

## Research Article

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# Concentrations of heavy metals in PM<sub>2.5</sub> and health risk assessment around Chinese New Year in Dalian, China

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**Abstract:** Twelve samples of heavy metals were analyzed by using a 1108A-1 mid-current particle sampler in Dalian, Liaoning Province, for 31 days before and after the spring festival 2019. The results showed that the concentrations of heavy metals were decreased by more than 25% during the spring festival, which was probably due to the shutdown of the factories and the decrease in people's travel. During the spring festival, the concentration of Ba was increased by 343.39% as compared to the concentration of Ba before the spring festival, which indicated that the fireworks had a great influence on the concentration of Ba. At the same time, this study also evaluated the health risk of heavy metals. For the heavy metals As, Cd, Co, Cr, and Ni, the lifetime cancer risk was found to be  $2.13 \times 10^{-4}$ ,  $2.08 \times 10^{-5}$ ,  $8.64 \times 10^{-7}$ ,  $4.39 \times 10^{-4}$  and  $7.93 \times 10^{-7}$ , respectively. The lifetime cancer risk of As, Cd, and Cr exceeds the threshold range of cancer risk ( $10^{-6}$ – $10^{-4}$ ), indicating that they are carcinogenic to humans. Also, during the spring festival, the non-carcinogenic risk value of V exceeded the limit value of environmental protection agency (EPA), and the lifetime carcinogenic risk value of As, Cd, and Cr exceeded the threshold range of carcinogenic risk; hence, they need to be carefully monitored and controlled.

**Keywords:** PM<sub>2.5</sub>, heavy metal concentration, spring festival, health risk assessment, cancer risk, non-cancer risk

