard conditions.

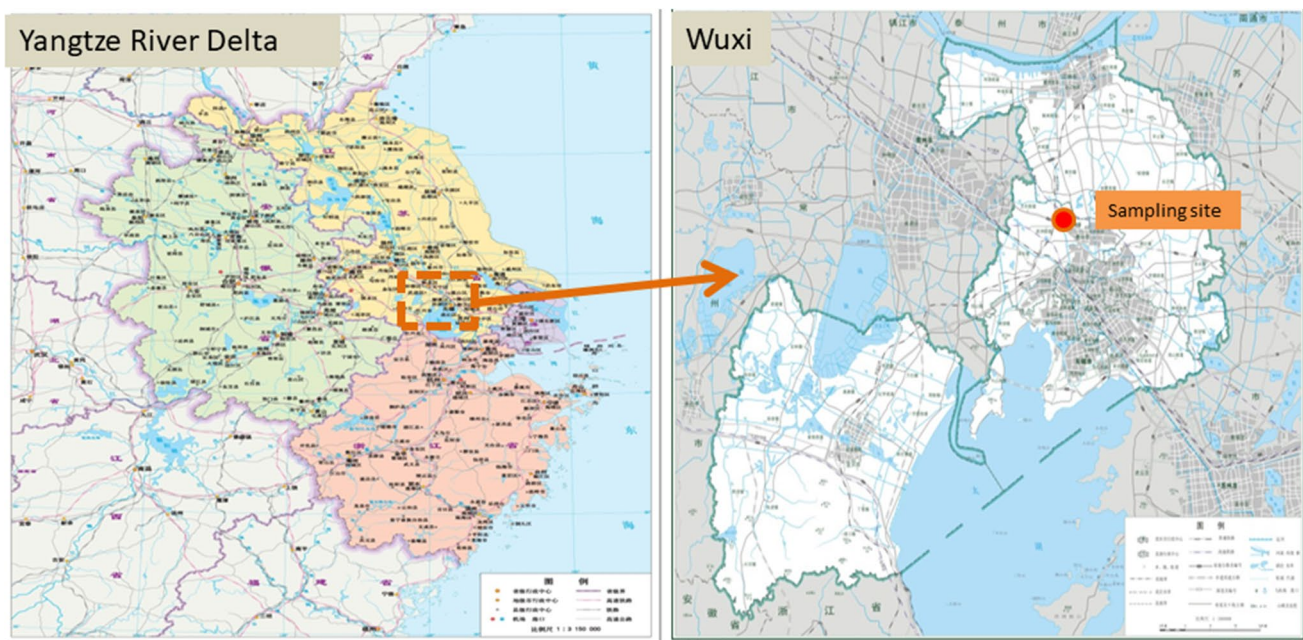


Fig. 1 PM_{2.5} sampling site in Wuxi

At the same time, field blanks were also collected the same mode as the samples, including transportation, storage, pre-treatment, and determination, except that filter films were not connected to sampler. And at least 3 blank samples were collected in each sampling batch. Samples were collected continuously for 7 days each month for 20 h per day. A total of 420 air samples were collected from January 10, 2016, to December 17, 2020.

Chemicals and reagents

Standard element solutions were obtained from Inorganic Ventures (Lakewood, NJ, USA). Deionized water was purified using the ELGA Purelab Ultra system (Vivendi Water Systems, Buckinghamshire, UK). Nitric acid and hydrochloric acid were purchased from Sigma-Aldrich Co. (LLC, USA). Individual stock solutions were prepared in pure water for ICP-MS analyses.

Sample preparation

The PM_{2.5} samples were prepared according to the requirements of the National Environmental Protection Standards of the People's Republic of China released by the Ministry of Environmental Protection of China in 2013. In summary, the PM_{2.5} samples were dissolved with acids for element measurement and the digestion was performed with an acid mixture (10 mL HNO₃ and HCl), with a temperature of 200 °C for a duration of 15 min. Subsequently, the extracts were cooled and diluted to 10 mL with 5% HNO₃, and 11 elements in ambient air particles were determined by inductively coupled plasma mass spectrometry (ICP-MS, Agilent Technologies 7900). Before sample determination, quality correction and resolution checks were performed using a tuning solution. Tuning should be performed more than for four instances to confirm that the relative standard deviation of signal strength of the elements contained in the tuning solution was less than 5%. The calibration, laboratory reagent, and field blanks were also analyzed throughout the analysis process. The elemental content of the air was calculated according to the standard sampling volume.

Health risk assessment

The health risk assessment was conducted by the standardized framework for human health risk assessment by US EPA (US EPA 2019). The four-step risk assessment process consists of hazard identification to identify the adverse health effects, dose response to document the relationship between dose and toxic effect, exposure assessment to calculate a numerical estimate of exposure or dose, and risk characterization to summarize an overall conclusion about risk. Inhalation was assumed to be the

only route of exposure. The PM_{2.5}-bound element health risk was implemented by the Crystal Ball software, Monte Carlo simulation uncertainty analysis, and sensitivity analysis.

The health risk assessment model caused by elements entering the human body through the respiratory tract was calculated using Eqs. (1) and (2):

$$HI = \sum HQ = \frac{EC}{RfC/1000} \quad (1)$$

$$CR = EC \times UR \quad (2)$$

where HI is the sum of HQ, which is used to assess the total potential of the non-carcinogenic risk caused by elements of concern. HQ is the hazard quotient (no unit), EC is the exposure concentration (µg/m³), RfC is the chronic inhalation reference concentration (mg/m³), CR is the carcinogenic risk (no unit), and UR is the inhalation unit risk ((µg/m³)⁻¹).

Human exposure concentration was calculated by Eq. (3):

$$EC = \frac{Ci \times ED \times EF \times ET}{AT} \quad (3)$$

where Ci is the concentration of PM_{2.5} (µg/m³) elements, ED is the exposure duration (years), EF is the exposure frequency (day/year), ET is the exposure time (h/day), and AT is the average exposure time (h) and is the amount of time over which exposure is averaged and is equal to ED for assessing non-carcinogenic risks. The exposure duration was 0–50 years, the frequency of exposure was 365 days/year, the exposure time was 24 h/day, and the average exposure time was 613,200 h for carcinogenic risks.

