



# Pollution characteristics and human health risks of PM<sub>2.5</sub>-bound heavy metals: a 3-year observation in Suzhou, China

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**Abstract** This study aimed to analyze the temporal trends, pollution levels, and health risks associated with eleven PM<sub>2.5</sub>-bound heavy metals (Sb, Al, As, Hg, Cd, Cr, Mn, Ni, Pb, Se and Tl). A total of 504 PM<sub>2.5</sub> samples were collected in Suzhou from January 2019 to December 2021. The pollution levels were estimated based on enrichment factors (EFs) which can be used to calculate the enrichment of heavy metals in PM<sub>2.5</sub> and determine whether the concentrations of PM<sub>2.5</sub>-bound heavy metals are influenced by the crustal or anthropogenic sources, and the health risk of PM<sub>2.5</sub>-bound heavy metals via inhalation was assessed following US EPA's Risk Assessment Guidance for Superfund (RAGS). The annual average concentration of PM<sub>2.5</sub> was 46.76 µg m<sup>-3</sup>, which was higher than the WHO recommended limit of 5 µg m<sup>-3</sup>. The average of the sum of eleven PM<sub>2.5</sub>-bound heavy metals was 180.61 ng m<sup>-3</sup>, dominated by Al, Mn, and Pb. The concentration of PM<sub>2.5</sub> in 2020 was significantly lower than that in 2019 and

2021. The PM<sub>2.5</sub> and PM<sub>2.5</sub>-bound heavy metal concentrations in winter and spring were significantly higher than those in autumn and summer. The EF of As, Cr, Cd, Hg, Ni, Pb, Sb, Mn, Se, and Tl was higher than 10, indicating they were mainly from anthropogenic sources. Exposure to a single non-carcinogenic heavy metal via inhalation was unlikely to cause non-carcinogenic effects (HQ < 1), but the integrated non-carcinogenic risks should be taken seriously (HI > 1). The cumulative carcinogenic risks from the carcinogenic elements were exceeding the lower limit (1 × 10<sup>-6</sup>) of the acceptable risk range. The carcinogenic risks of As and Cr(VI) contributed 60.98% and 26.77%, respectively, which were regarded as two key carcinogenic risk factors. Overall, the government policies and countermeasures for the PM<sub>2.5</sub> pollution control should be performed not only based on the PM<sub>2.5</sub> concentration but also based on the PM<sub>2.5</sub>-bound heavy metals and their health risks for the local residents.

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## Introduction

Ambient particulate matter pollution, especially particulate matter with a diameter of 2.5 µm or less (PM<sub>2.5</sub>) pollution, is one of the most serious environmental problems to be addressed worldwide

(Kasimov et al., 2020; Onishi et al., 2012; Zhang et al., 2020). The data from the Global Burden of Diseases Study have shown that long-term exposure to ambient  $PM_{2.5}$  was ranked fifth among the risk factors contributing to global deaths in 2015 (Cohen et al., 2017). Approximately 86% of urban inhabitants (2.5 billion) lived in urban areas where the  $PM_{2.5}$  concentration exceeds WHO's 2005 guideline annual average  $PM_{2.5}$  ( $10 \mu\text{g m}^{-3}$ ), resulting in an excess of 1.8 million deaths in 2019 (Southerland et al., 2022). Because of its small particle size, good chemical stability, and complex harmful chemical compositions,  $PM_{2.5}$  can be easily absorbed by the human pulmonary alveoli and blood through respiratory system, then has a severe negative impact on human health (Cao et al., 2012), such as increasing the risk of premature death from respiratory diseases (Wang et al., 2021a), cardiovascular diseases (Grande et al., 2020; Wang et al., 2015), lower respiratory infection (Horne et al., 2018), and lung cancer (Hamra et al., 2014). In addition to exposure to high concentration of  $PM_{2.5}$ , exposure to low pollution levels below the current WHO guideline values also has a negative impact on public health. For example, Strak et al. (2021) have shown that an increase of  $5 \mu\text{g m}^{-3}$   $PM_{2.5}$  above the US standard of  $12 \mu\text{g m}^{-3}$  was associated with 13% increase in natural deaths of participants while a  $5 \mu\text{g m}^{-3}$   $PM_{2.5}$  increase below the same US standard was associated with 29.6% increase in natural death rate. Hence, with rapid economic development and acceleration of urbanization, exposure to  $PM_{2.5}$  pollution has become a serious concern globally.

Due to its large specific surface area,  $PM_{2.5}$  may act as a fine carrier of various chemical components mainly including heavy metals, polycyclic aromatic hydrocarbons, element carbon, water-soluble inorganic ions, organic matter, and so on (Cheng et al., 2015; Zhang et al., 2021). Among them, heavy metals, such as Sb, Al, As, Hg, Cd, Cr, Mn, Ni, Pb, Se, and Tl, have induced great interest because of their complicated sources, high toxicity, biological accumulation, and persistence. Excessive exposure to  $PM_{2.5}$ -bound heavy metals via the pathways of inhalation, dermal contact, and ingestion could threaten human health by penetrating respiratory system and accumulating in different tissues and organs (Kim et al., 2015; Sacks et al., 2011; Tsai et al., 2020). In recent years, with the implementation of the Action Plan for Air Pollution Prevention and Control

(Chai, 2020), the concentration of  $PM_{2.5}$  has presented a downward trend all the time, especially in China (Yang et al. 2020). In China, the annual average  $PM_{2.5}$  concentrations have decreased from 61.8 to  $42.0 \mu\text{g m}^{-3}$  from 2013 to 2017 (Zhang et al. 2019). China will be able to achieve  $PM_{2.5}$  air quality annual standard ( $35 \mu\text{g m}^{-3}$ ) by 2030 by fulfilling its National Determined Contributions pledges and continuing air pollution control policies (Cheng et al., 2021). However, a low concentration of  $PM_{2.5}$  does not signify a low health risk (Strak et al., 2021). In particular, the  $PM_{2.5}$ -bound heavy metals, including As, Cr(VI), and Cd, have been classified as group 1 carcinogenic agents, Pb and Ni have been classified as group 2a carcinogenic agents by the International Agency for Research on Cancer (2016). These carcinogens have no thresholds on adverse health effects and are harmful to humans even at very low concentrations. To some extent, the characteristics and health risk assessment of  $PM_{2.5}$ -bound heavy metals can offer more valuable information than the analysis of  $PM_{2.5}$  concentration to the policymaker. Hence, it is necessary to renew the assessment about the human health risks of  $PM_{2.5}$ -bound heavy metals in China.

As a famous tourist city with high economic and industrial activities in the Yangtze River Delta (YRD) of China, Suzhou had a permanent residential population of 12.84 million people and 4.79 million vehicles in 2021. According to our knowledge, few investigations of the characteristics and health risk assessment of atmospheric  $PM_{2.5}$ -bound heavy metals in Suzhou have been conducted recently. Given that emission sources, meteorological patterns, and economic structures differ in different cities of China, the characteristics and health risk levels of atmospheric  $PM_{2.5}$ -bound heavy metals were distinct in each city (Wang et al., 2021b). As one of the cities with a low  $PM_{2.5}$  concentration in China, the risk levels of  $PM_{2.5}$ -bound heavy metals in Suzhou were worthy of in-depth investigation. In this study,  $PM_{2.5}$  samples were collected in two representative regions including one urban district and one county-level city of Suzhou from January 2019 to December 2021, with the main objectives as follows: (1) to investigate the temporal distribution characteristics and the exceedance of  $PM_{2.5}$  and 11  $PM_{2.5}$ -bound heavy metals during 2019 and 2021 in Suzhou; and (2) to assess the carcinogenic and noncarcinogenic risk posed to the general population via inhalation. Our study about  $PM_{2.5}$  and

PM<sub>2.5</sub>-bound heavy metals could provide scientific basis for the formulation of targeted pollution-control and risk-mitigation strategies to improve the ambient air quality and the public health of Suzhou, China.