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## 1 Introduction

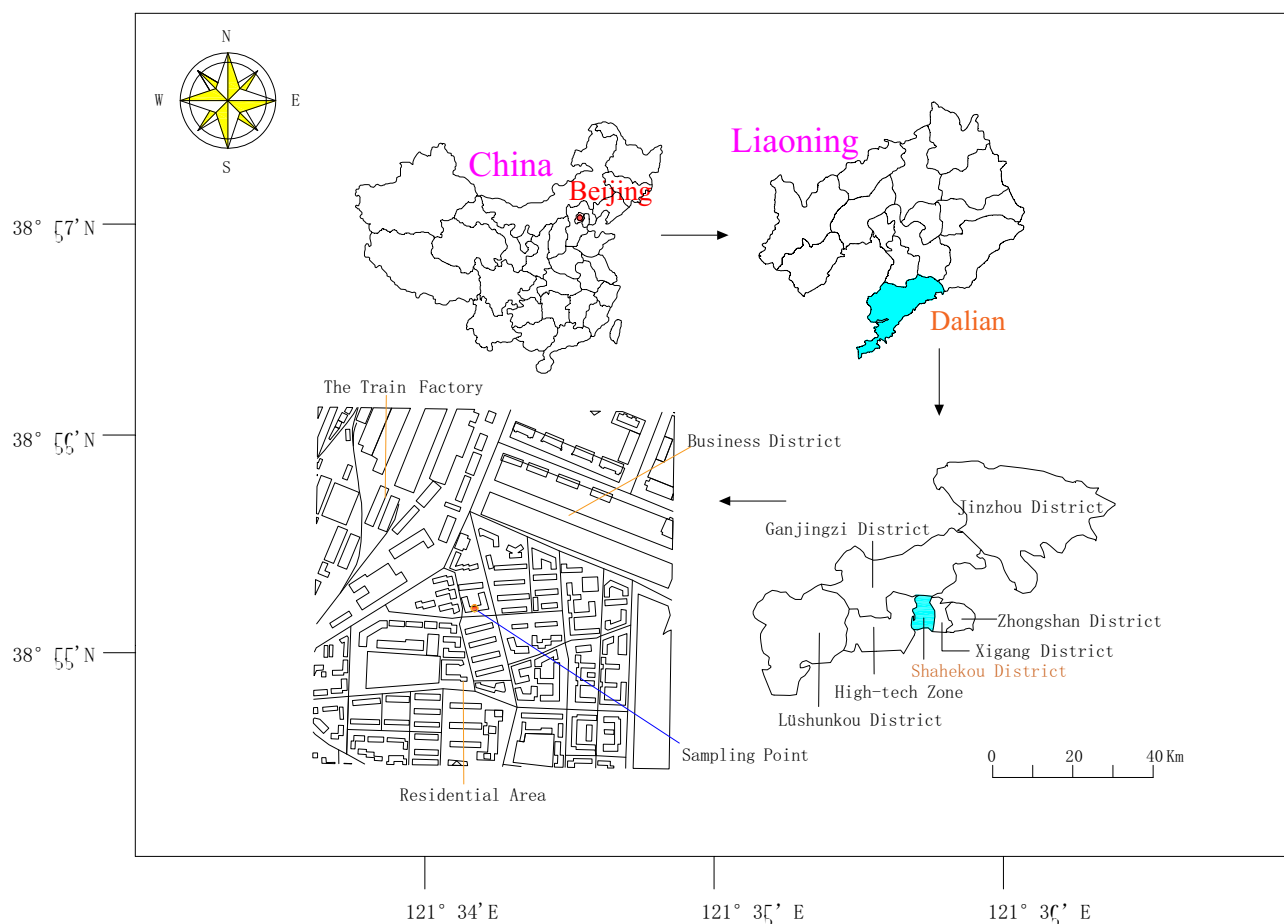
During the past few years in China, due to the rapid increase in urbanization, industrialization, and economic growth, the existence of smog or haze episodes as reflected by the high fine particulate matter levels and reduced visibility has been spotlighted in national-scale China, particularly in the most advanced and highly populated cities [1]. Particulate matter (PM<sub>2.5</sub>) having a diameter less than or equal to 2.5  $\mu\text{m}$  has been considered as major particulate air pollution. PM<sub>2.5</sub> has been recognized as a potential carrier (because of its higher surface area to mass ratio) of biologically accessible transition metal or metalloids that can easily enter into the human body via ingestion, dermal contact absorption, and inhalation [2–4]. Atmospheric fine particulate matter has become the primary pollutant in most Chinese cities [5]. Generally, PM<sub>2.5</sub> originates mainly from the sources such as dust blowing, organic matter burning, automobile exhaust, coal burning, metal smelting, regional transported aerosols, and other industrial processes [6–11]. However, still, it was challenging to identify the contributions from each source and to understand the mechanism for the formation of particulate matter [12,13]. Inorganic elements are an important part of atmospheric particulate matter, and most of them have the characteristics of refractory degradation and easy enrichment, which can cause functional obstacles to human organs and cause irreversible damage [14,15]. For example, exposure to high levels of arsenic for long periods of time can cause serious damage to the body and can even lead to lung and skin cancer [16]. Heavy metals in PM<sub>2.5</sub> are mainly derived from road dust, traffic emissions, fuel combustion, and industrial processes [17,18].

Dalian is the largest port city in Northeast China, with an annual cargo throughput of 200 million tons. The Spring Festival is one of the most important festivals in China. In the traditional concept, in order to celebrate the Spring Festival, people set off a large number of fireworks

and firecrackers during Spring Festival. Spring Festival generally refers to a total of 16 days from the 30th day of the lunar new year to the 15th day of the first month of the lunar new year. In recent years, the state has increased restrictions on the discharge of fireworks during the Spring Festival and advocated reducing the discharge of fireworks. However, there are still a lot of fireworks during the Spring Festival. Fireworks and firecrackers have a great impact on the concentration of  $PM_{2.5}$ , and because of the harsh weather conditions in winter, air particulate matter pollution easily occurs. This causes some particularity in the  $PM_{2.5}$  concentration and heavy metal content during the spring festival [19]. At the same time, heavy metals in  $PM_{2.5}$  in the atmosphere can cause serious damage to people when they enter the body, and so, the primary task of air quality management is to study their sources and determine their sources [20]. According to a 2010 study, more than 200,000 people worldwide die

