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Short Communication

Arsenic Accumulation and Speciation of $PM_{2.5}$ and Relevant Health Risk Assessment in Jinan, China

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Abstract

Arsenic (As) is a ubiquitous carcinogen in the environment. Its bio-toxicity is significantly correlated with its chemical forms. In this study, As accumulation and speciation of $PM_{2.5}$ was investigated in Jinan. The high $PM_{2.5}$ levels fluctuated between 69.67 $\mu g \cdot m^{-3}$ and 211.25 $\mu g \cdot m^{-3}$ in winter, and ranged from 63.46 $\mu g \cdot m^{-3}$ to 125.50 $\mu g \cdot m^{-3}$ in summer. The total As concentration of $PM_{2.5}$ in winter and summer varied from 4.23 to 15.47 $ng \cdot m^{-3}$ and from 4.59 to 11.69 $ng \cdot m^{-3}$. As(V) accounted for 70~100% of the extractable speciation in the $PM_{2.5}$ samples. Exposure levels of total As for the general public were 63.45~232.05 ng d^{-1}, and 68.85~175.35 ng d^{-1} by inhalation in winter and summer. The mean of cancer risk (CR) of As in $PM_{2.5}$ for winter and summer were $7.30\times10^{-6}\pm0.89\times10^{-6}$ and $5.26\times10^{-6}\pm0.58\times10^{-6}$, respectively.

Keywords: $PM_{2.5}$, arsenic, speciation distribution, health risk, Jinan

Introduction

Arsenic is a ubiquitous carcinogen and mutagen that has created serious human health problems around the world. It is released into the environment from both anthropogenic and natural sources [1]. Many studies have demonstrated anthropogenic sources such as the combustion of fuels, and the use of pesticides contributes As at three times higher levels than natural ones [2]. As toxicity and migration are significantly correlated with its speciation and oxidation state in the environment. In general, inorganic As species are more toxic than organic As species, As_2O_3(As(III)) is more toxic than As_2O_5(As(V)). As exposure has been reported to be related to several types of cancer, including skin, lung, and urinary bladder [3-4]. Therefore, it has aroused a global concern and substantial investigation in various environments in past decades.

Humans are exposed to As through oral ingestion, such as food and water, but inhaled exposure is thought to be another important As source in industrial areas. It was reported that As concentration in air from urban and industrial districts was significantly higher than those in rural areas [4]. Previous studies indicated that more than 90% of atmospheric As exists mainly in fine airborne

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particles with diameters smaller than or equal to 3.5 μm. Compared with coarse grains, more than 80% of the airborne particles smaller than 2.5 μm can get into the pulmonary alveoli, where they can be retained and stay for months to years [5]. Therefore, it is essential to get a better understanding of the As real polluted levels and relative risk assessment, especially the distribution of different As species in $PM_{2.5}$. As speciation studies in total suspended particles (TSP) and PM_{10} in Huelva suggested that the presence of As(III) was directly related to an anthropogenic source [2]. To data, relative studies have focused on total As concentrations and species in TSP and PM_{10}. Information on As levels and speciation in $PM_{2.5}$ is limited.

Shandong Province is located in eastern China, adjacent to Korea and Japan. And although it occupies only 1.6% of China, it contributed 9% of China's $PM_{2.5}$ emissions in 2006 [6]. In the past few years, it has caused international attention for its $PM_{2.5}$ pollution because of its special geographical position. Regional migration of atmospheric pollutants from the Shandong area was thought to be a source of aerosol pollution in Beijing under prevailing south and southeast winds [7]. Furthermore, Shandong was considered to be a potential source of secondary inorganic aerosols in Seoul, Korea [8]. As the capital of Shandong Province, Jinan suffered a serious $PM_{2.5}$ pollution in past decades, and posed harmful health risks on citizens. Cheng et al. (2011) reported that the annual average concentration of $PM_{2.5}$ in Jinan was as high as 148.71 $\mu g \cdot m^{-3}$, which was one of the highest levels reported in the world [9]. In past decades, the consumption of fossil fuels was increasing with rapid industrialization and urbanization. However, coal was still the major fuel widely used for industrial processes and daily life in Jinan, and much more coal was combusted during the heating period (from November to March). Coal burning is mainly responsible for the severe air pollution in northern Chinese cities, e.g., by As. A large amount of coal combustion may lead to atmospheric As pollution in Jinan. Up to now, studies about $PM_{2.5}$ in Jinan were mainly on chemical characteristics of $PM_{2.5}$ and the formation mechanism of secondary aerosols [10]. Research on As concentrations and speciation in $PM_{2.5}$ is limited.