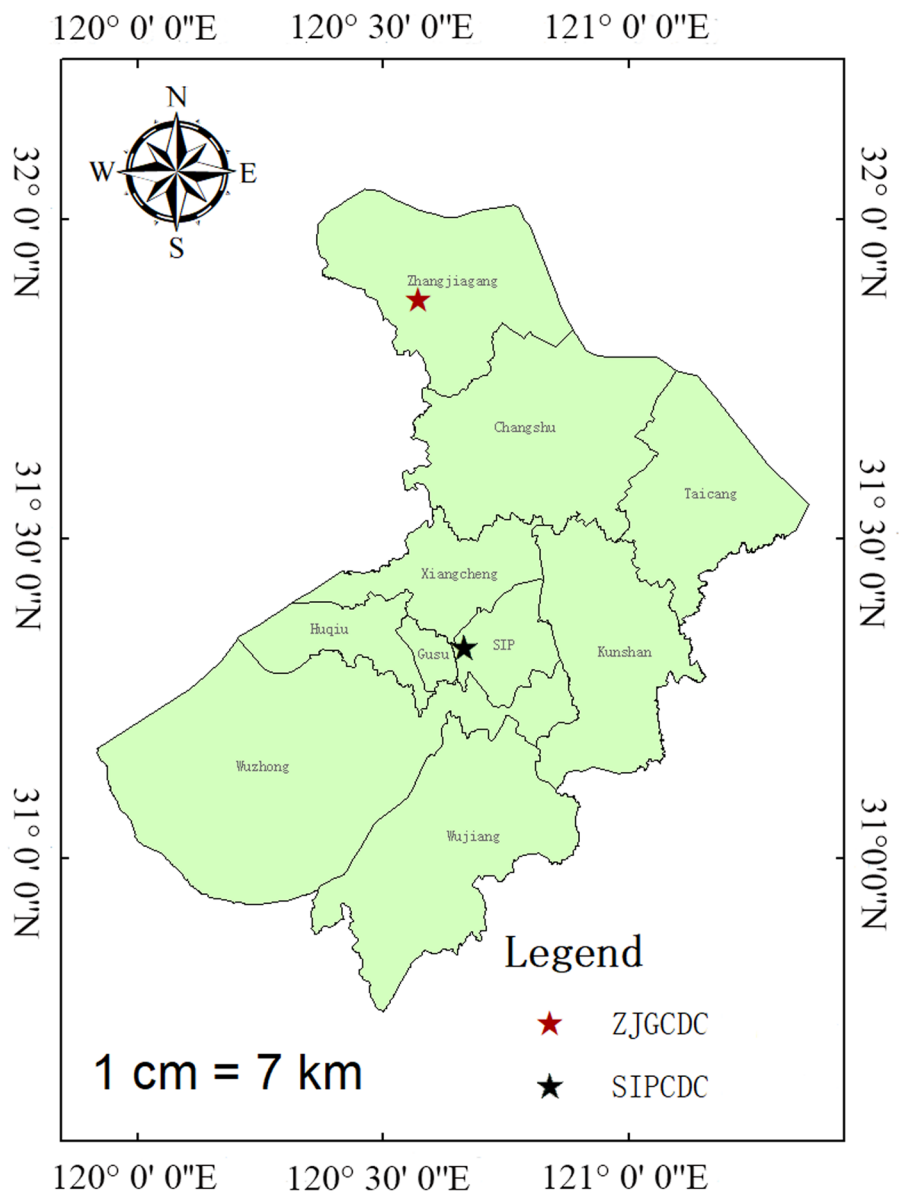
**Methodology**

**Sampling sites**

Two representative sampling sites were chosen in Suzhou, including one urban district and one

county-level city, as shown in Fig. 1. As one of the six urban districts of Suzhou, Suzhou Industrial Park has become one of the regions with the highest degree of openness, the best quality and efficiency of development and the best business environment in China, so this district could represent one type of the atmospheric environment in Suzhou. The sampling point in Suzhou Industrial Park was located on Suzhou Industrial Park Center for Disease Control and Prevention (31.33°N, 120.66°E), which was away from the major roads and surrounded by industrial and residential areas, but there were no serious industrial pollution

**Fig. 1** Locations of sampling sites in Suzhou, China (Notes: ZJGCDC: Zhangjiagang Municipal Center for Disease Control and Prevention; SIPCDC: Suzhou Industrial Park Center for Disease Control and Prevention.)



sources within two kilometers. Another sampling site was set on Zhangjiagang Municipal Center for Disease Control and Prevention (31.87°N, 120.57°E) in Zhangjiagang city which was one of the four county-level city of Suzhou. Likewise, this site was away from the major roads but surrounded by residential and commercial areas. The above two sampling sites both were set at the building rooftops approximately fifteen meters above the ground level, and near the national or municipal control point of China Environmental Monitoring Station.

### PM<sub>2.5</sub> sampling

PM<sub>2.5</sub> samples were collected simultaneously at two sampling sites from the 10th to the 16th of every month for three years from January 2019 to December 2021, and the collection duration for each sample was 22 h starting at 9 a.m every day. PM<sub>2.5</sub> and PM<sub>2.5</sub>-bound heavy metals were collected onto 90 mm quartz fiber filters (Whatman, UK) by a mid-volume PM<sub>2.5</sub> flow sampler (TH-150D, Tianhong Instrument Co., Ltd., Wuhan, China) at a flow rate of 100 L min<sup>-1</sup>. Blank control samples and parallel control samples were obtained once every month at two sampling sites, respectively. Meteorological parameters, such as temperature, barometric pressure and relative humidity were recorded at the time of sample collection. A total of 504 valid PM<sub>2.5</sub> samples were collected at two sampling sites from 2019 to 2021. All collected filters were subsequently stored at -18 °C for later analysis.

### PM<sub>2.5</sub>-bound heavy metals analysis and quality assurance/quality control (QA/QC)

The filters were equilibrated under constant temperature (22 °C ± 1 °C) and constant relative humidity (30% ± 5%) for 24 h before and after being weighed by a one ten-thousandth ultra-microbalance (FA2004, Tianping Instrument Co., Ltd., Shanghai, China). A quarter of the sampling filter was cut up by the ceramic scissors and then placed in a 50-mL centrifuge tube and extracted by concentrated nitric acid (65% GR grade, 20 mL). The tube was then incubated in a water bath at 70 °C with ultrasonic extraction for 3 h. After cooling down to the room temperature, the sample extract was filtrated through a 0.45 mm pore size hydrophilic membrane. The tube was rinsed

thrice with ultrapure water, and the rinsing solution was also filtrated the same way as the extract. This final extract was stored in a metal-free polyethylene bottle at 4 °C for subsequent analysis. Finally, inductively coupled plasma mass spectrometry (ICP-MS) (X-Series II, Thermo Scientific, USA) was chosen to detect the eleven PM<sub>2.5</sub>-bound heavy metals including Sb, Al, As, Hg, Cd, Cr, Mn, Ni, Pb, Se and Tl. The specific detected methods for the mass concentrations of PM<sub>2.5</sub> and PM<sub>2.5</sub>-bound heavy metals were referred to the relevant standard documents (MEEPRC, 2011, 2013).

QA/QC included blank control trial, parallel control trial and recovery analysis of the standard reference material. The detected values of all heavy metals in blank control samples which obtained every month were lower than their method detection limits, respectively. The relative errors between the detected values of all heavy metals in the samples and the parallel control samples obtained once every month at the same time were less than 10%. The certified standard mix of the twelve heavy metals was spiked on the acid-washed blank filters. Then the element recovery percentage from the standard reference material was between 82 and 110%.