each year from diseases caused by air pollution [21]. Meanwhile, the International Agency for research on cancer, the World Health Organization's specialized cancer agency, officially listed outdoor air pollution as the first group of human carcinogens in October 2013 [22]. Therefore, the research on air pollution is very important. In recent years, most of the studies are primarily concentrated on the analysis of health risks for the preferred toxic elements that are released from specific sources [23,24]. But, there were some reports that have detected the main sources of the risk using the combination of the health risk assessment model and positive matrix factorization, and such research studies are still limited [25,26]. More importantly, there are a few studies on atmospheric  $PM_{2.5}$  and the concentration of heavy metals in Dalian

Therefore, based on this, a 31 day study was conducted in Dalian, Liaoning Province, China, during the 2019 spring festival. The concentrations of heavy metals



**Figure 1:** Sampling point of atmospheric deposit in Dalian.

in PM<sub>2.5</sub> before and after the spring festival in Dalian were determined and a health risk assessment was conducted.

## 2 Materials and methods

### 2.1 Sample collection

The longitude of Dalian is in between 120°58' and 123°31'E and latitude is in between 38°43' and 40°10'N. The topography of Dalian was characterized by the central height, and the east and west sides descend in steps to the seashore, forming the landform of the mountain and Hilly Peninsula. According to the national environmental protection standards of the People's Republic of China (HJ618-2011), the sample was taken from the top of the 7th floor of a residential building in the Shahekou district of Dalian, about 24 m above ground level, with no buildings higher than 200 m in the vicinity. Dalian Locomotive Works was 500 m to the northwest of the sampling site, and the commercial center was about 200 m to the northeast side. The south and southwest are the residential areas. In general, the study area around the increased human activities and traffic can be considered as a region of high activity (Figure 1).

The sampling time was from January 22 to February 22, 2019, of which February 5, 2019, was New Year's Day, and February 19, 2019, was the lantern festival. The medium flow intelligent particle sampler of type Lao1108a-1 was adopted. A polypropylene filter of 90 mm was selected, and the sampler was calibrated by flow calibrator before use, and the flow rate was set to 100 L/min. The sampling time is from 9 a.m. to 8:30 a.m. the next day, and each sampling time is 23.5 h. The polypropylene filter membrane was balanced in the environment with temperature (20 ± 1)°C and humidity (50% ± 1%) for 48 h before and after sampling.

### 2.2 Chemical analysis

First, ¼ polypropylene filter membrane was cut with the help of ceramic scissors and placed in the digester along with 5.6 mL of nitric acid (pH = 5.6), 0.05 mL of 40% hydrofluoric acid (pH = 5.3), and 5 mL of dilute nitric acid (pH = 5.4). A 0.45 µm needle filter was used one or two times. The volume was set to 50 mL with 1% nitric acid. Inductively coupled plasma massspectrometry (ICP-MS) was used to analyze the 12 elements such as V, Cr, Mn, Ni, Cu, Zn,

Pb, As, Ba, Sb, Co, and Cd. The analyte was introduced into an argon stream as an aerosol in an aqueous solution, and then it entered the central region of an argon plasma excited by radio frequency energy at atmospheric pressure. The sample was ionized, dissociated, vaporized, and dissolvable because of the higher temperature of the plasma. The plasma enters the vacuum system through different pressure regions. In the vacuum system, the MS section (quadrupole rapid scanning mass spectrometer) detects all the ions by high-speed sequential scanning separation and by high-speed double-channel separation.

The membrane was changed before and after each sampling to ensure that the filter membrane is flat, free of burrs and damage. The sampling head needs to be cleaned once in 168 h. For every 10 samples measured, a blank filter membrane was set and a single-point calibration was conducted to ensure that the blank control samples and quality control samples in each batch of experiments were measured synchronously. Each batch (≤20) was tested for spiked recovery. The recovery, the average relative standard deviation (RSD), and the standard curve  $R^2$  of 12 ions are 85.5–115.5%, <10%, and 0.999, respectively.