The main objectives of this study were:

1) To present As levels and speciation in $PM_{2.5}$ with the optimized extraction method in February and August 2014.
2) Principal component analysis (PCA) was used to reveal the influence of factors on As levels in $PM_{2.5}$.
3) To estimate cancer risk (CR) and respiratory exposure value of As.

Our results may be of great importance for knowing toxicological properties of As in $PM_{2.5}$, and provide powerful data to assess the potential human health risk of As by inhaled exposure.

Materials and Methods

Sample Collection

Samples were collected in two periods in 2014: winter (February 12-20) and summer (August 23-31). The location of the sampling site was shown in Fig. 1. It is located in the atmospheric monitoring station in Quancheng Square in central Jinan city (117°01′10″E, 36°39′40″N). The site was about 10 m above ground level. Air samples were typically collected daily using a modified moderate-volume air sampler (Tianhong instrument Co., Ltd, China), from 08:00 to 06:00 the next day. The calibrated airflow was 131~135 $m^3 \cdot d^{-1}$. The glass fiber membrane was barked for 6 h in the muffler furnace at 600ºC to remove the organic compositions before sampling. The membrane was dried for 24 h in the dryer before and after sampling, and then was weighed after being placed at room temperature for 24 h. After weighting, the membrane was placed in the refrigerator at -20ºC under conditions of seal preservation.

Chemical Analysis

Analysis of Total As in $PM_{2.5}$

The filters were digested with 10 mL nitric acid overnight and then transferred to a heating plate. The remainder of the processing and analysis steps

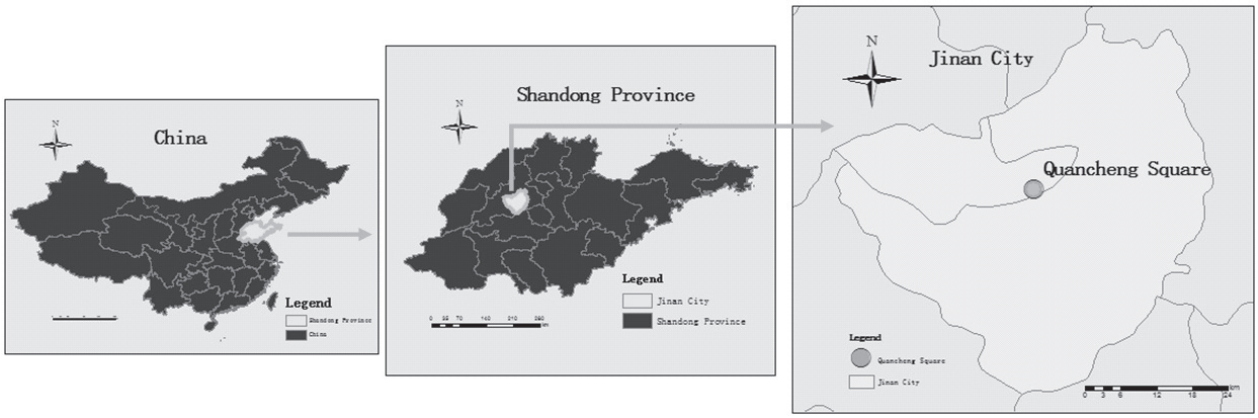

Fig. 1. The sampling location in Jinan, China.

were described in detail by Sun et al. [12]. Total As concentrations were determined by hydride generation atomic fluorescence spectrometry (HG-AFS) (PF52, Beijing Puxi Analytical Instruments Co., Beijing, China).