### Enrichment factor (EF)

The EF method can be used to calculate the enrichment of heavy metals in PM<sub>2.5</sub> and determine whether the concentrations of PM<sub>2.5</sub>-bound heavy metals are influenced by the crustal or anthropogenic sources. The EF values are calculated by Eq. (1) (Xu et al., 2021) below:

$$EF_i = \frac{(C_i/C_{Al})_{\text{sample}}}{(C_i/C_{Al})_{\text{crust}}} \quad (1)$$

where  $C_i$  represents the concentration of each heavy metal;  $C_{Al}$  represents the concentration of Al which was chose as the reference element in this study because of its high concentration in crustal dust, and relatively less contribution from other sources (Arimoto et al., 2006);  $(C_i/C_{Al})_{\text{sample}}$  is the ratio of the concentration of heavy metal  $i$  and Al in aerosol samples, and  $(C_i/C_{Al})_{\text{crust}}$  is the ratio of their concentrations in the upper continental crust. As reported by China's National Environmental Monitoring Centre (CNEMC, 1990), the concentrations of heavy metals

in the upper continental crust in Suzhou city were 7.64% for Al, and 83, 700, 35.2, 9.2, 0.082, 23.9, 0.14, 0.65, 0.06, and 0.77 mg kg<sup>-1</sup> for Cr, Mn, Ni, As, Cd, Pb, Se, Tl, Hg, and Sb, respectively. When the EF is lower than 10, it indicates that the PM<sub>2.5</sub>-bound heavy metal is mainly influenced by natural sources and crustal sources, when the EF is greater than 10, it indicates that the PM<sub>2.5</sub>-bound heavy metal is mainly influenced by anthropogenic sources.

### Health risk assessment

According to the related studies of Intergrated Risk Information System (IRIS) and IARC, pollutants can be divided into non-carcinogens and carcinogens. In this study, the PM<sub>2.5</sub>-bound heavy metals of Cr(VI), Mn, Ni, As, Cd, Pb, Se, Tl, Hg and Sb are non-carcinogens, and As, Cd, Cr(VI), Pb, Ni have carcinogenic effects on the public, simultaneously. The Cr (VI) was considered as 1/7th of Cr (Peter et al., 2018) Because inhalation is the major pathway of residential exposure to PM<sub>2.5</sub>-bound heavy metals (Li et al., 2015), the health risks from inhalation exposure to PM<sub>2.5</sub>-bound heavy metals for the general population in Suzhou were estimated following US EPA’s Risk Assessment Guidance for Superfund (RAGS) (USEPA, 2009) and the relevant Chinese standard document (WS/T 777-2021) (NHCPRC, 2021) in this study. According to the RAGS, the average daily doses (ADD) of non-carcinogenic heavy metals and the life-time average daily doses (LADD) of carcinogenic heavy metals via inhalation are calculated by Eq. (2) and Eq. (3), respectively:

$$ADD = (EC \times EF \times ED \times ET) / AT_{nc} \tag{2}$$

$$LADD = (EC \times EF \times ED \times ET) / AT_{ca} \tag{3}$$

where ADD was the average daily dose (mg/m<sup>3</sup>); EC was the exposure concentration of PM<sub>2.5</sub>-bound heavy metals (mg m<sup>-3</sup>). The EC of a heavy metal in PM<sub>2.5</sub> used the annual mean concentration; however, according to EPA’s recommendation, the 95% upper confidence limit (95%UCL) of the EC was adopted to yield a conservative exposure estimate in this study. EF was the exposure frequency (365 days per year was chosen in this study); ED was the exposure duration (30 years for adult was considered in this study); ET was the exposure time (24 h/day); AT<sub>nc</sub>

was the averaging time for non-carcinogens which was equal to ED×365 days×24 h, and AT<sub>ca</sub> was the averaging time for carcinogens which was equal to 80 years×365 days×24 h in this study.

Then, the non-carcinogenic risk of a heavy metal was evaluated with the hazard quotient (HQ) which is calculated by Eq. (4):

$$HQ = ADD/RfC \tag{4}$$

where RfC was the reference concentration for inhalation (mg m<sup>-3</sup>). The RfC values of the non-carcinogenic heavy metals are shown in Table 1. If HQ is less than 1, it indicates that the heavy metal’s non-carcinogenic risk is unlikely or negligible; If HQ is greater than 1, it implies that the heavy metal may have non-carcinogenic effect on the public (Mohm- and et al., 2015).

The carcinogenic risk of a heavy metal via inhalation pathway was evaluated with the carcinogenic risk (CR) which is calculated by Eq. (5):

$$CR = LADD \times IUR \times 10^3 \tag{5}$$

where IUR was the inhalation unit risk [(μg m<sup>-3</sup>)<sup>-1</sup>]. The IUR values of the carcinogenic heavy metals are shown in Table 1. A CR < 10<sup>-6</sup> indicates the carcinogenic risk is negligible, and a CR < 10<sup>-4</sup> and > 10<sup>-6</sup> indicates the carcinogenic risk is acceptable, and a CR > 10<sup>-4</sup> indicates the carcinogenic risk exceeds the acceptable risk range (Hou et al., 2019).

## Results and discussion

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

#### *The annual mean concentration of PM<sub>2.5</sub>*

In order to control air pollution, the China National Ambient Air Quality Standards (CNAAQS) has been developed by China through the local situation of air quality and economic development in 2012. The limit values of PM<sub>2.5</sub> were set to control PM<sub>2.5</sub> pollution in two interim target stages including the secondary standard concentration limit of 75 μg m<sup>-3</sup>, defined as CNAAQS Interim Target 2 (CNAAQS IT-2), and the primary standard concentration limit of 35 μg m<sup>-3</sup>, defined as CNAAQS Interim Target 1 (CNAAQS IT-1). The annual mean concentration of PM<sub>2.5</sub> in

**Table 1** Specific RfC and IUR values of PM<sub>2.5</sub>-bound heavy metals via inhalation

Elements	Non-carcinogen		Carcinogen	
	RfC (mg m <sup>-3</sup> )	Data source <sup>a</sup>	IUR [(μg m <sup>-3</sup> ) <sup>-1</sup> ]	Data source
Sb	3.0 × 10 <sup>-4</sup>	ATSDR	– <sup>b</sup>	–
Al	5.0 × 10 <sup>-3</sup>	PPRTVs	–	–
As	1.5 × 10 <sup>-5</sup>	CALEPA	4.3 × 10 <sup>-3</sup>	IRIS
Cd	1.0 × 10 <sup>-5</sup>	ATSDR	1.8 × 10 <sup>-3</sup>	IRIS
Cr(VI)	1.0 × 10 <sup>-4</sup>	IRIS	1.2 × 10 <sup>-2</sup>	IRIS
Hg	3.0 × 10 <sup>-4</sup>	IRIS	–	–
Pb	1.5 × 10 <sup>-4</sup>	NAAQS	8.0 × 10 <sup>-5</sup>	CALEPA
Mn	5.0 × 10 <sup>-5</sup>	IRIS	–	–
Ni	5.0 × 10 <sup>-5</sup>	ATSDR	2.4 × 10 <sup>-4</sup>	ATSDR
Se	2.0 × 10 <sup>-2</sup>	CALEPA	–	–
Tl	–	–	–	–

<sup>a</sup> RfC and IUR were cited from Integrated Risk Information System (IRIS), California Environmental Protection Agency (CALEPA), Agency for Toxic Substances and Disease Registry (ATSDR), US EPA's National Ambient Air Quality Standard (NAAQS) or Provisional Peer-Reviewed Toxicity Values (PPRTVs)

<sup>b</sup> There are no specific studies showing that the element has carcinogenic risk or non-carcinogenic risk