Based on published data from the US Environmental Protection Agency (US EPA), the International Agency for Research on Cancer (IARC, WHO), and Agency for Toxic Substances and Disease Registry (ATSDR, USA), the reference values of the elemental toxicity parameters are listed in Table 1.

Statistical analysis

For data analysis, Microsoft Excel 2012 using embed Crystal Ball, SPSS 18.0, and GraphPad Prism 9 were employed. Comparisons among the different groups were analyzed using the homogeneity test of variance and one-way ANOVA. Most of the literature reported the mean and standard deviation of element concentrations, and the sample sizes were frequently greater than 50. Therefore, the comparison of element levels between different regions based on the literature was analyzed using a *t*-test methodology. To improve statistical robustness, the bootstrap method was also applied. The criterion for significance was $p < 0.05$.

Table 1 Reference values of elements toxicity parameters

	Chronic inhalation reference concentration (mg/m ³)	Inhalation unit risk (µg/m ³) ⁻¹	Data sources
Al	5.00E-03	-	PPRTV ^a
Sb	3.00E-04	-	IRIS ^b
As	1.50E-03	4.30E-03	IARC ^c , IRIS
Cd	1.00E-05	1.80E-03	IRIS, ATSDR ^d
Cr	1.00E-04	-	IRIS
Pb	-	1.20E-05	CalEPA ^e
Mn	5.00E-05	-	IRIS
Hg	3.00E-04	-	IRIS
Ni	1.40E-05	2.40E-04	IRIS
Se	2.00E-02	-	CalEPA

^aProvisional peer reviewed toxicity value, US Environmental Protection Agency (US EPA), <https://hhpprtv.ornl.gov/>

^bIRIS—Integrated Risk Information System, US EPA, https://iris.epa.gov/AtoZ/list_type=alpha

^cInternational Agency for Research on Cancer (IARC), The World Health Organization (WHO), <https://www.iarc.fr/>

^dATSDR—Agency for Toxic Substances and Disease Registry, USA, <https://www.atsdr.cdc.gov/>

^eCalEPA—California Environmental Protection Agency, USA, <https://oehha.ca.gov/>

Results

Distribution characteristics of elements in PM_{2.5}

From January 2016 to December 2020, the average concentration of PM_{2.5} at major atmospheric monitoring sites in

Wuxi was 56.1 ± 31.0 µg/m³. According to the PRC National Standard (The Ministry of Environmental Protection of China 2012), the average 24-h ambient PM_{2.5} concentration limit is 75 µg/m³. 91 samples exceed the limit, accounting for 0.06% of the annual days and 21.6% of the total samples. The total content of 11 elements was 0.38 ± 0.33 µg/m³, comprising 0.68% of the total PM_{2.5} mass. The 11 elements listed in descending order of average concentration were Al, Pb, Mn, Cr, As, Ni, Se, Sb, Cd, Tl, and Hg. The average content of As in PM_{2.5} for 2016 (6.57 ± 5.25 ng/m³) exceeded the Ambient air quality standard of China and the air pollutant concentration limit of EU by 6 ng/m³. The other elements did not exceed the limit values. The concentration of As in PM_{2.5} in Wuxi was 5.01 ± 3.50 ng/m³ in recent 5 years, slightly less than other areas in China. These areas included Baoding, Hebei (8.5–113.8 ng/m³, 2016); Wuhan, Hubei (8.18 ng/m³, 2017); Beijing (7.84 ± 7.90 ng/m³, 2016); Shenzhen (6.40–7.06 ng/m³, 2018); and Taiyuan, Shanxi (13.03–15.56 ng/m³, 2018) (Xie et al. 2020; Mao et al. 2020a, b; Liu et al. 2018; Qin et al. 2020). The PM_{2.5} concentrations and those of the 11 elements detected in the atmosphere during the monitoring period are detailed in Table 2.

Figure 2a and b show the annual distribution of PM_{2.5}, and elements in the atmospheric samples. PM_{2.5} concentrations and those of the 11 elements detected in the atmosphere showed a significant decreasing trend during the last 5 years, with a *p* value less than 0.05. The contents of As, Cr, Pb, Mn, and Tl were found to differ significantly by year per the ANOVA analysis, whereas the other elements had no such characteristics.

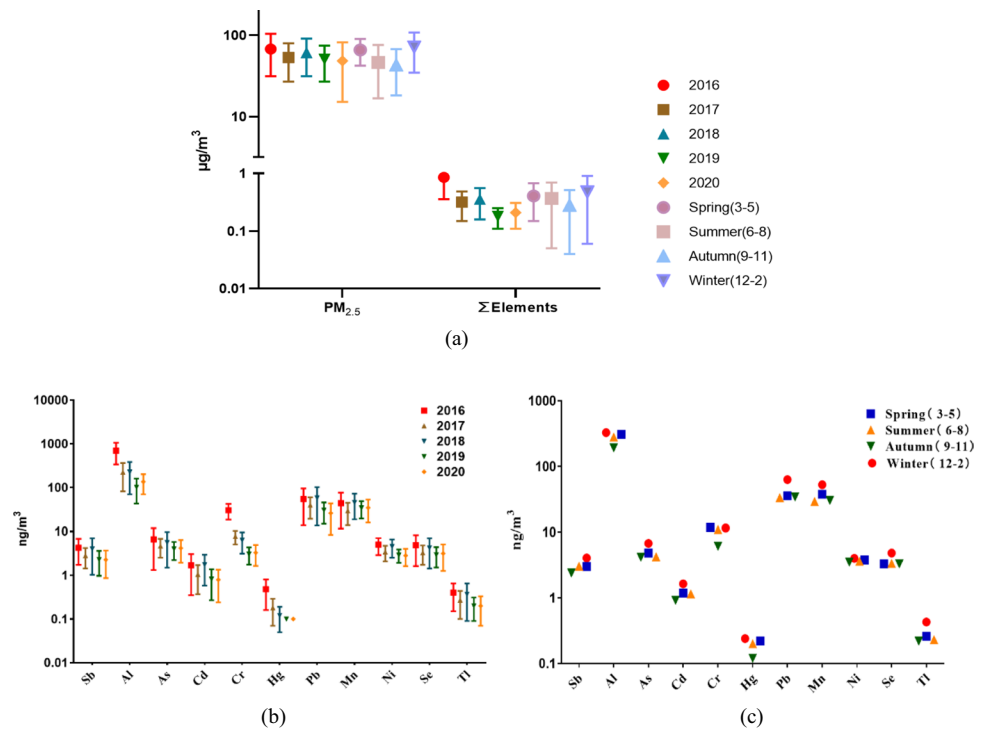
The seasonal distribution characteristics are shown in Fig. 2a and c. In winter and spring, pollutants are not