### 2.3 Health risk assessment of heavy metals

The human health risk assessment model was developed by the United States Environmental Protection Agency in 1983, which divides risk assessment into four steps: (i) hazard identification, (ii) dose response, (iii) exposure assessment, and (iv) risk characterization [27]. Among the 12 elements used in the study, cobalt, arsenic, nickel, cadmium, and chromium were carcinogens and their risks were assessed using the lifetime average daily exposure (LDD), while the remaining non-carcinogens were represented by the daily average exposure metric (ADD) [28]. The formulae used for the calculation of ADD and LADD are as follows:

$$\text{ADD} = \frac{C \times \text{IR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{ATF}} \quad (1)$$

$$\text{LADD} = \frac{C \times \text{EF}}{\text{AT}} \times \left( \frac{\text{InhR}_{\text{child}} \times \text{ED}_{\text{child}}}{\text{BW}_{\text{child}}} + \frac{\text{InhR}_{\text{adult}} \times \text{ED}_{\text{adult}}}{\text{BW}_{\text{adult}}} \right) \quad (2)$$

$$\text{ILCR} = \text{LADD} \times \text{SF} \quad (3)$$

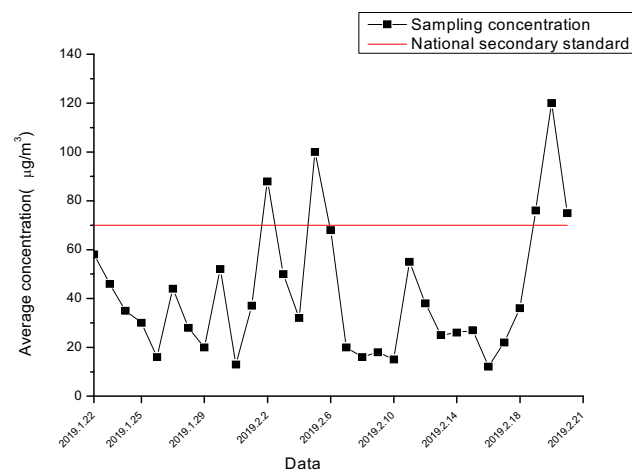
$$\text{HQ} = \frac{\text{ADD}}{\text{RfD}}. \quad (4)$$

In the above equations,  $C$  is the concentration of pollutants in  $\text{mg}/\text{m}^3$ ;  $\text{InhR}$  refers to the respiratory rate in  $\text{m}^3/\text{day}$ ;  $\text{EF}$  refers to the exposure frequency in  $\text{d}/\text{a}$ ;  $\text{ED}$  refers to the exposure duration in a BW;  $\text{AT}$  refers to the average exposure time in day; and  $\text{HQ}$  refers to the risk coefficient: when  $\text{HQ} > 1$ , it indicates the non-cancer risk. reference dose (RFD) is the reference dose, which represents the maximum amount of pollutants in  $\text{mg}/(\text{kg day})$ ; this dose of intake of heavy metals per unit weight of the human body will not cause adverse reactions. ILCR represents the lifetime cancer risk, indicating the probability of causing cancer;  $\text{SF}$  refers to the slope coefficient, in  $(\text{mg}/[\text{kg day}])$ , of respiratory exposure, indicating the maximum probability of a person being exposed to a given dose of a contaminant to produce a carcinogenic effect [29].

### 3 Results and discussion

#### 3.1 PM<sub>2.5</sub> concentration

The subsampling period was divided into three parts: before the spring festival (January 22–February 4), during the spring festival (February 5–February 11), and after the spring festival (February 12–February 21). As can be seen from Figure 2, during the sampling period, the concentration of PM<sub>2.5</sub> exceeded the secondary standard ( $75 \mu\text{g}/\text{m}^3$ ) of the average daily concentration of PM<sub>2.5</sub> for 4 days, accounting for 13% of the total sampling days. During the whole sampling period, the average concentration of PM<sub>2.5</sub> was  $41.87 \mu\text{g}/\text{m}^3$ , which did not exceed the second