Suzhou during 2019 and 2021 was  $46.76 \pm 29.84 \mu\text{g}/\text{m}^3$  which was lower than the concentration limit of CNAAQs IT-2, but higher than the concentration limit of CNAAQs IT-1 (MEEPRC, 2012) and the WHO recommended limit of  $5 \mu\text{g}/\text{m}^3$  in the WHO global air quality guidelines in 2021 which is defined as AQG2021 (WHO, 2021) (Table 2). However, as a city in YRD of China, the concentration of PM<sub>2.5</sub> in Suzhou was lower than the other cities in YRD, such as Hangzhou ( $63.0 \mu\text{g}/\text{m}^3$ ) (Xu et al., 2021) and Changzhou ( $50.5 \mu\text{g}/\text{m}^3$ ) (Yi et al., 2022). The annual mean concentrations of PM<sub>2.5</sub> from 2019 to 2021 were  $51.24 \pm 30.06$ ,  $45.15 \pm 33.52$ , and  $43.89 \pm 24.97 \mu\text{g}/\text{m}^3$ , respectively. The concentration of PM<sub>2.5</sub> in 2019 was significantly higher than that in 2020 and 2021. In order to control the spread of the 2019 novel coronavirus (COVID-19) which broke out intermittently during 2020 and 2021, the lockdown policy, such as going out of business and production, was implemented in Suzhou which reduced the emission of PM<sub>2.5</sub> (Wang et al. 2021b). As shown in Table 2 and Fig. 3, The seasonal variation of PM<sub>2.5</sub> concentrations showed that the concentrations of PM<sub>2.5</sub> in Suzhou during 2019 and 2021 were significantly higher in winter and spring than that in summer and autumn. The results were similar to the data reported by Wang et al. (2018). The reason might be ascribed to the increase in air pollutant emissions

and the suitable meteorological conditions in winter and spring. As the outdoor ambient temperature decreases, coal consumption by local heating demand increases simultaneously, and a large amount of air pollutants generated by fossil fuel combustion and traffic emission is released into the atmosphere. In addition, the meteorological conditions including low temperature, low relative humidity, low wind speed, low precipitation and high atmospheric pressure were not conducive to the dispersion of these air pollutants (Hong et al., 2013). It was also confirmed in this study that temperature, relative humidity, wind speed and precipitation were negatively correlated with PM<sub>2.5</sub> concentrations ( $P < 0.05$ ), and atmospheric pressure was positively correlated with PM<sub>2.5</sub> concentrations ( $P < 0.05$ ) (Table 4).

#### Exceedance of PM<sub>2.5</sub>

At present, the concentration limit of CNAAQs IT-2 is the minimum requirement for enforcement in China. Meanwhile, the best limit value of  $5 \mu\text{g}/\text{m}^3$  for the public's health is recommended by WHO (2021). In this study, an exceedance is defined when the concentration of PM<sub>2.5</sub> exceeds the air quality standards or limit values.

In this section, we have analyzed the PM<sub>2.5</sub> concentrations in Suzhou during 2019 and 2021 to

**Table 2** Annual mean concentration ( $\mu\text{g m}^{-3}$ ) and percentage exceedance (%) of  $\text{PM}_{2.5}$  in Suzhou from 2019 to 2021

Time <sup>a</sup>	Mean concentration (Mean $\pm$ SD)	Percentage exceedance <sup>b</sup>			
		CNAAQS IT-2	CNAAQS IT-1	AQG2021	
2019	51.24 $\pm$ 30.06 (All year (n = 168))	16.07(51.24 $\pm$ 30.06)	65.48(16.07)	100.00(65.48)	100.00
	Spring (n = 42) <sup>c</sup>	63.43 $\pm$ 32.14	21.43	92.86	100.00
	Summer (n = 42)	45.16 $\pm$ 21.86	7.14	64.29	100.00
	Autumn (n = 42)	42.33 $\pm$ 30.93	14.29	40.48	100.00
	Winter (n = 42)	54.05 $\pm$ 30.56	21.43	64.29	100.00
2020	45.15 $\pm$ 33.52 (All year(n = 168))	14.88 (45.15 $\pm$ 33.52)	51.79 (14.88)	100.00 (51.79)	100.00
	Spring (n = 42)	53.88 $\pm$ 24.87	21.43	76.19	100.00
	Summer (n = 42)	37.76 $\pm$ 20.52	4.76	52.38	100.00
	Autumn (n = 42)	27.79 $\pm$ 10.18	0.00	26.19	100.00
	Winter (n = 42)	61.19 $\pm$ 52.22	33.33	52.38	100.00
2021	43.89 $\pm$ 24.97 (All year(n = 168))	14.88 (43.89 $\pm$ 24.97)	56.55 (14.88)	99.40 (56.55)	99.40
	Spring (n = 42)	51.81 $\pm$ 27.05	23.81	64.29	100.00
	Summer (n = 42)	36.81 $\pm$ 19.93	2.38	47.62	100.00
	Autumn (n = 42)	31.17 $\pm$ 22.74	4.76	35.71	97.62
	Winter (n = 42)	55.79 $\pm$ 21.63	28.57	78.57	100.00
2019–2021	46.76 $\pm$ 29.84 (All year(n = 504))	15.28 (46.76 $\pm$ 29.84)	57.94 (15.28)	99.80 (57.94)	99.80
	Spring (n = 126)	56.37 $\pm$ 28.42	22.22	77.78	100.00
	Summer (n = 126)	39.91 $\pm$ 20.96	4.76	54.76	100.00
	Autumn (n = 126)	33.76 $\pm$ 23.59	6.35	34.13	99.21
	Winter (n = 126)	57.01 $\pm$ 36.92	27.78	65.08	100.00

<sup>a</sup>Spring is from March to May, summer is from June to August, autumn is from September to November, and winter is from December to February

<sup>b</sup>The secondary standard concentration limit of the China National Ambient Air Quality Standards (CNAAQS IT-2) is  $75 \mu\text{g m}^{-3}$ ; the primary standard concentration limit of the China National Ambient Air Quality Standards (CNAAQS IT-1) is  $35 \mu\text{g m}^{-3}$ ; and the WHO recommended limit (AQG2021) is  $5 \mu\text{g m}^{-3}$

<sup>c</sup>The number of valid samples

study the number of threshold exceedances of daily mean  $\text{PM}_{2.5}$  concentrations as per the limit values of CNAAQS IT-1 and IT-2, and the limit values of AQG2021, respectively. The average number of percentage exceedances for each year and season are summarized in Table 2. As can be seen from Table 2, in terms of CNAAQS IT-2 and IT-1, and AQG2021, the percentage exceedances during 2019 to 2021 were 15.28%, 57.94% and 99.80%, respectively. The percentage exceedance in terms of the AQG2021 was more than six times higher than that in terms of CNAAQS IT-2. According to all the three concentration limit values of  $\text{PM}_{2.5}$ , the

percentage exceedances were significantly higher in 2019 than that in 2020 and 2021, and the percentage exceedances were significantly higher in winter and spring than that in summer and autumn. The above results showed that the number of exceedances increases as the threshold was decreased for each year and season, and the variation trends of  $\text{PM}_{2.5}$  exceedances were consistent with the  $\text{PM}_{2.5}$  concentrations. Burnett et al. (2018) have reported that it still has the risk of lower respiratory tract infection, stroke and other diseases when the public expose to low concentration of  $\text{PM}_{2.5}$  for a long time. Zhang et al. (2019) proved that it had been

achieved significant health benefits when the annual mean concentration of PM<sub>2.5</sub> in China decreased from 61.8 µg m<sup>-3</sup> in 2013–42.0 µg m<sup>-3</sup> in 2017. Furthermore, they also predicted that the number of premature deaths caused by exposure to related pollution would be significantly reduced when the annual concentration of PM<sub>2.5</sub> reached the WHO recommended limit of 5 µg m<sup>-3</sup> in China.