Table 2 PM_{2.5} and 11-element concentration in atmosphere sample

	Total sample	2016	2017	2018	2019	2020	Limits ^a
Sample size	420	84	84	84	84	84	
PM _{2.5} (µg/m ³)	56.1 ± 31.0	67.7 ± 36.4	53.3 ± 26.4	61.0 ± 29.7	50.7 ± 23.9	48.3 ± 33.2	75
Al (ng/m ³)	277 ± 287	698 ± 359	224 ± 142	226 ± 156	102 ± 58.7	136 ± 65.9	-
Pb (ng/m ³)	41.5 ± 32.6	54.5 ± 40.8	39.4 ± 20.1	57.6 ± 44.1	30.2 ± 15.4	25.9 ± 17.5	500
Mn (ng/m ³)	37.4 ± 23.3	43.9 ± 32.3	29.4 ± 15.6	45.3 ± 26.6	34.1 ± 14.5	34.5 ± 18.6	-
Cr (ng/m ³)	10.1 ± 11.7	10.8 ± 2.57	7.70 ± 2.65	6.33 ± 3.20	3.05 ± 1.29	3.28 ± 1.64	-
As (ng/m ³)	5.01 ± 3.50	6.57 ± 5.25	4.67 ± 2.14	5.60 ± 4.10	4.01 ± 1.77	4.17 ± 2.23	6
Ni (ng/m ³)	3.73 ± 1.81	4.99 ± 2.09	3.41 ± 1.33	4.55 ± 2.03	2.90 ± 0.97	2.83 ± 1.21	-
Se (ng/m ³)	3.70 ± 2.41	4.91 ± 3.29	3.27 ± 1.53	4.21 ± 2.78	2.95 ± 1.44	3.16 ± 1.90	-
Sb (ng/m ³)	3.13 ± 2.21	4.27 ± 2.54	2.82 ± 1.38	4.01 ± 2.98	2.29 ± 1.32	2.27 ± 1.41	-
Cd (ng/m ³)	1.22 ± 1.01	1.70 ± 1.35	1.04 ± 0.67	1.76 ± 1.18	0.82 ± 0.55	0.79 ± 0.55	5
Tl (ng/m ³)	0.29 ± 0.22	0.40 ± 0.25	0.27 ± 0.17	0.37 ± 0.28	0.20 ± 0.11	0.20 ± 0.13	-
Hg (ng/m ³)	0.20 ± 0.21	0.48 ± 0.32	0.18 ± 0.11	0.12 ± 0.07	0.10 ± 0.00	0.10 ± 0.00	50
∑Elements (µg/m ³)	0.38 ± 0.33	0.83 ± 0.39	0.32 ± 0.17	0.36 ± 0.20	0.18 ± 0.07	0.21 ± 0.10	

^aThe limits specified by the Ambient air quality standard of China, GB3095-2012

Fig. 2 Temporal distribution characteristics of PM_{2.5}-bound elements. **a** Temporal distribution of PM_{2.5} and Σ Elements. **b** Annual distribution of PM_{2.5}-bound elements. **c** Seasonal distribution of PM_{2.5}-bound elements



easily transferred due to cold weather, low precipitation, and strong atmospheric stability. Consequently, the observed PM_{2.5} concentration listed seasonally in descending order was: winter, spring, summer, and autumn. During autumn, the PM_{2.5} concentration was 42% less than that observed during winter. The Sb, As, Cd, Pb, Mn, Se, and Tl contents in winter were significantly greater than those in other seasons by one-way ANOVA multiple comparisons ($p < 0.05$), whereas the contents of Al, Cr, and Hg in winter were significantly greater than those in autumn. Compared with winter, the Sb, Al, As, Cd, Cr, Hg, Pb, Mn, Se, and Tl contents during autumn decreased by 40.7%, 41.1%, 38.3%, 43.6%, 46.9%, 49.4%, 45.5%, 42.3%, 31.8%, and 48.0%, respectively. The As, Cd, Pb, Mn, Ni, Se, and Tl contents were stable in spring, summer, and autumn, and no significant differences were found among the seasons. However, the Ni content did not vary significantly among seasons.