Analysis of Arsenic Speciation in $PM_{2.5}$

The filter membrane was digested in 5 mL 1% nitric acid in a centrifuge tube oscillating overnight, and transferred to a microwave-accelerated reaction system (ETHOS ONE, Milestone, Italy) with the following temperature program: 55ºC for 10 min, 75ºC for 10 min, and 95ºC for 30 min. The digest solutions were centrifuged and passed through a 0.45 μm nylon filter. In order to minimize species transformation, samples were determined within a few hours of filtration and kept in the dark and in a refrigerator. As speciation was assayed simultaneously by HPLC-ICP-MS (NexION 300, PerkinElmer, USA). The analytical procedures and conditions were adapted from a previous study [13].

PCA Analysis

PCA is a widely used statistical technique originating from multivariate statistical analysis, which allows us to identify the major factors within a certain multidimensional dataset [14]. It can also be applied for explaining the data in a way that highlights similarities and differences. In this study, PCA was conducted by software CANOCO 4.5 bundled with CanoDraw for Windows.

Health Risk Assessment

Higher As pollution levels via inhalation may result in higher cancer risks for residents in Jinan; therefore, cancer risk (CR) had been done in this study. The calculation was followed by the U.S. Environmental Protection Agency's Exposure Factors Handbook [15]:

$$ECinh = (C \times ET \times EF \times ED)/AT \quad (1)$$

$$CR = IUR \times ECinh \quad (2)$$

…where C is contaminant concentration in $PM_{2.5}$ ($\mu g/m^3$), ET is exposure time (hours/day), EF is exposure frequency (180 days /year in this study), ED is exposure duration (24 years for adults), and AT is average aging time = 70 years×365 days/year ×24 h. All parameters used in the calculation of EC were found in reports published by the U.S. EPA during different periods (U.S. EPA, 2011a). CR is carcinogenic risk and IUR us inhalation unit risk; for arsenic it's 4.3×10^{-3} $(\mu g \cdot m^{-3})^{-1}$ from EPA's integrated risk information system.

Statistical Analysis

All statistical analyses were performed with the use of origin 9.0. PCA was performed by CANOCO 4.5.

Quality Control

Blank and standard membranes were used to check accuracy during sampling and weighing, with one sample corresponding to one blank. The detection limits were calculated using three times the signal-to-noise ratio. The detection limits of the four arsenic species (As(III), DMA, MMA, and As(V)) were in the range of 0.6~1 $\mu g \cdot L^{-1}$. The determination precisions of four arsenic species were 1.26%, 1.24%, 1.00%, and 0.79%, respectively. The standard solutions were added into the sample to check the stability of four arsenic species during the digestion process. The recoveries of As(III), DMA, MMA, and As(V) were 87.4%, 94.4%, 96.8%, and 98.0%, respectively. Therefore, all arsenic species were kept stable during the course of the determination.

Results and Discussion

$PM_{2.5}$ Levels in Winter and Summer

The average contents of $PM_{2.5}$ in two sampling periods were shown in Fig. 2. The $PM_{2.5}$ concentrations fluctuated between 69.67 μg m^{-3} and 211.25 μg m^{-3}, with a mean level of 136.34±15.23 μg m^{-3} in winter. The levels ranged from 63.46 μg m^{-3} to 125.50 μg m^{-3}, with a mean value of 87.29±7.18 μg m^{-3} in summer.