#### Concentrations of PM<sub>2.5</sub>-bound heavy metals in Suzhou

The annual mean concentrations of PM<sub>2.5</sub>-bound heavy metals in Suzhou during 2019 and 2021 are summarized in Table 3.  $\Sigma$ Heavy metals, defined as the sum concentration of eleven heavy metals, was observed with a annual mean concentration of 180.61 ± 176.95 ng m<sup>-3</sup> and accounted for 0.47% of PM<sub>2.5</sub> concentration in Suzhou. (Table 4)  $\Sigma$ Heavy metals was dominated by Al, Mn, and Pb in this city. The annual mean concentrations of the three dominant metals were 109.77 ± 156.00, 30.48 ± 24.73, and 21.92 ± 18.94 ng m<sup>-3</sup>, respectively, the sum of which accounted for 89.79% of  $\Sigma$ Heavy metals. The highest concentration of Al among these heavy metals suggested a predominant contribution from crustal sources such as fugitive dust to the fine aerosols in Suzhou (Ma et al., 2019; Xu et al., 2021). While the dominance of Mn and Pb could be attributed to coal combustion (Deng et al., 2014) and traffic emissions

including exhaust and non-exhaust emissions (Charron et al., 2018). Other heavy metals were ranked as: Cr > Sb > As > Ni > Se > Cd > Hg > Tl. The annual mean concentrations of Mn, Pb, As, Cd, Se, Tl, Sb were lower than those measured in Hangzhou, but the annual mean concentrations of Al, Cr, Hg, Ni were higher than that in Hangzhou (Wang et al., 2018). According to the China National Ambient Air Quality Standards, the concentrations of As, Pb, Cd, Hg in Suzhou were 3.51 ng m<sup>-3</sup>, 21.92 ng m<sup>-3</sup>, 0.71 ng m<sup>-3</sup>, 0.36 ng m<sup>-3</sup>, respectively, which were significantly lower than the China standards of 6 ng m<sup>-3</sup>, 500 ng m<sup>-3</sup>, 5 ng m<sup>-3</sup>, 50 ng m<sup>-3</sup>, respectively. This indicated the effectiveness of energy-saving and emission-reduction in Suzhou. However, it was noteworthy that the concentration of Cr(VI) in Suzhou was 0.66 ng m<sup>-3</sup>, which was lower than some cities of Northern China such as Beijing (2.5 ng m<sup>-3</sup>) (Fan et al., 2021), Baoding (8.3 ng m<sup>-3</sup>) (Liu et al., 2018), and some cities of Eastern China such as Shanghai (3.2 ng m<sup>-3</sup>) (Chen et al., 2008), and some cities of Southern China such as Xiamen (3.7 ng/m<sup>3</sup>) (Wang et al., 2017), and some other Asian cities such as Siheung (2.43 ng m<sup>-3</sup>) (Lee et al., 2022), but more than twenty times higher than the China standards of 0.025 ng m<sup>-3</sup>. Many studies showed that the sources of Cr included brake and engine wear (Charron et al., 2018), industrial metalworking activities (Dai et al., 2015), and the incineration of plastic materials.

**Table 3** Annual mean concentrations (ng m<sup>-3</sup>) of PM<sub>2.5</sub>-bound heavy metals in Suzhou during 2019 and 2021

Types	Min–Max	Concentration (Mean ± SD)	Heavy metals concentration/PM <sub>2.5</sub> (%)
PM <sub>2.5</sub> <sup>a</sup>	4.00–209.00	46.76 ± 29.84	–
Sb	ND–77.20	4.19 ± 3.97	0.010
Al	3.00–2045.00	109.77 ± 156.00	0.288
As	0.60–38.90	3.51 ± 3.57	0.009
Cd	0.03–7.77	0.71 ± 0.81	0.002
Cr	0.51–55.70	4.63 ± 4.78	0.013
Hg	ND <sup>b</sup> –5.81	0.36 ± 0.50	0.001
Pb	0.87–153.00	21.92 ± 18.94	0.057
Mn	2.32–197.00	30.48 ± 24.73	0.075
Ni	0.32–21.40	3.22 ± 2.48	0.009
Se	0.41–34.70	2.61 ± 3.01	0.007
Tl	ND–4.70	0.23 ± 0.49	0.001
$\Sigma$ Heavy metals	17.04–2204.99	180.61 ± 176.95	0.472

<sup>a</sup>The unit of PM<sub>2.5</sub> mass concentration is µg m<sup>-3</sup>

<sup>b</sup>The value of heavy metal is below the method detection limit

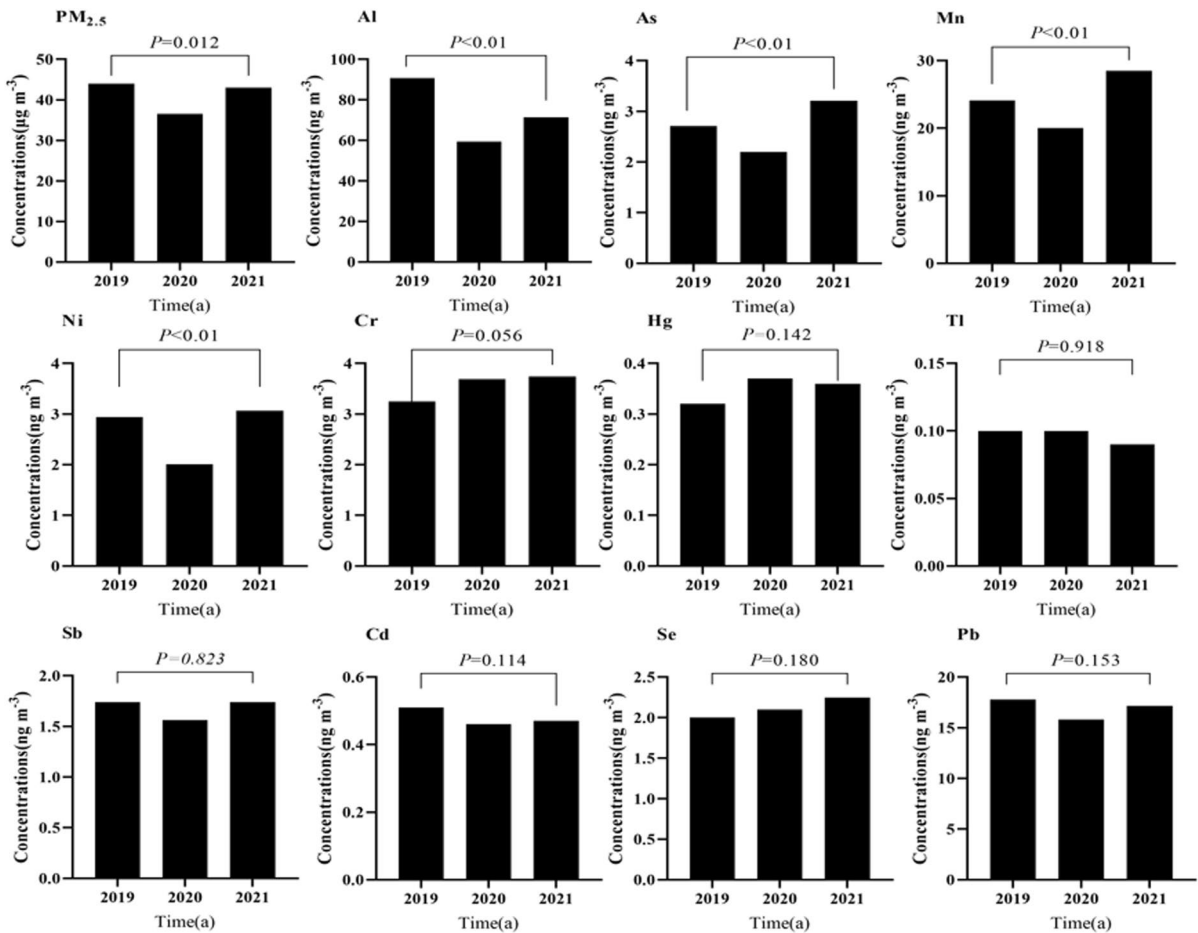


**Table 4** Spearman's correlation coefficients of the concentration of trace elements in PM<sub>2.5</sub> with meteorological parameters in Suzhou during 2019–2021