**Figure 2:** Characteristic chart of the PM<sub>2.5</sub> concentration change during sampling.

level of the daily average concentration of PM<sub>2.5</sub> in China. This concentration differed from the observations acquired in Nanjing and Beijing [30,31]. The average concentration of PM<sub>2.5</sub> was  $112.6 \mu\text{g}/\text{m}^3$  in the former and  $104.5 \mu\text{g}/\text{m}^3$  in the latter during the spring festival. Dalian was considered as a coastal city and could be one of the reasons for the above changes in the average concentration; also the wind power and air humidity during the sampling period were higher as compared to inland cities as mentioned above. The north wind was the direction of the dominant wind, which was favorable for the circulation of wind and the blowing of the pollutants to the sea; hence, the concentration of PM<sub>2.5</sub> was lower than those of inland cities.

During the sampling period, the PM<sub>2.5</sub> concentration had a total of six low values, including two before the festival, three on the festival, one after the festival, and the lowest value appeared after the festival. The main reason for the low PM<sub>2.5</sub> concentration before the festival may be related to the strong wind that appeared on that day. The reason for such a situation in the festival may be that it was during the spring festival holidays when factories were closed and people's travel was reduced; the low concentration of PM<sub>2.5</sub> during the spring festival holidays may be due to the shutdown of all factories, reduced road transport and the decreased travel of people. After the festival, maybe due to snow, which caused PM<sub>2.5</sub> deposition in the air, in addition to the strong wind at that time, so the concentration of PM<sub>2.5</sub> appeared low. Three peaks of the PM<sub>2.5</sub> concentration were observed during the sampling period, which was on 2, 5, and 20 February. On February 2, the concentration of PM<sub>2.5</sub> was  $88 \mu\text{g}/\text{m}^3$ , which was 137.8% higher than the previous day; however, on February 5 (New Year's Eve), the PM<sub>2.5</sub> concentration was  $100 \mu\text{g}/\text{m}^3$ , which was 212.5% higher than the previous day. The acquired concentration of PM<sub>2.5</sub> on February 20 was  $120 \mu\text{g}/\text{m}^3$ , which was 58% higher before the day. The peak of PM<sub>2.5</sub> concentration obtained on February 2 may be attributed to the combination of high humidity (72%) and increased ozone concentration ( $83 \mu\text{g}/\text{m}^3$ ). The last two peaks of the PM<sub>2.5</sub> concentration were related to the custom of people setting off fireworks and firecrackers. Besides the influence of people setting off fireworks, there was also a firework party on the day of the Lantern Festival; a large number of huge fireworks also had an impact on the PM<sub>2.5</sub> surge.

From February 5 to February 6 (the New Year's Eve and the first day of the Lunar New Year), the average concentration of PM<sub>2.5</sub> was  $84 \mu\text{g}/\text{m}^3$ , which was 12% higher than the national secondary standard for PM<sub>2.5</sub>. The average concentration of PM<sub>2.5</sub> obtained on February 7 to 10 was  $17 \mu\text{g}/\text{m}^3$ , which was 80% lower than the

previous two days. These data were consistent with the observations in Nanjing and Beijing, suggesting that  $PM_{2.5}$  concentrations would gradually increase and peak on New Year's Eve as, all factories were closed and less travel by the people during the Lunar New Year break, in turn, it shows a gradual downward trend. From February 11 to 14, with the end of the Chinese New Year holidays, again the concentration of  $PM_{2.5}$  began to increase slowly. There was a partial snowfall from February 15 to 16, which caused the wet deposition of  $PM_{2.5}$  in the air to be removed. On February 17, the level of  $PM_{2.5}$  increases slowly and finally returns to their pre-spring festival levels, probably because of the resumed production by the factories, increased industrial emissions, return of the people to their normal routine, and the increased level of vehicle exhaust emissions.

