Environmental Science and Pollution Research (2020) 27:21654–21665
https://doi.org/10.1007/s11356-020-08626-2

RESEARCH ARTICLE

Temporal trend of arsenic in outdoor air $PM_{2.5}$ in Wuhan, China, in 2015–2017 and the personal inhalation of PM-bound arsenic: implications for human exposure

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Received: 14 January 2020 / Accepted: 26 March 2020 / Published online: 11 April 2020

Abstract

Arsenic in fine air particulate matter ($PM_{2.5}$) has been identified as an important factor responsible for the morbidity of lung cancer, which has increased sharply in many regions of China. Some reports in China have shown that arsenic in the air exceeds the ambient air quality standard value, while long-term airborne arsenic concentrations in central China and human exposure via inhalation of PM–bound arsenic (inhalable airborne PM) have not been well characterized. In this study, 579 outdoor air $PM_{2.5}$ samples from Wuhan, a typical city in central China, were collected from 2015 to 2017, and arsenic was measured by inductively coupled plasma-mass spectrometry. Personal exposure to PM-bound arsenic via inhalation and urinary arsenic concentration were also measured. The concentrations of arsenic in $PM_{2.5}$ were in the range of 0.42–61.6 ng/m^3 (mean 8.48 ng/m^3). The average concentration of arsenic in 2015 (10.7 ng/m^3) was higher than that in 2016 (6.81 ng/m^3) and 2017 (8.18 ng/m^3), exceeded the standard value. The arsenic concentrations in spring and winter were higher than those in summer and autumn. No significant differences ($p > 0.05$) were found among different sites. The daily intake of arsenic inhalation based on PM_{10} samples collected by personal samplers (median, 10.8 ng/m^3) was estimated. Urban residents inhaled higher levels of PM-bound arsenic than rural residents. Daily intake of arsenic via inhalation accounted for a negligible part (< 1%) of the total daily intake of arsenic (calculated based on excreted urinary arsenic); however, potential associations between the adverse effects (e.g., lung adenocarcinoma) and inhaled PM-bound arsenic require more attention, particularly for those who experience in long-term exposure. This study is the first report of a 3-year temporal trend of airborne $PM_{2.5}$-bound arsenic in central China.

Keywords Arsenic · $PM_{2.5}$ · Human exposure · Urine · Personal exposure

Xiang Mao and Xun Hu contributed equally to this work.

Responsible editor: Philippe Garrigues

Electronic supplementary material The online version of this article (https://doi.org/10.1007/s11356-020-08626-2) contains supplementary material, which is available to authorized users.

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Introduction

Assessing human exposure to arsenic is of considerable interest for public health, particularly in relation to the long-term exposure. To date, arsenic has been widely studied in ambient air (Duan and Tan 2013; Huang et al. 2014a; Li et al. 2015; Sánchez-Rodas et al. 2015; Santos and Fernández-Olmo 2016), water (IARC 2004; Liao et al. 2018; Rahman et al. 2017), soil and dust (Cao et al. 2016), and foodstuffs (Davis et al. 2017; Jiang et al. 2015; Mantha et al. 2016) because of its adverse effects, including cancers (Abernathy et al. 1999).

Based on data from laboratory experiments on animals and from occupational experiments from smelters, inorganic arsenic compounds were classified as group 1 human carcinogens (IARC 1987; IARC 2004). The lung is the principal site for arsenic carcinogenicity (European Commission 2001). When arsenic is inhaled, the accumulation level can be as high as 8 times; thus, a higher risk of lung cancer is expected to be

related to a higher amount of inhaled particulate matter (PM)–bound arsenic (European Commission 2001). Inhaled PM-bound arsenic exposure was high during the past decades in mainland China (Duan and Tan 2013), and it could be associated with the increasing morbidity of lung cancer (She et al. 2013; Yoshikawa et al. 2008), particularly lung adenocarcinoma (Guo et al. 2004; Kuo et al. 2017).

Additionally, arsenic in air PM may pose risks to vulnerable populations (Liao et al. 2018; Liu et al. 2018; Rahman et al. 2017; Yang et al. 2015); it has also been associated with heart diseases (Keil and Richardson 2017), and acute lower respiratory infections in children (Wang et al. 2018).