	PM <sub>2.5</sub>	Sb	Al	As	Cd	Cr	Hg	Pb	Mn	Ni	Se	Tl
<i>T</i> <sup>a</sup>	-0.279**	-0.198**	-0.272**	-0.155**	-0.229**	-0.109*	-0.263**	-0.341**	-0.314**	-0.137**	-0.165**	-0.378**
<i>RH</i> <sup>b</sup>	-0.164**	-0.134**	-0.266**	-0.032	-0.260**	-0.094*	0.001	-0.194**	-0.226**	0.042	-0.116*	-0.209**
<i>AP</i> <sup>c</sup>	0.088*	0.160**	0.181**	0.062	0.148**	0.040	0.145**	0.270**	0.250**	0.058	0.119**	0.296**
<i>WS</i> <sup>d</sup>	-0.309**	-0.170**	0.020	-0.067	-0.140**	-0.125**	-0.115**	-0.154**	-0.206**	-0.141**	-0.238**	-0.167**
<i>P</i> <sup>e</sup>	-0.220**	-0.164**	-0.218**	-0.106*	-0.238**	-0.100*	0.009	-0.207**	-0.233**	-0.108*	-0.206**	-0.206**

<sup>a</sup>*T* is temperature, <sup>b</sup>*RH* is relative humidity, <sup>c</sup>*AP* is atmospheric pressure, <sup>d</sup>*WS* is wind speed, <sup>e</sup>*P* is precipitation; \* Showed significant correlation at 0.05 level (double side); \*\* Showed significant correlation at 0.01 level (double side). Significance values at *p* < 0.05 are marked bold

Annual variations of the heavy metal concentrations in PM<sub>2.5</sub> from 2019 to 2021 are presented in Fig. 2. The concentrations of PM<sub>2.5</sub>, Al, As, Mn, Ni were significantly lower in 2020 than that in 2019 and 2021. The main reason might be ascribed to the 2019 novel coronavirus (COVID-19) epidemic which broke out intermittently in 2020. Due to the seriousness of the COVID-19 epidemic, lots of industrial activities and pollutant emissions were not exactly at full capacity, leading to the improvement of the air quality globally (Chen et al., 2021; Liu et al., 2020). For example, Li et al.(2020) reported that human activities such as industrial operations were lowered significantly during the COVID-19 lockdown over the Yangtze River Delta Region of China, leading to lowered PM<sub>2.5</sub> emissions by 27–46%. Mahato et al.(2020) reported that the concentration of PM<sub>2.5</sub> during the COVID-19 lockdown in Delhi decreased by 50% compared to pre-lockdown levels. The current studies have shown that the sources of heavy metals in PM<sub>2.5</sub> are different and characteristic, and the proportions of these heavy metals in PM<sub>2.5</sub> are very small (Table 3). The variation trends of some heavy metals in PM<sub>2.5</sub> maybe inconsistent with the variation trends of PM<sub>2.5</sub> and the other heavy metals because of the variations of characteristic pollution sources. Previous studies showed that the sources of Al mainly included fugitive dust and related industrial activities, and As was main from coal combustion (Tian et al., 2010), and Mn and Ni mainly came from vehicle emissions and metallurgical industries (Chang et al., 2009). The decrease in Al, As, Mn, Ni concentrations in 2020 were primarily attributable to the strict anti-epidemic measures which resulted in reduction in coal combustion, industrial operations and constructions (Li et al., 2020). It was noteworthy that many studies showed that Cd, Pb and As were the key tracers of coal combustion (Ambade 2014; Pan et al., 2015). However, the concentrations of Cd and Pb in PM<sub>2.5</sub> in Suzhou during 2019 and 2021 didn't decrease when the coal combustion was reduced. One possible reason was that the contribution from coal combustion emissions to atmospheric Cd and Pb was not considered unique. In addition to coal combustion, sources of Cd and Pb included brake and engine wear and motor oil combustion (Charron et al., 2018). In order to control the speed of the COVID-19, the local authorities advised citizens to drive to work, resulting in elevation in

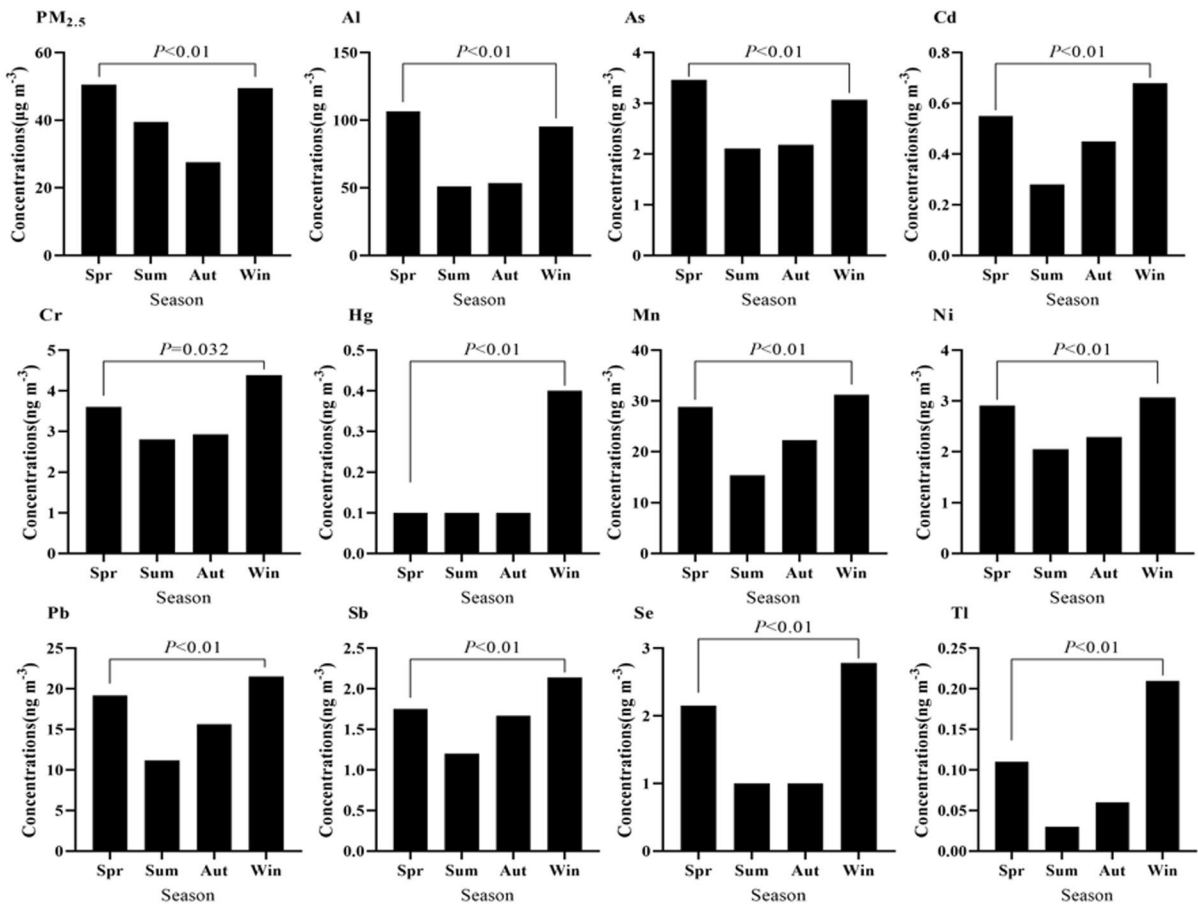


**Fig. 2** Annual variations of heavy metal concentrations in PM<sub>2.5</sub> in Suzhou from 2019 to 2021

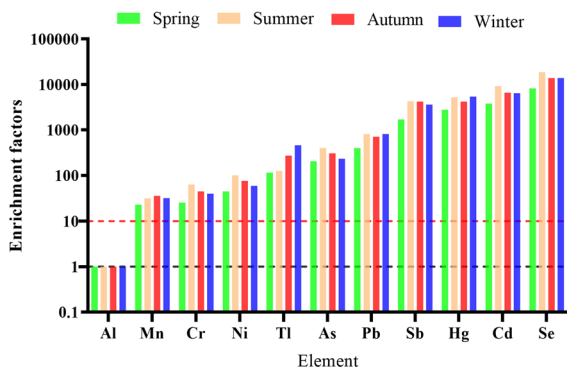
traffic pollution emissions including Cd and Pb emissions.