In addition, a PM_{2.5} content greater than 75 $\mu\text{g}/\text{m}^3$ was defined as a day exceeding the standard, while a PM_{2.5} content less than 75 $\mu\text{g}/\text{m}^3$ was defined as the clean day. From January 2016 to December 2020, there were 91 days which exceeded the standard. The average PM_{2.5} content on exceeding standards was $102 \pm 25.1 \mu\text{g}/\text{m}^3$, while the average PM_{2.5} content on clean days was $43.0 \pm 16.7 \mu\text{g}/\text{m}^3$. As shown in Fig. 3, the content of each element on the exceeded standard day was significantly greater than that on the clean day, and the ratio of element content on

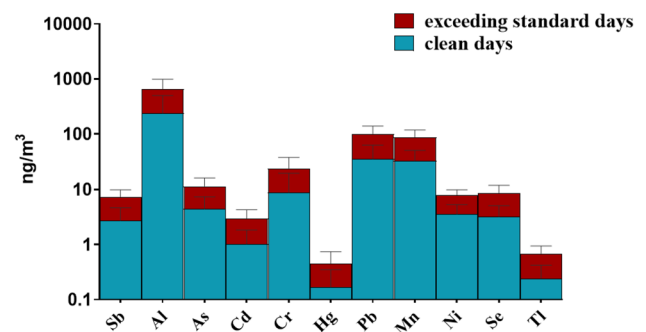


Fig. 3 Distribution characteristics of PM_{2.5}-bound elements on exceeding standard and clean days

the exceeded standard day to the clean day ranged from 1.24(Ni) to 1.93(Cd).

Literature review

Through database retrieval, the contents of atmospheric PM_{2.5} elements in other regions were obtained. By *t*-test analysis, the differences in pollution of various elements in PM_{2.5}, during the same period, were compared with other studies, and the regional distribution characteristics were obtained. Studies regarding industrial polluted areas, green spaces, rural areas, and haze days were excluded. PM_{2.5} samples in urban areas from 2016 to 2020 were selected to obtain the mean and standard deviation of the element

content and sample size information. Based on the corresponding inclusion and exclusion criteria, detailed data from the included studies are shown in Table 3. Individual *t*-tests were performed between each location and Wuxi on the data obtained. Results indicated that PM_{2.5} and its elements in the air of Europe and the USA were lower than those in Asia, such as some industrial cities in India and China. Besides, what was noteworthy was that taking Xi'an, an industrialized

city in China, as an example, its atmospheric PM_{2.5} content ($50.1 \pm 30.4 \mu\text{g}/\text{m}^3$) was less than other Asian cities, such as Taiyuan ($122.1 \pm 67.2 \mu\text{g}/\text{m}^3$) and Agra ($133 \pm 54 \mu\text{g}/\text{m}^3$), but the Cr content in PM_{2.5} was greater than that of Agra by more than a factor of 17. Currently, hexavalent Cr compounds have been classified as class I carcinogens by the IARC, which can invade the human body through digestion, the respiratory tract, skin, and mucous membrane, and may cause

Table 3 Elemental and PM_{2.5} concentrations in other countries and regions

	Average	Standard deviation	Unit	Sample size	Country/regions	<i>P</i> value
PM _{2.5}	6.2	4.1	$\mu\text{g}/\text{m}^3$	113	Toronto, Canada ^(Celo, V et al.,2021)	<0.05
	4.6	2.4	$\mu\text{g}/\text{m}^3$	70	Vancouver, Canada	<0.05
	122.1	67.2	$\mu\text{g}/\text{m}^3$	24	Taiyuan, China ^(Wang,Y et al.,2022)	<0.05
	66.25	35.73	$\mu\text{g}/\text{m}^3$	112	Henan, China ^(Liu, HJ et al.,2021)	<0.05
	50.1	30.4	$\mu\text{g}/\text{m}^3$	46	Xi'an, China ^(Wu,T et al.,2021)	<0.05
	8	8.3	$\mu\text{g}/\text{m}^3$	24	Thulamela, South Africa ^(Edlund, K. K et al.,2021)	<0.05
	133	54	$\mu\text{g}/\text{m}^3$	48	Agra, India ^(Sah, D et al., 2019)	<0.05
As	0.45	0.4	ng/m^3	113	Toronto, Canada	<0.05
	0.38	0.3	ng/m^3	70	Vancouver, Canada	<0.05
	117.2	18.6	ng/m^3	46	Xi'an, China	<0.05
Cd	0.09	0.34	ng/m^3	113	Toronto, Canada	<0.05
	0.08	0.12	ng/m^3	70	Vancouver, Canada	<0.05
	16.3	6	ng/m^3	46	Xi'an, China	<0.05
	54.4	36	ng/m^3	48	Agra, India	<0.05
Cr	0.43	0.3	ng/m^3	113	Toronto, Canada	<0.05
	0.4	0.6	ng/m^3	70	Vancouver, Canada	<0.05
	342.6	76.7	ng/m^3	46	Xi'an, China	<0.05
	19.3	9.9	ng/m^3	48	Agra, India	<0.05
Cu	3.6	2	ng/m^3	113	Toronto, Canada	<0.05
	4.2	3.6	ng/m^3	70	Vancouver, Canada	<0.05
Mn	1.9	1.6	ng/m^3	113	Toronto, Canada	<0.05
	1.5	1.2	ng/m^3	70	Vancouver, Canada	<0.05
	3.9	4.4	ng/m^3	24	Thulamela, South Africa	<0.05
	146.7	77	ng/m^3	48	Agra, India	<0.05
Ni	0.21	0.2	ng/m^3	113	Toronto, Canada	<0.05
	0.5	0.5	ng/m^3	70	Vancouver, Canada	<0.05
	11.3	5	ng/m^3	46	Xi'an, China	<0.05
	14	5	ng/m^3	24	Thulamela, South Africa	<0.05
	123.9	78	ng/m^3	48	Agra, India	<0.05
Pb	2	1.3	ng/m^3	113	Toronto, Canada	<0.05
	1.6	2.4	ng/m^3	70	Vancouver, Canada	<0.05
	102.4	67.3	ng/m^3	24	Taiyuan, China	<0.05
	35	5	ng/m^3	46	Xi'an, China	0.1779
	139.5	102	ng/m^3	48	Agra, India	<0.05
Sb	0.55	0.54	ng/m^3	113	Toronto, Canada	<0.05
	0.56	0.38	ng/m^3	70	Vancouver, Canada	<0.05
	25	18	ng/m^3	24	Thulamela, South Africa	<0.05
Se	0.58	0.5	ng/m^3	113	Toronto, Canada	<0.05
	0.14	0.1	ng/m^3	70	Vancouver, Canada	<0.05
Al	3180.6	679	ng/m^3	46	Xi'an, China	<0.05

cancer after long-term exposure. Therefore, the air quality cannot be effectively controlled by limiting the concentration of PM_{2.5}.