### 3.2 Concentration of heavy metals

During the Spring Festival in 2019, the order of heavy metals by concentration were as follows:  $Zn > Ba > V > Pb > As > Mn > Cr > Cu > Cd > Ni > Sb > Co$ . The limits of As and Cd are  $0.006$  and  $0.005 \mu g/m^3$ , respectively. The results showed that the average concentration of As was  $0.0774 \mu g/m^3$ , which was 12.9 times the national standard, and the average concentration of Cd was 3.62 times of the national standard. These findings indicate that the pollution of As and Cd was severe during the sampling

period. This was the same as that observed in Nanjing and Kunming. It is generally accepted that As is the identifying element of coal combustion, usually from fly ash [32–34], and Cd is usually thought to come from industrial smelting processes and waste incineration [35–37].

During the spring festival, the concentrations of many heavy metals decrease as compared to those before the spring festival; the changes in the concentration are as follows: Cd (25.16%), Cr (25.46%), Co (25.64%), Cu (33.45%), Ni (37.33%), As (37.44%), V (40.04%), Pb (43.32%), Zn (50.15%) and Mn (59.79%). V and Ni are commonly used as identifying elements for the burning of heavy oil from ships [34,35]. As an important port in northern China, in the Dalian's port industry, the emissions during the spring festival period are reduced because of the shutdown of the port, and hence, the concentrations of V and Ni decrease during the spring festival. The main sources of Cu, Pb, and Zn are the wear of brake pads associated with road dust and the marked elements of automobile exhaust. The concentrations of these three elements were decreased in different aspects [36–39], which may be due to a decrease in the number of cars driven by the people and less travel during the spring festival. The concentrations of Mn, Cr, and Co, which are the marking elements of metal smelting and industrial combustion [40], were much lower than those before the spring festival because of the shutdown of factories during the spring festival. As is a marked element of coal combustion and the main source is metal smelting [41,42]; in addition to

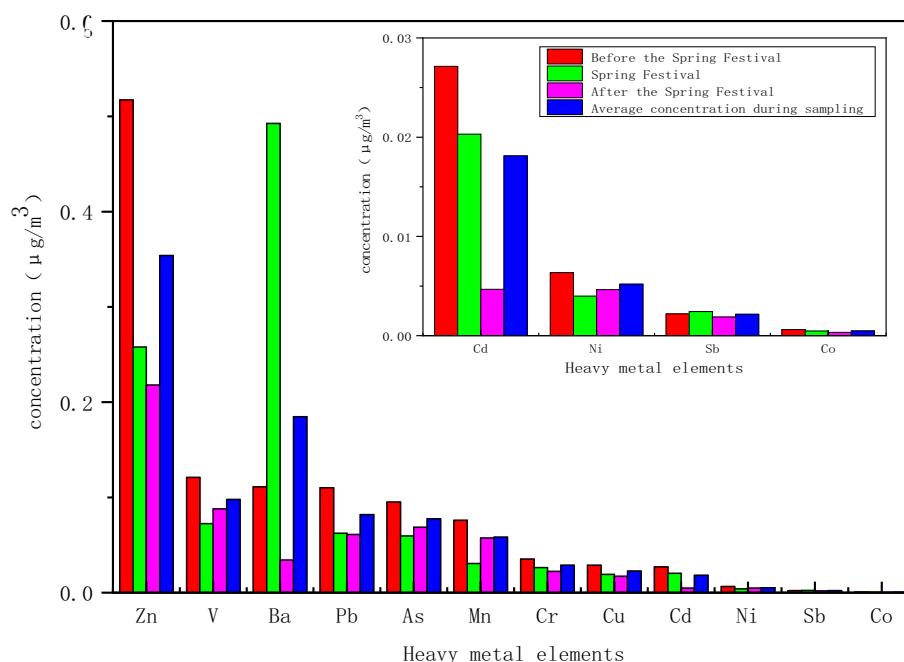


Figure 3: Average concentrations of heavy metals during sampling.



the use of coal for factory boiler combustion and heating, the Dalian area also has a considerable part of the small bath boiler for water heating. During the spring festival, factories and small baths do not produce, so the amount of coal burned will drop and the concentration of As during the spring festival will be lower than that before the festival (Figure 3).