As can be seen in Fig. 3, the concentrations of PM<sub>2.5</sub>-bound heavy metals in Suzhou displayed obvious seasonality. Overall, the concentrations of most heavy metals were consistent with the seasonal distribution of PM<sub>2.5</sub>, showing a pattern of high concentrations in winter and spring, and low concentrations in summer and autumn. This pattern was consistent with previous observations in Nanjing (Sun et al., 2014) and Hangzhou (Xu et al., 2021). The seasonal patterns of the heavy metals were attributable to source emissions, meteorological conditions, and environmental transport. Because of the lower temperature, lower planetary boundary layer and prevalent stagnant atmosphere

in winter, a larger amount of particulate matters generated by local fossil fuel combustion and vehicular emissions was released into the atmosphere. Meanwhile, the prevailing northwest wind in winter promoted the long-distance transmission of plentiful particulate matters which generated by industrial activities and house-heating in Northern China. The lower concentrations in summer and autumn could be explained by less fossil fuel combustion, better dispersion due to higher planetary boundary layer, and effective wet scavenging by more precipitations (Xu et al., 2019). It was noteworthy that the concentration of Hg in PM<sub>2.5</sub> showed a pattern of high concentrations in winter, and low concentrations in spring, summer and autumn, which was not exactly consistent with the seasonal distribution of the other ten PM<sub>2.5</sub>-bound heavy metals. One possible reason



**Fig.3** Seasonal variations of heavy metal concentrations in PM<sub>2.5</sub> in Suzhou from 2019 to 2021



**Fig.4** The seasonal EF values of PM<sub>2.5</sub>-bound heavy metals in Suzhou during 2019 and 2021

was that the contribution from coal combustion emissions to atmospheric Hg was not considered unique. The atmospheric Hg is mainly from coal combustion. In addition to the coal combustion, source of Hg also includes waste incineration and metallurgical industries. Because of the high temperature in spring in Suzhou, the coal combustion might be reduced in spring, resulting in lower concentration of Hg in PM<sub>2.5</sub>.

#### Health risks posed by PM<sub>2.5</sub>-bound heavy metals

As shown in Fig. 4, the EFs of Mn, Cr, Ni, Tl, As, Pb, Sb, Hg, Cd, and Se in PM<sub>2.5</sub> in Suzhou were all greater than 10 in all seasons, indicating that these heavy metals were mainly influenced by anthropogenic sources, such as fossil fuel combustion and vehicular emissions (Chen et al., 2008; Jena and

**Table 5** Non-carcinogenic and carcinogenic risks of PM<sub>2.5</sub>-bound trace elements via inhalation exposure in Suzhou

Elements	95%UCL of EC (ng m <sup>-3</sup> ) <sup>a</sup>					CR								
	Spring	Summer	Autumn	Winter		Spring	Summer	Autumn	Winter					
Sb	3.95	4.19	4.32	6.15		1.32×10 <sup>-2</sup>	1.40×10 <sup>-2</sup>	1.44×10 <sup>-2</sup>	2.05×10 <sup>-2</sup>		- <sup>b</sup>	-	-	-
Al	220.63	75.11	96.15	134.89		4.41×10 <sup>-2</sup>	1.50×10 <sup>-2</sup>	1.92×10 <sup>-2</sup>	2.70×10 <sup>-2</sup>		-	-	-	-
As	5.03	3.91	3.49	4.04		0.34	0.26	0.23	0.27		<b>8.11×10<sup>-6</sup></b>	<b>6.30×10<sup>-6</sup></b>	<b>5.63×10<sup>-6</sup></b>	<b>6.51×10<sup>-6</sup></b>
Cd	0.85	0.83	0.69	1.02		0.09	0.08	0.07	0.10		5.74×10 <sup>-7</sup>	5.60×10 <sup>-7</sup>	4.66×10 <sup>-7</sup>	6.89×10 <sup>-7</sup>
Cr(VI)	0.79	0.80	0.66	0.86		7.90×10 <sup>-3</sup>	8.00×10 <sup>-3</sup>	6.60×10 <sup>-3</sup>	8.60×10 <sup>-3</sup>		<b>3.56×10<sup>-6</sup></b>	<b>3.60×10<sup>-6</sup></b>	<b>2.97×10<sup>-6</sup></b>	<b>3.87×10<sup>-6</sup></b>
Hg	0.45	0.31	0.37	0.64		1.50×10 <sup>-3</sup>	1.03×10 <sup>-3</sup>	1.23×10 <sup>-3</sup>	2.13×10 <sup>-3</sup>		-	-	-	-
Pb	24.08	19.86	20.63	34.39		0.16	0.13	0.14	0.23		7.22×10 <sup>-7</sup>	5.96×10 <sup>-7</sup>	6.19×10 <sup>-7</sup>	1.03×10 <sup>-7</sup>
Mn	41.66	21.43	29.85	44.60		0.83	0.43	0.60	0.89		-	-	-	-
Ni	3.93	3.50	3.30	3.84		0.08	0.07	0.07	0.08		3.54×10 <sup>-7</sup>	3.15×10 <sup>-7</sup>	2.97×10 <sup>-7</sup>	3.46×10 <sup>-7</sup>
Se	3.12	2.79	2.64	3.87		1.56×10 <sup>-4</sup>	1.40×10 <sup>-4</sup>	1.32×10 <sup>-4</sup>	1.94×10 <sup>-4</sup>		-	-	-	-
Tl	0.21	0.09	0.25	0.63		-	-	-	-		-	-	-	-
HI						<b>1.56<sup>c</sup></b>	<b>1.01</b>	<b>1.14</b>	<b>1.63</b>		<b>1.33×10<sup>-5</sup></b>	<b>1.14×10<sup>-5</sup></b>	<b>9.98×10<sup>-6</sup></b>	<b>1.25×10<sup>-5</sup></b>
∑CR														

<sup>a</sup>The 95% upper confidence limit (UCL) of the arithmetic average concentrations of the daily mean element concentration in Suzhou from 2019 to 2021

<sup>b</sup>There are no specific studies showing that the element has cancer risk or non-cancer risk

<sup>c</sup>HQ value of the non-carcinogen above 1 or CR value of the carcinogen above 10<sup>-6</sup> is marked bold

Singh 2017). Hence, it is necessary to assess the non-carcinogenic and carcinogenic risks posed by these heavy metals via inhalation exposure for the general population in Suzhou.

Regarding non-carcinogenic risks (Table 5), the HQs of Mn, Cr(VI), Ni, As, Pb, Sb, Hg, Cd and Se were all less than 1 in Suzhou, indicating long-term exposure to these heavy metals were unlikely to cause adverse health effects. The non-carcinogenic risks to the residents in Suzhou were lower than the other cities in YRD of China, such as Hangzhou and Ningbo ( $HQ_{As}=8.7$ ) (Xu et al., 2021). However, the HI values which represent the sum of HQ values via inhalation exposure for inhabitants were all greater than 1 in all seasons, indicating that the integrated effects of multi-elemental exposure, especially Mn which observed with the highest HI value, might result in adverse non-carcinogenic risks to local residents.