Health risk assessment of PM_{2.5} and elemental components

Based on the health risk assessment model and the reference values of element toxicity parameters, the non-carcinogenic hazard index and carcinogenic risk of adults exposed to the ten elements comprising PM_{2.5}, through the respiratory pathway in Wuxi city, were calculated. The human exposure concentration data for the ten elements are shown in Table 4. After 10,000 simulation tests, it was found that the exposure concentration of Al was significantly greater than that of the other elements ($0.199 \pm 0.081 \mu\text{g}/\text{m}^3$), followed by Pb and Mn. Owing to the presence of Al in the earth's crust, a variety of human production and life activities can release large amounts of Al, such as vehicle fuel, exhaust gas, decorative materials, batteries, indoor smoking, paint used for painting the walls, erosion and corrosion of automobile rubber; consequently, its concentration in the air is significantly greater than that of other elements (Kermani et al. 2021).

The US EPA recommends that the HQ of individual chemicals and the total non-carcinogenic HI should be less than one (US EPA, 1989), which indicates that the non-carcinogenic risk borne by the recipient is within an acceptable range. If it is greater than one, further the health risk assessment should be conducted. In the present health risk assessment, the total hazard index of nine elements was 0.743, with a range of 2.54E–05 to 5.33, indicating that the potential risk of exposure to a mixture of elements through respiratory should not be ignored. The individual HQ of each element were shown in Fig. 4a. Y-axis label

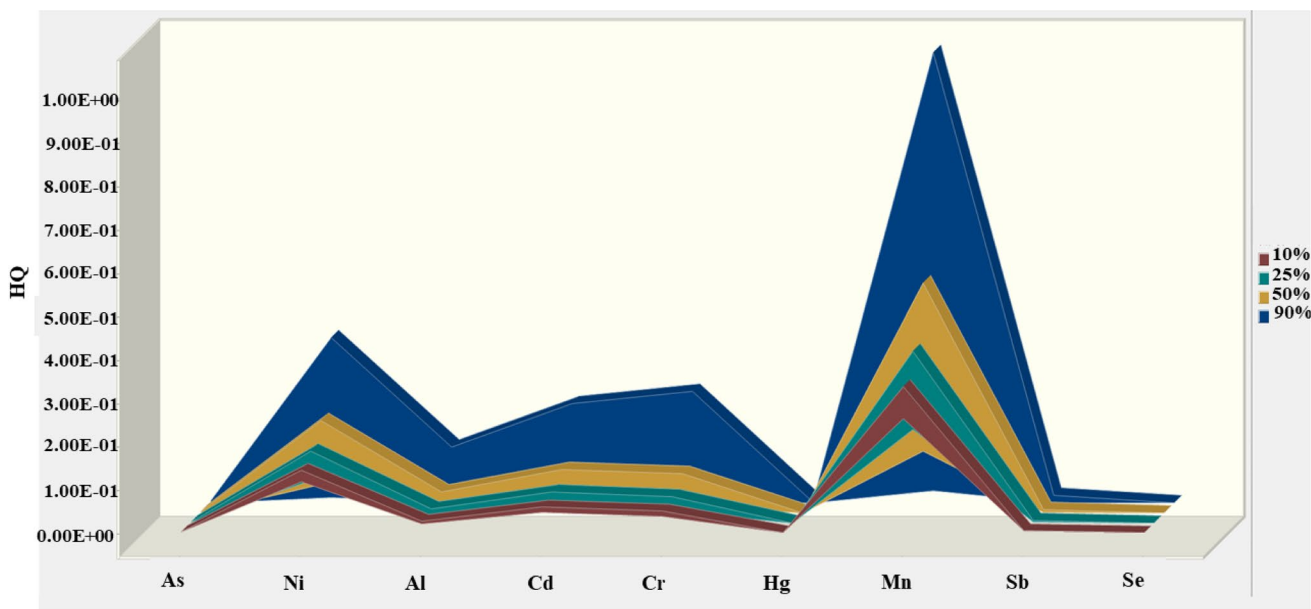
was the prediction value of HQ for each element, and the band width showed the amount of uncertainty at each percentile level with different colors. The non-carcinogenic risk of Mn was significantly greater than that of other elements with an average of 0.390, followed by Ni (0.154). Also, it must be noted out that the non-carcinogenic risk caused by oral and skin exposure is significantly higher than that caused by respiratory exposure. Direct oral ingestion is the main pathway of element exposure, and the risk of respiratory and inhalation exposure is less than that of the other two exposure pathways by a factor of 1/1000 or 1/10,000 (Wu et al. 2021). Therefore, the non-carcinogenic risk of human exposure to multiple environmental elements cannot be ignored.

In Fig. 4b, scatter charts show correlations, dependencies, and other relationships between pairs of forecasts and assumptions plotted against each other. The lower left to the upper right show positive relationships. Y-axis label is HI, and X-axis label is variable name which is related to HI forecast in present study. The *r* value means correlation between HI and variables. The sensitivity analysis showed that the correlation between exposure duration and the HI was the highest with a *r* value of 0.8797, and the correlation between elements and HI listed in descending order was Mn, Ni, Cr, Cd, Al, Se, Sb, Hg, and As.