Average concentration of $PM_{2.5}$ in winter was almost two times more than the Chinese national secondary standard limit value of 75 μg m^{-3} (GB 3095–2012) for mixed commercial, traffic, and residential areas, and the average concentration in summer also exceeded the standard limit value as shown in Fig. 2. Similar results were found in urban Beijing during the heating (122.09 μg m^{-3}) and non-heatings period (88.99 μg m^{-3}) [16]. Ye et al. (2003) reported that $PM_{2.5}$ concentrations for Shanghai in summer and winter were 64.8 and 88.6 μg m^{-3}, respectively. As a developing city in China, Jinan has a relatively higher concentration of $PM_{2.5}$ than other developed cities [6]. Seasonal patterns of $PM_{2.5}$ in

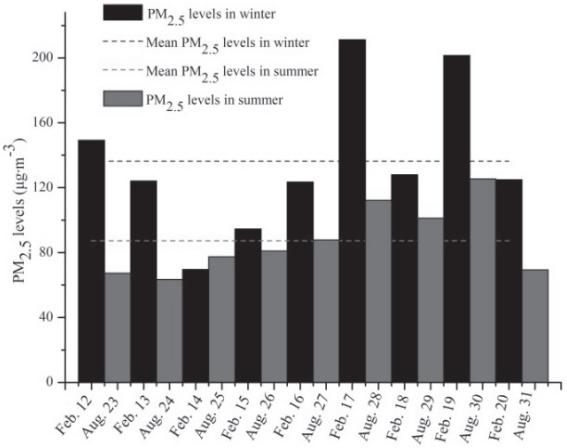

Fig. 2. $PM_{2.5}$ levels in winter and summer in Jinan.

Jinan are similar to other cities. Several factors may effect $PM_{2.5}$ concentrations between winter and summer. Firstly, coal is the primary energy source for Jinan, accounting for more than 70% of all energy consumed in Jinan. Continued increases in coal consumption and incomplete pollution control measures during the heating period led to higher $PM_{2.5}$ pollution [6, 10]. Secondly, the sampling period in winter fell during the traditional Chinese Spring Festival, and numerous studies have demonstrated that firework and firecracker emissions were another source of $PM_{2.5}$ [17-18]. Thirdly, Jinan is surrounded by mountains, which makes pollutants difficult to disperse [19]. Finally, the wind speed and temperature were both low in winter, thus favoring the accumulation of pollutants. Otherwise, more precipitation led to a lower $PM_{2.5}$ concentration in summer.

Total As and its Species in $PM_{2.5}$

The total As concentrations of $PM_{2.5}$ in two sampling periods are shown by Fig. 3 to vary from 4.23 to 15.47 ng m^{-3} in winter and from 4.59 to 11.69 ng m^{-3} in summer. The average concentration of As in winter and summer was 10.04±1.22 ng m^{-3} and 7.23±0.80 ng m^{-3}, respectively. The As concentrations in winter and summer exceeded an estimated target concentration for $PM_{2.5}$ (6 ng m^{-3}) in a previous study [19]. Higher $PM_{2.5}$-bound As concentrations were observed at megacities, such as Guangzhou (17.58 ng m^{-3}), Beijing (20~60 ng m^{-3}), Xi'an (80 ng m^{-3}), and Shanghai (30 ng m^{-3}) [18, 22-25]. Relatively lower As concentrations were found in Aspropyrgos, Greece (1.9±0.3 ng m^{-3}); Huelva, Spain (7.7 ng m^{-3}); St. Louis, USA (3 ng m^{-3}); Indianapolis, USA (2 ng m^{-3}); and Toronto, Canada (0.4 ng m^{-3}) [5, 24-25]. As a developing city in China, the total As levels in Jinan were relative lower than larger cities in China. However, compared with cities in developed countries, the As pollution in Jinan may be worse, which indicated that there was ahigher risk of respiration exposure for humans in Jinan.

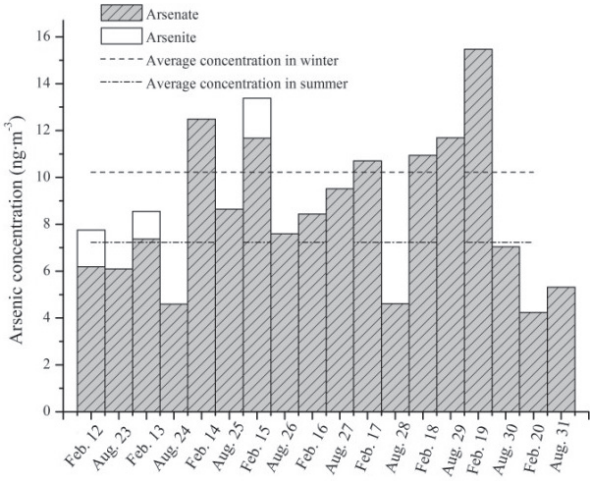

Fig. 3. Arsenic species of $PM_{2.5}$ in Jinan.