In this study, the concentration of Ba increased by 343.39% during the spring festival as compared with the concentration before the spring festival but decreased by 93.07% after the spring festival. These findings were consistent with the results observed in India [19], Shenyang [40], Kunming [43], etc. All showed significant increases in the concentration of Ba during the fireworks display. It was also confirmed from the other study that, during the spring festival in Beijing, the concentration of Ba was also increased because of the fireworks and firecrackers [44]. This may be related to the raw materials of fireworks and firecrackers, as well as the mode of action. The main raw materials of fireworks and firecrackers are strong oxidants like potassium nitrate, sulfur, charcoal, flame colorant, and flash additives Sr, Ba, Al, Cu, and other metal powders. Metal powders are metallic substances or metallic salts that decompose at high temperatures to produce different lights. For example, aluminum/magnesium alloys when heated give off a brilliant white light, strontium nitrate and lithium burn with a red light, sodium nitrate gives off yellow light, and barium nitrate gives off green light. When fireworks and firecrackers are ignited, charcoal powder, sulfur powder, metal powder, and so on burn rapidly under the action of an oxidant, producing gas containing carbon, nitrogen, sulfur, metal oxide dust, and produce a lot of light and heat [45,46].

The concentrations of Ba and Cd were decreased by 93.07 and 87.64%, respectively, whereas the concentration

of Mn was increased by 87.64% after the spring festival. Ba mainly comes from the fireworks discharge, Cd is usually used as a marking element, and Mn is used as a marking element in metal smelting and industrial combustion; therefore, the concentration of Mn increased obviously during the spring festival.

### 3.3 Health risk assessment of heavy metals

The health risk assessment model was developed by the United States Environmental Protection Agency in 1983, which divides risk assessment into four steps: (i) hazard identification, (ii) dose response, (iii) exposure assessment, and (iv) risk characterization [27]. Of the 12 elements studied in this article, except for cobalt, arsenic, nickel, cadmium, and chromium that are carcinogens, none of them are carcinogenic. The risk assessment for cobalt, arsenic, nickel, cadmium, and chromium is, therefore, expressed by the lifetime average daily exposure level of LADD, and the risk for the remaining elements is expressed by the daily average exposure level of ADD [28].

Table 1 shows the risk of respiratory exposure to heavy metals in PM<sub>2.5</sub> during the 2019 Chinese New Year in Dalian. The results showed that the non-carcinogenic risk values of heavy metals were HQ from 2.90 to  $2.86 \times 10^{-5}$ , and the order of non-carcinogenic risk values of heavy metals was as follows: V (2.90), Mn ( $8.63 \times 10^{-1}$ ), Cr ( $2.05 \times 10^{-1}$ ), As ( $5.35 \times 10^{-2}$ ), Co ( $1.76 \times 10^{-2}$ ), Pb ( $4.85 \times 10^{-3}$ ), Sb ( $1.12 \times 10^{-3}$ ), Zn ( $2.44 \times 10^{-4}$ ), Ba ( $1.92 \times 10^{-4}$ ), Cu ( $1.17 \times 10^{-4}$ ), Ni ( $5.38 \times 10^{-5}$ ), and Cd ( $3.76 \times 10^{-5}$ ). The non-carcinogenic risk for V was 2.90, exceeding the EPA limit of 1 [29]. This was different from the results observed in Shenyang, Nanchang, Nanjing,