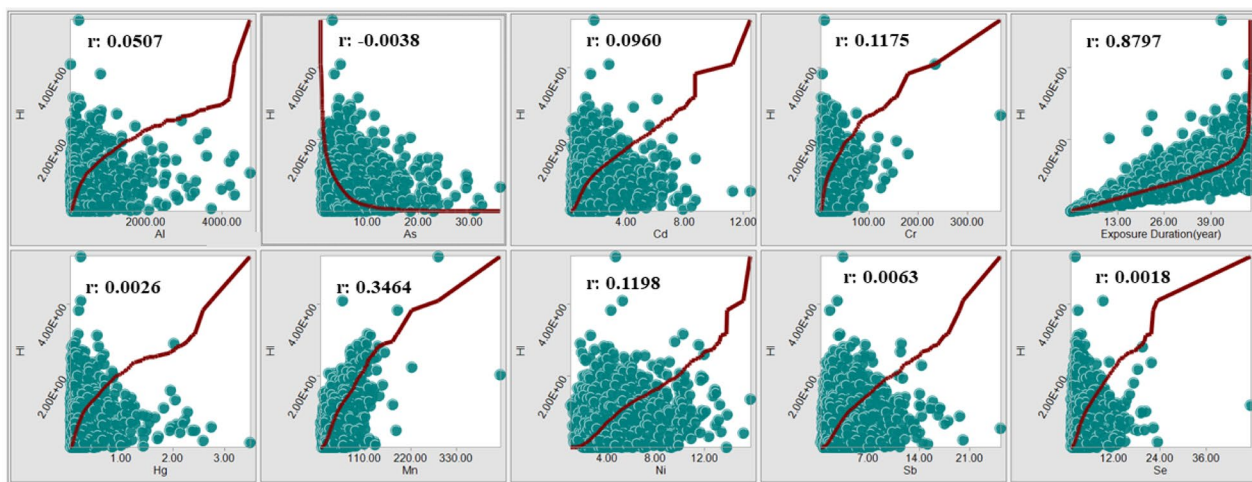
Furthermore, the Tornado chart tool in the Crystal Ball software was used to verify the results. The Tornado chart tool measures the impact of each model variable one at a time on a target forecast, which displays the result in two ways, the Tornado chart and Spider chart. In the Tornado chart, the variables listed higher have a greater predicted impact and variables listed near the bottom have smaller predicated oscillation. The Spider chart quantifies the slope of the curve obtained from the numerical tests of all

Table 4 Exposure concentration data for PM_{2.5}-bound elements

	Trials (10,000)	Average	Median	Variance	Min	Max	Range	Standard error
Carcinogenic effect	EC-As	1.97E–03	1.45E–03	3.83E–06	1.37E–08	3.69E–02	3.69E–02	1.96E–05
	EC-Cd	5.40E–04	3.65E–04	3.59E–07	2.34E–08	1.07E–02	1.07E–02	5.99E–06
	EC-Ni	1.53E–03	1.30E–03	1.41E–06	1.87E–08	1.05E–02	1.05E–02	1.19E–05
	EC-Pb	1.80E–02	1.32E–02	3.20E–04	3.93E–07	2.07E–01	2.07E–01	1.79E–04
Non-carcinogenic effect	EC-Al	1.99E–01	1.13E–01	8.11E–02	1.64E–06	5.08E+00	5.08E+00	2.85E–03
	EC-As	2.77E–03	2.04E–03	7.09E–06	5.99E–08	3.14E–02	3.14E–02	2.66E–05
	EC-Cd	7.58E–04	5.14E–04	6.86E–07	7.64E–08	1.09E–02	1.09E–02	8.28E–06
	EC-Cr	7.55E–03	4.32E–03	1.16E–04	9.77E–08	2.32E–01	2.32E–01	1.08E–04
	EC-Hg	1.26E–04	7.59E–05	2.70E–08	6.38E–09	2.26E–03	2.26E–03	1.64E–06
	EC-Mn	1.95E–02	1.51E–02	2.99E–04	5.69E–07	2.39E–01	2.39E–01	1.73E–04
	EC-Ni	3.02E–03	2.15E–03	2.70E–06	6.96E–08	1.17E–02	1.17E–02	1.64E–05
	EC-Pb	2.53E–02	1.86E–02	6.25E–04	8.15E–07	4.02E–01	4.02E–01	2.50E–04
	EC-Sb	1.85E–03	1.39E–03	3.00E–06	2.49E–08	2.94E–02	2.94E–02	1.73E–05
	EC-Se	2.04E–03	1.57E–03	3.72E–06	5.79E–08	4.15E–02	4.15E–02	1.93E–05



(a)



(b)

Fig. 4 HI of non-carcinogenic risk analysis of PM_{2.5}-bound elements exposure. **a** Non-carcinogenic risk profile for PM_{2.5}-bound elements. **b** Sensitivity analysis scatter plot

variables. The greater the slope, the greater the impact on prediction. The predicted results of this simulation were consistent with the scatter plot of sensitivity analysis, as shown in Fig. 5.

Considering that the carcinogenic toxicity was only confirmed by hexavalent Cr and the current study data focused on total Cr, the prediction of carcinogenic risk excluded Cr. As shown in Table 5, the average carcinogenic risk of other four carcinogens (As, Cd, Pb and Ni) was 1.01×10^{-5} , and the median value is 7.97×10^{-6} . According to the relevant reference value of EPA, the acceptable

range of carcinogenic risk level is between 10^{-6} and 10^{-4} , indicating that the carcinogenic risk of elements pollution in air was within the acceptable range. Among them, As was found the highest carcinogenic risk, with an average of 8.53×10^{-6} , followed by Cd (9.72×10^{-7}), Ni (3.71×10^{-7}), Pb (2.13×10^{-7}). It is worth noting that the carcinogenic risk value was only 2.25×10^{-9} in Tehran, Iran, 2016 (MohseniBandpi et al. 2018). And in the heating period of Baoding, located at the middle of Hebei province, China, the carcinogenic risk of As for male adult is 9.40×10^{-7} (Liang et al. 2019).