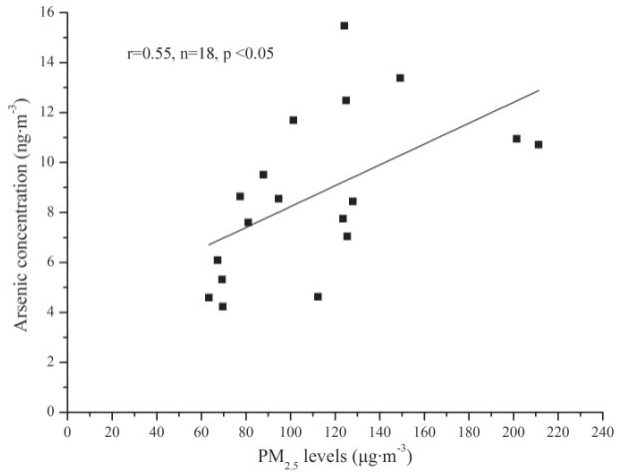

Fig. 4. Relationship of $PM_{2.5}$ levels with As concentrations in winter and summer in Jinan.

Arsenic species of $PM_{2.5}$ were analyzed for the first time in Jinan. As(III) and As(V) were found synchronously in winter samples. However, only As(V) was detected in summer samples. Organoarsenic was not found (Fig. 3). The results indicate that As(V) was always the main As speciation in all the samples, accounting for more than a 70% of total arsenic, with a maximum concentration 15.47 ng m^{-3}. However, As(III) was detected only in some winter samples. As we know, As(III) is not stable and can be relatively rapidly oxidized to As(V) in the atmosphere by such oxidants as ozone [24]. Finding As(V) to be the dominant As species in samples was consistent with studies from other locations [4, 26]. Emissions of arsenic from coal burning are primarily the reduced oxide As_2O_3, and are associated with fine particles [23]. Since coal sources dominate $PM_{2.5}$ in Jinan, especially in winter, the resulting As(III) released must be relatively rapidly oxidized to As(V). A small amount of As(III) found in samples in winter may be attributed to incomplete oxidation. It is noteworthy that As(III) is more toxic than As(V); therefore, the relatively low percentage of As(III)

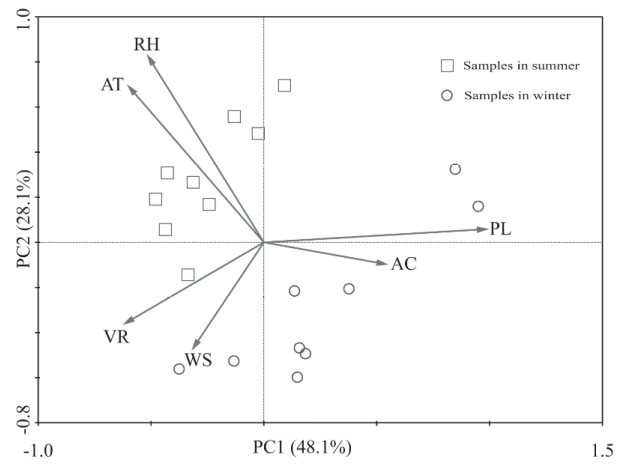

Fig. 5. Principal component analysis (PCA).

Table 1. Comparison of excess cancer risks of As in $PM_{2.5}$ of this study with values from other cities.