For carcinogenic risks (Table 5), the CR values of Cd, Pb, and Ni were less than  $1 \times 10^{-6}$  in all seasons, indicating the carcinogenic risks posed by these trace elements to local residents via inhalation were negligible. However, the CR values of Cr(VI) and As in all seasons were greater than  $1 \times 10^{-6}$  and less than  $1 \times 10^{-4}$ . Hence, Cr(VI) and As could be considered two key carcinogenic risk factors which was consistent with previous observations in Zhejiang Province (Xu et al., 2021).

In this study, probability method was also used to assess the non-carcinogenic and carcinogenic risks posed by these heavy metals. Compared with point estimation, probability method can directly reflect the temporal distribution of the health risks. The screening concentration of PM<sub>2.5</sub>-bound heavy metals was calculated by Eq. 2, 3 and 4. The screening concentration refers to the concentration of non-carcinogenic or carcinogenic pollutant when its HQ is 1 or CR is  $1 \times 10^{-6}$ , respectively (NHCPRC, 2019). An exceedance was defined when the concentration of the trace element exceeded the screening concentration. As shown in Table 6, the HQ values of Sb, Al, Cd, Cr(VI), Hg, Ni, and Se were all less than 1 in all seasons. It was worth noting that the percentage exceedances of the HQ value of Mn in winter and spring were higher which were 15.87% and 33.33%, respectively. This implies the non-carcinogenic risks to Suzhou residents posed by Mn in PM<sub>2.5</sub> in winter and spring should be taken seriously. For carcinogenic risks, the CR values of Cr(VI) and As in

all seasons were greater than  $1 \times 10^{-6}$  and less than  $1 \times 10^{-4}$ , with percentage exceedances of 79.96% and 78.57%, respectively, indicating the carcinogenic risks to Suzhou residents posed by Cr(VI) and As in PM<sub>2.5</sub> in all seasons should be taken seriously.

The health risk assessment indicated that PM<sub>2.5</sub>-bound carcinogenic elements posed potential carcinogenic risks to the general population in Suzhou. In particular, Cr(VI) and As were the key carcinogenic risk factors. At the same time, the potential non-carcinogenic risks should be taken seriously because of the integrated effects of multi-elemental exposure. Hence, it is necessary for the local government to take pertinent actions to prevent and control emissions of PM<sub>2.5</sub>-bound toxic elements. As the key carcinogenic risk factors, sources of Cr(VI) and As should be targeted by effective controls on activities, such as traffic emissions, industrial Cr plating operations (Karar et al., 2006), and coal combustion emissions.

## Conclusions

To our knowledge, this is the first study that analyzed pollution characteristics and health risk of PM<sub>2.5</sub>-bound heavy metals in Suzhou. The annual mean concentration of PM<sub>2.5</sub> in Suzhou during 2019 and 2021 was  $46.76 \pm 29.84 \mu\text{g m}^{-3}$ , with percentage exceedances of 15.28% and 91.07% in terms of the China national ambient air quality standard limit of  $75 \mu\text{g m}^{-3}$  and the WHO recommended limit of  $15 \mu\text{g m}^{-3}$ , respectively. The sum of concentrations of 11 heavy metals in Suzhou averaged  $180.61 \pm 176.95 \text{ ng m}^{-3}$ , dominated by Al, Mn, and Pb. The concentrations of PM<sub>2.5</sub> and PM<sub>2.5</sub>-bound heavy metals displayed significant seasonality, showing a pattern of high concentrations in winter and spring and low concentrations in summer and autumn. The EF approach revealed that all the heavy metals were mostly influenced by anthropogenic sources, except for Al, which was mainly from crustal source. All the single non-carcinogenic heavy metals were unlikely to cause non-carcinogenic effects. However, the HI value of these elements was slightly greater than 1, indicating that the integrated non-carcinogenic risks to local residents via inhalation should be taken seriously. The highest and lowest cumulative carcinogenic risk from As, Cd, Cr(VI),

**Table 6** Percentage exceedance of the non-carcinogenic and carcinogenic risks of PM<sub>2.5</sub>-bound trace elements via inhalation exposure in Suzhou

Elements	CR = 1 × 10 <sup>-6</sup>												
	HQ = 1					CR = 1 × 10 <sup>-6</sup>							
	Screening Concentration (ng/m <sup>3</sup> ) <sup>a</sup>	Percentage exceedance (%) <sup>c</sup>				Total	Screening Concentration (ng/m <sup>3</sup> ) <sup>b</sup>	Percentage exceedance (%)				Total	
Spring		Summer	Autumn	Winter	Spring			Summer	Autumn	Winter			
Sb	300	0.00	0.00	0.00	0.00	0.00	— <sup>d</sup>	—	—	—	—	—	—
Al	5000	0.00	0.00	0.00	0.00	0.00	—	—	—	—	—	—	—
As	15	0.79	1.59	0.79	0.00	0.79	0.62	83.33	76.19	75.40	79.37	78.57	78.57
Cd	10	0.00	0.00	0.00	0.00	0.00	1.48	7.94	12.70	6.35	15.87	10.71	10.71
Cr(VI)	100	0.00	0.00	0.00	0.00	0.00	0.22	85.71	73.02	80.95	79.37	79.96	79.96
Hg	300	0.00	0.00	0.00	0.00	0.00	—	—	—	—	—	—	—
Pb	150	0.00	0.00	0.00	0.79	0.20	33.33	11.90	11.90	8.73	31.75	16.07	16.07
Mn	50	15.87	2.38	7.94	33.33	14.88	—	—	—	—	—	—	—
Ni	50	0.00	0.00	0.00	0.00	0.00	11.11	1.59	2.38	1.59	0.79	1.59	1.59
Se	20,000	0.00	0.00	0.00	0.00	0.00	—	—	—	—	—	—	—
Tl	—	—	—	—	—	—	—	—	—	—	—	—	—

<sup>a</sup>The concentration of the trace element when HQ is 1<sup>b</sup>The concentration of the trace element when CR is 1 × 10<sup>-6</sup><sup>c</sup>An exceedance is defined when the concentration of the trace element exceeds the concentration at HQ = 1 or CR = 1 × 10<sup>-6</sup><sup>d</sup>There are no specific studies showing that the element has cancer risk or non-cancer risk

Pb, and Ni in PM<sub>2.5</sub> in Suzhou were  $3.57 \times 10^{-5}$  and  $2.78 \times 10^{-5}$ , respectively, more than ten times of the lower limit ( $1 \times 10^{-6}$ ) of the acceptable risk range. Two key carcinogenic risk factors (Cr(VI) and As) in this study contributed 74.73% and 20.22% of the cumulative carcinogenic risk, respectively, and their percentage exceedances were as more as 79.96% and 78.57%, respectively. Therefore, government policies and countermeasures for the PM<sub>2.5</sub> pollution control should be performed not only based on the PM<sub>2.5</sub> concentration but also based on the PM<sub>2.5</sub>-bound heavy metals and their health risks for the local residents. For example, it is desperately necessary for Suzhou to reduce the concentrations of Cr and As and to manage their characteristic emission sources.

**Author contributions** All authors contributed to the study conception and design. PM<sub>2.5</sub> samples collection and data analysis were performed by XZ, MX, MZ, YW, JL and SL. The detection of samples were performed by JL. The technological guidances were provided by QL. The first draft of the manuscript was written by XZ and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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**Data availability** The datasets used and/or analyzed during the current study are available on reasonable request.

#### Declarations

**Competing interests** The authors declare no competing interests.

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