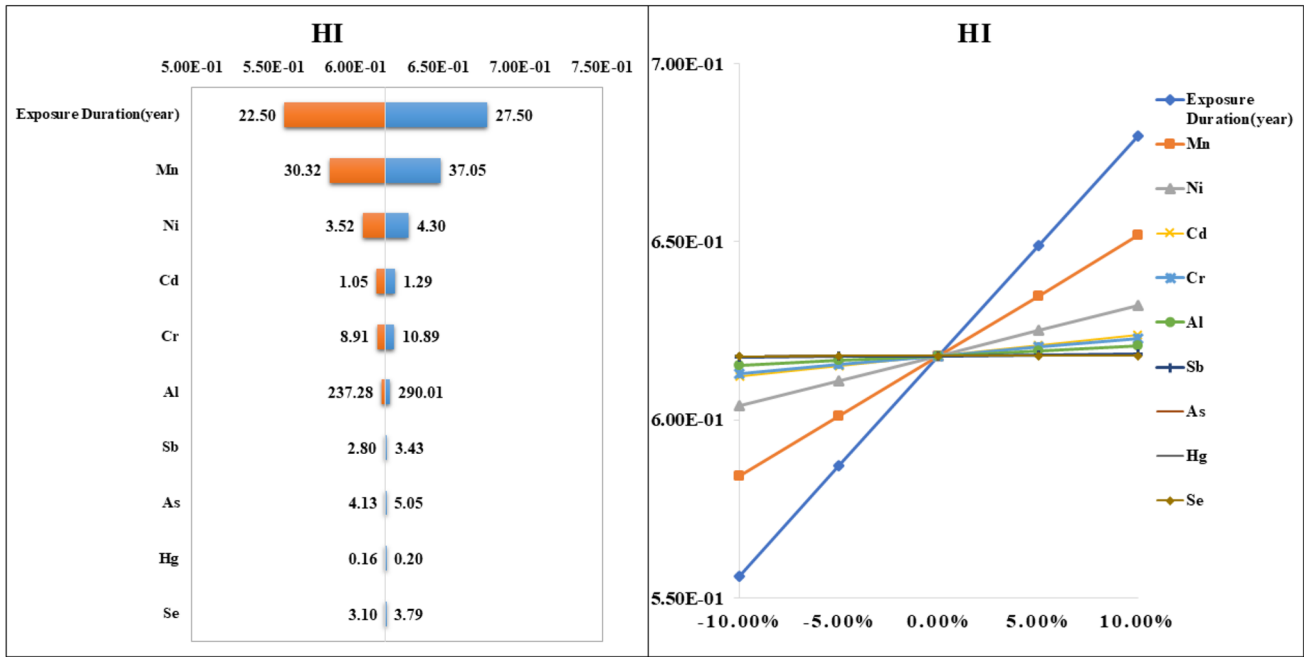


Fig. 5 Sensitivity analysis of non-carcinogenic risk analysis of PM_{2.5}-bound element exposure

Table 5 Carcinogenic risk of PM_{2.5}-bound elements

	CR	As	Cd	Ni	Pb
Average	1.01E-5	8.53E-06	9.72E-07	3.71E-07	2.13E-07
Median	7.97E-6	6.28E-06	6.66E-07	3.12E-07	1.59E-07
Variance	7.58E-11	6.54E-11	1.10E-12	8.31E-14	4.41E-14
Min	1.95E-9	1.68E-09	2.02E-10	4.37E-11	7.73E-12
Max	1.01E-4	9.19E-05	1.34E-05	2.99E-06	2.41E-06
Standard error	8.71E-8	8.09E-08	1.05E-08	2.88E-09	2.10E-09

Discussion

Ambient air pollution is a major environmental risk factor in the incidence of serious diseases, such as cardiovascular and respiratory diseases. PM_{2.5} is the predominant air pollutant since of its ability infiltrate deeply into the gas-exchange region of the lungs and cross the alveolar membrane into the blood vessels (He et al. 2010). China is known to be the largest developing country, with a heavy reliance on coal as a major energy source for maintaining essential industrial activities. A previous study showed an explicit causal relationship between coal burning and increasing PM_{2.5} in China (Cai et al. 2018). Generally, PM_{2.5} pollution in the Jing-Jin-Ji region is best characterized area for PM_{2.5} pollution in China. Long-term and large-scale monitoring studies have been conducted mainly in major cities such as Beijing, Guangzhou, Nanjing, and Shanghai. There have been

few studies regarding small-and medium-sized cities, and only simple statistical methods have been employed (Yin et al. 2020; Wang et al. 2019; Shen et al. 2016). Yu et al. reported PM_{2.5}, PM₁₀, SO₂, O₃, and NO₂ data from ten cities in the Yangtze River Delta between 2014 and 2017. Wuxi’s air quality was worse than that of other cities because 90% of the comprehensive energy consumption in Wuxi was occupied by heavy industry (Yu et al. 2020). A few relevant studies have been conducted in Wuxi, China.

In the current study, we performed a 5-year environmental surveillance of PM_{2.5} and 11 major elements from 2016 to 2020. Surveillance data were analyzed by calculating the temporal differences and spatial distribution compared to other regions of elements. Toxicological and carcinogenic evaluations were performed using the hazard index (HI) and carcinogenic risk (CR) quantities. The average PM_{2.5} concentration was 56.1 ± 31.0 µg/m³ with a range of 6.9 (August 13, 2020) to 181 µg/m³ (November 14, 2016). According to

the ambient air quality standards in China, the daily $PM_{2.5}$ concentration in the ambient air quality function zones Class II should not exceed $75 \mu\text{g}/\text{m}^3$ and Wuxi has yet to achieve the goal for the entire year. As for the respective recommended limits of daily $PM_{2.5}$, only 116 samples satisfied Class I ambient air quality function zones. The $PM_{2.5}$ content was greater than the WHO air quality guidelines of $25 \mu\text{g}/\text{m}^3$ by a factor of two. Our previous study found that the percentage increase caused an increase in the $PM_{2.5}$ -death rate twice as large among women than among men (Zhu et al. 2017). In addition, the $PM_{2.5}$ concentration was significantly greater during winter than during other seasons. This phenomenon was a result of the low boundary layer height and abundant $PM_{2.5}$ transported from North China by monsoon flows (Li et al. 2018). In contrast, we found that $PM_{2.5}$ pollution was severe during the Spring Festival as consequence of fireworks. Discharging fireworks during the Chinese Spring Festival celebrations is a deep-rooted custom in China. Although most cities have introduced policies to restrict the use of fireworks, the extensive firework displays during Spring Festival still result in a significant $PM_{2.5}$ concentration increase nationwide. This increase was 159–223% of the average level, indicating the instantaneous effect far exceeds that of any other factor over the whole year (Zhang et al. 2020). Therefore, prohibited fireworks are beneficial in protecting the environment.