City	Mediums	Excess cancer risk	Reference
Jinan, China	$PM_{2.5}$	$5.26\pm0.58\times10^{-6}$~$7.30\pm0.89\times^{10-6}$	This study
Seoul, Korea	$PM_{2.5}$	6.5×10^{-6}	Park et al. (2008)
Xining, China	$PM_{2.5}$	2.16×10^{-6}	Shi et al. (2014)
Nanjing, China	$PM_{2.5}$	9.16×10^{-6}	Hu et al. (2012)

cannot be ignored. Previous studies have demonstrated that chronic exposure to inorganic arsenic may give rise to severe health effects on humans [18]. Hence the composition of inorganic arsenic in the air may cause certain toxic effects on human in Jinan.

Determining Factors that Influence As

The levels of $PM_{2.5}$ depended on many meteorological factors, such as wind speed, humidity, etc. [22]. Therefore, these meteorological factors may also influence the arsenic concentration of $PM_{2.5}$. There was no significant correlation between the total content of As and relative humidity, temperature, and wind speed (data unshown). Similar results were found in-between As in TSP and meteorological factors [11]. However, a positive correlation between $PM_{2.5}$ levels and As concentrations was shown in Fig. 4. This may explain why higher As concentrations occurred in winter. Coal combustion was also identified as a predominant source of arsenic [6], which contributed to higher As pollution in winter.

Two principal components were obtained and accounted for 76.2% of the total variance (Fig. 5). Principal component 1 accounted for 48.1% of the total variance, and principal component 2 could explain 28.1%. This indicated that $PM_{2.5}$ levels were the main controlling factor of As in $PM_{2.5}$, and a significantly positive relation was shown between $PM_{2.5}$ levels and As concentrations in $PM_{2.5}$. Meteorological factors such as wind speed, temperature, and relative humidity may have a negative correlation with As in $PM_{2.5}$. Frequent high wind speed would make the local As pollution easy to disperse [27].

Determining Cancer Risk

The mean As pollution levels in Jinan exceeded the European Union limit (6 ng m^{-3}) [28] and a nearly six times the WHO value (0.66 ng m^{-3}). Up to now, major concerns were paid to the As contamination in drinking water. It has been reported that more than 20,000 arsenicosis patients are confirmed annually in China [9]. In fact, more people could be at risk from aerosol As pollution via inhaled exposure. Here we assumed 100% of As entered lungs through respiration. Assuming that about 15 m^3 of air was inhaled per day for adult, then the exposure of total As for the general public in Jinan was 63.45~232.05 ng d^{-1} and 68.85~175.35 ng d^{-1} by inhalation in winter and summer, respectively. The values were higher than the U.S. standard (40~90 ng d^{-1}).

Table 1 compared city-specific results with CR of As in $PM_{2.5}$ from other cities (i.e., Seoul, Xining, and Nanjing). The CRs of these cities were all in a magnitude of 1×10^{-6}~1×10^{-5}, indicating that the cancer risks lower than 1×10^{-4} are generally considered to be acceptable [29], but should not be negligible (>1×10^{-6}) [30]. The mean CR of As in $PM_{2.5}$ for winter and summer were $7.30\times10^{-6}\pm0.89\times10^{-6}$ and $5.26\times10^{-6}\pm0.58\times10^{-6}$, respectively. Despite the differences between seasons, average CR in this study ranged from 2 to 3 times higher than the values in Xining and had no significant differences with Seoul and Nanjing [31-32]. Since As in $PM_{2.5}$ can access lung cells without any resistance, it can pose a health risk for months to years. Further research should be developed to realize the underlying long-term risks of different As species in $PM_{2.5.}$

Conclusion

In this study, we investigated As accumulation and speciation of $PM_{2.5}$ in Jinan, which is suffering from severe $PM_{2.5}$ pollution. The average concentrations of As in winter and summer were 0.04 ± 1.22 ng m^{-3} and 7.23 ± 0.80 ng m^{-3}, respectively. As(V) was the predominant speciation in $PM_{2.5}$, accounting for 70~100% of total arsenic. Meanwhile, the cancer risk (CR) from As in $PM_{2.5}$ was assessed. The results showed that more concerns should be paid to the As-caused potential health risks of $PM_{2.5}$ in Jinan.

Acknowledgements

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