**Table 1:** Risk of respiratory exposure to heavy metals in PM<sub>2.5</sub> during spring festival 2019 in Dalian

Heavy metal	HQ adult male	HQ adult female	HQ children	ILCR
V	2.90	2.62	2.20	—
Mn	$8.63 \times 10^{-1}$	$7.80 \times 10^{-1}$	$6.57 \times 10^{-1}$	—
Cr	$2.05 \times 10^{-1}$	$1.86 \times 10^{-1}$	$1.56 \times 10^{-1}$	$4.39 \times 10^{-4}$
As	$5.35 \times 10^{-2}$	$4.84 \times 10^{-2}$	$4.07 \times 10^{-2}$	$2.13 \times 10^{-4}$
Co	$1.76 \times 10^{-2}$	$1.59 \times 10^{-2}$	$1.34 \times 10^{-2}$	$8.64 \times 10^{-7}$
Pb	$4.85 \times 10^{-3}$	$4.39 \times 10^{-3}$	$3.69 \times 10^{-3}$	—
Sb	$1.12 \times 10^{-3}$	$1.01 \times 10^{-3}$	$8.51 \times 10^{-4}$	—
Zn	$2.44 \times 10^{-4}$	$2.21 \times 10^{-4}$	$1.86 \times 10^{-4}$	—
Ba	$1.92 \times 10^{-4}$	$1.73 \times 10^{-4}$	$1.46 \times 10^{-4}$	—
Cu	$1.17 \times 10^{-4}$	$1.06 \times 10^{-4}$	$8.90 \times 10^{-5}$	—
Ni	$5.38 \times 10^{-5}$	$4.86 \times 10^{-5}$	$4.09 \times 10^{-5}$	$7.93 \times 10^{-7}$
Cd	$3.76 \times 10^{-5}$	$3.40 \times 10^{-5}$	$2.86 \times 10^{-5}$	$2.08 \times 10^{-5}$

and other inland cities [47–49]. The non-cancer risk of V in these inland cities does not exceed 1; on the contrary, the non-cancer risk of V was very low. The biggest difference was likely to be geographical. Dalian is a large coastal city in the north with a well-developed ship-building industry and a large port. The Port of Dalian is the largest open port in northeast China, handling 467 million tons of cargo in 2018. The fuel used by a large number of cargo ships is heavy oil. The marking element of heavy oil burning is V, so the corresponding V is higher than other heavy metals.

For the heavy metals As, Cd, Co, Cr, and Ni, the lifetime cancer risk values were  $2.13 \times 10^{-4}$ ,  $2.08 \times 10^{-5}$ ,  $8.64 \times 10^{-7}$ ,  $4.39 \times 10^{-4}$  and  $7.93 \times 10^{-7}$ , respectively. The lifetime cancer risk of As, Cd, and Cr exceeds the threshold range of cancer risk ( $10^{-6}$ – $10^{-4}$ ), indicating that they are carcinogenic to humans. As, Cd, and Cr all come from coal combustion and so the lifetime cancer risk values of As, Cd and Cr will exceed the threshold value of ILCR. However, these results are different from those in Guiyang and Guangzhou [50,51], which may be because the South does not need heating as much as the North, and the South does not have as many heavy industries as the North. Therefore, the elements like As, Cd, and Cr, which were found in heavy industry or coal-burning businesses, were not in high concentrations in the South, so they do not have a high lifetime cancer risk of ILCR.

In summary, of all the heavy metals examined in this study, only V had a higher non-carcinogenic risk, while As, Cd, and Cr had a carcinogenic risk to humans. Therefore, it is necessary to strengthen the control of these four heavy metals to reduce their risks to the human body.

## 4 Conclusion

During the spring festival, the concentrations of many heavy metals in  $PM_{2.5}$  throughout the time of sampling period were lower as compared to the concentrations of the heavy metals before the spring festival. The lower concentrations of Cd, Cr, Co, Cu, Ni, As, V, Pb, Zn, and Mn were found to be 25.16, 25.46, 25.64, 33.45, 37.33, 37.44, 40.04, 43.32, 50.15, and 59.79%, respectively. This may be due to the shut down of factories and the reduced travel by the people during the Lunar New Year holidays. The health risk assessment of heavy metals displayed that among 12 heavy metals in  $PM_{2.5}$  during the spring festival in the Dalian atmosphere, the non-carcinogenic risk of V exceeded the EPA limit value of one, and there was a non-carcinogenic risk to human health. Among As, Cd, Co, Cr,

and Ni, the lifetime cancer risk values of As, Cd, and Cr were higher than the threshold ( $10^{-6}$ – $10^{-4}$ ), so it is necessary to monitor and control them.

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