Al was found to have the highest exposure concentration among all elements. It is well known that Al is the most common metal element and the third most common element behind oxygen and silicon in nature, accounting for 8.3% of the total weight of the Earth's crust. Owing to its excellent physical and chemical properties, Al is widely used in production and in daily life activities (Xu et al. 2021a, b). The Al $PM_{2.5}$ component was found to be high in other regions, such as Seoul, South Korea ($566 \pm 517 \text{ ng}/\text{m}^3$, Min: $98 \text{ ng}/\text{m}^3$, Max: $2535 \text{ ng}/\text{m}^3$); Alborz, Iran ($278.91 \text{ ng}/\text{m}^3$); and Wuhan, China ($113.41 \text{ ng}/\text{m}^3$) (Kang et al. 2004; Kermani et al. 2021; Wang et al. 2021a, b). However, Al levels were much lower in America, such as Ohio, USA ($42.1 \pm 28.2 \text{ ng}/\text{m}^3$) and New York, USA ($26.6 \pm 8.99 \text{ ng}/\text{m}^3$) (Brokamp et al. 2017; Ito et al. 2016). In addition, Al can be ingested in a variety of pathways other than respiration, such as milk formula for infants, and vaccination (Alasfar and Isaifan 2021). Since Siem first reported the neurotoxicity of Al in 1886, subsequent studies have confirmed the neurotoxicity of Al and its compounds. In recent years, elevated Al content has been detected in the brain tissues of patients with Alzheimer's disease, autism, and epilepsy (Ghosh et al. 2021; Skalny et al. 2021). Hence, overexposure to Al urges the need for more studies to explore mitigation actions, especially in Asia.

During the health risk evaluation and sensitivity analysis, Mn was found to have a greater non-carcinogenic risk, as shown in Figs. 4 and 5. Mn is a trace metal that is ubiquitous

in the environment and essential for maintaining human biochemistry and cellular reactions. Mn is also a cofactor for many enzymes, including glutamine synthase, arginase, pyruvate decarboxylase, and mitochondrial superoxide dismutase. However, the toxic effect of excessive Mn on the central nervous system has also been proven, and excessive accumulation of Mn in the central nervous system can cause neurotoxicity, leading to neurological and brain disorders (Wang et al. 2008; Lucchini et al. 2009; Sidoryk-Wegrzynowicz and Aschner 2013). In current study, Mn content in $PM_{2.5}$ samples was $37.4 \pm 23.3 \text{ ng}/\text{m}^3$, and was well below the WHO guideline of $150 \text{ ng}/\text{m}^3$. However, the non-carcinogenic risk posed by Mn cannot be ignored. A cohort study conducted by South Korea found that subtle structural changes in the brain may be induced by exposure to airborne pollutants such as, Mn in $PM_{2.5}$ with a content of $10.59 \pm 1.76 \text{ ng}/\text{m}^3$ (Jang et al. 2021). In addition, $PM_{2.5}$ -Mn exposure may be associated with greater depression symptomatology and is positively associated with the stimulated production of inflammatory mediators by stimulated immune cells, such as IL-1 β and TNF- α (Racette et al. 2021; Tripathy et al. 2021).

In summary, ambient $PM_{2.5}$ pollution is a critical issue in China and Asia. Several comprehensive environmental regulations have been initiated, and the low-carbon city pilot policy (LCCP) may have an impact on haze pollution. A study constructed multiple models to test the effect of the LCCP based on panel data from 271 cities in China from 2005 to 2018. The findings show that LCCP has significantly reduced the urban haze pollution, and the average annual concentration of $PM_{2.5}$ in pilot cities decreased by 14.29% (Yan et al. 2021). Another study showed that China's Emissions Trading Scheme reduced $PM_{2.5}$ concentrations by 4.8% using monthly $PM_{2.5}$ concentration and weather data for 297 Chinese cities from January 2005 to December 2017. This reduction effect was most significant during summer (Liu et al. 2021a, b). In addition, to increase the efficiency of air filters in reducing PM emissions, metal-organic frameworks (MOFs, here, Zr-MOFs, especially with functional groups (FGs) such as $-\text{NO}_2$) were coated when MOFs have FGs that can achieve large charge separation (Woo et al. 2020). With the effective reduction of $PM_{2.5}$ emissions, the associated elements were also well controlled.

Conclusions

This study was the first to carry out long-term monitoring of various elements in atmospheric $PM_{2.5}$, and the results showed that $PM_{2.5}$ and element pollution concentrations in ambient-air within Wuxi have been decreasing from 2016 to 2020. $PM_{2.5}$ pollution had significant temporal and spatial distribution variation. The numbers of $PM_{2.5}$ and elemental samples which exceeded guideline limits were greater in

winter and spring than in other seasons. The total content of 11 elements was $0.38 \pm 0.33 \mu\text{g}/\text{m}^3$, and no other elements exceeded the limit except for As in 2016. The mean value of the hazard index caused by exposure to the nine elements through the respiratory tract was 0.743, and the carcinogenic risk was 1.01×10^{-5} . The health risk values were within acceptable guidelines ranges. However, considering that humans are exposed to elements in a variety of combinations, the potential risk of long-term exposure to Al, Mn, and As of atmospheric $\text{PM}_{2.5}$ should not be ignored. Therefore, the $\text{PM}_{2.5}$ and element pollution levels in Wuxi should be closely monitored for the foreseeable future, especially in winter and spring.

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Data availability Not applicable.

Declarations

Ethical approval Not applicable.

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