Arsenic contained in airborne PM has been attributed to coal combustion (Liu et al. 2002), resuspended dust, and industrial and vehicle exhaust (Tao et al. 2014). Urbanization and industrialization increase the concentration of arsenic contained in airborne PM, resulting in increased human exposure to airborne PM-bound arsenic through inhalation (Fishbein 1984). More importantly, inorganic arsenic was identified as the predominant species in airborne PM, with a proportion as high as 98% in different cities among various countries (Gonzalez-Castanedo et al. 2015; Huang et al. 2014a; Oliveira et al. 2005; Tsopelas et al. 2008; Yang et al. 2012). Based on the genotoxic properties of arsenic, the WHO has recommended 6.6 ng/m^3 as the target value of arsenic in ambient air; China (GB 3095-2012) (Duan and Tan 2013) and several countries in the European Union (EU) have suggested a target value of 6 ng/m^3 (European Commission 2004); and the United States (US) recommends a target value of 4.29 ng/m^3 (Lewis et al. 2015). However, a study in Japan suggested the association between lung cancer and air PM-bound arsenic concentration of 1.77 ng/m^3 or higher (Yoshikawa et al. 2008). Recently, some studies in China (Huang et al. 2014a; Li et al. 2015; Tao et al. 2014) reported that the arsenic concentration in PM has exceeded the recommended standard value (Duan and Tan 2013), while other toxic metals did not. Very high concentrations of arsenic in air (66–70 ng/m^3) have been reported in Wuhan, mainly due to the industries, including 11 coal-fired power plants, one of the largest steel companies in China and 11 cement companies (Lv et al. 2006; Querol et al. 2006). High levels of arsenic in atmospheric air (34.7–49.4 ng/m^3) were still observed before 2014 (Zhang et al. 2015); however, the current level and trend of airborne PM-bound arsenic in Wuhan has not been reported after the strategy of cutting excess production capacity in the steel and coal industries was implemented in China.

The temporal (several years) profile of $PM_{2.5}$-bound arsenic in central China has not yet been well documented (Duan and Tan 2013). Since human health can be impacted by chronic exposure to arsenic, long-term and systematic observation is needed to assess dynamic human exposure. In this study, 579 outdoor air samples from Wuhan, central China, collected over 3 years were analyzed to identify whether the general population is still being exposed to high concentrations of $PM_{2.5}$-bound arsenic and whether there are differences in the exposure levels among various districts. Additionally, human exposure to PM-bound arsenic was calculated via PM_{10} samples collected by personal samplers ($n = 54$), and the amount was compared with the total daily intake of arsenic calculated based on their total urinary arsenic concentrations.

Materials and methods

Reagents

Standards for arsenic (single-species pentavalent arsenic standard, NIST SRM# 3103a) (Yu et al. 2006) measurement (IV-ICPMS-71A) and internal standard (yttrium in IV-ICPMS-71D) were purchased from Inorganic Ventures (Christiansburg, VA, USA). Nitric acid (68%) was purchased from Fisher Scientific (Gent, Belgium). Ultrapure water (Millipore, Burlington, MA, USA) was used for analysis.

Sample collection

From January 2015 to December 2017, 579 outdoor air $PM_{2.5}$ samples were collected from 5 sites in Wuhan, the capital city of Hubei Province and a representative city in central China, including Qingshan District ($n = 237$) and Wuchang District ($n = 243$) from 2015 to 2017 (5 to 7 samples evenly every month) and Dongxihu, Hanyang, and Jiang'an ($n = 33$ each, during 2017 only, typically 3 samples each month) (Table S1). The sampling was conducted simultaneously at different sampling sites on days without rain (Table S1 and Figure S1).

Medium-volume samplers (Wuhan Tianhong Environmental Protection Industry Co., Ltd., TH-150C; flow rate 100 L/min) were placed 10–15 m above the ground (HJ/T 194-2005, China) in urban areas characterized by a lack of major industrial activities within a 5-km range from the sampling point. Quartz fiber filters were applied to collect $PM_{2.5}$ samples for 22 h (90 mm, Whatman International Ltd., Maidstone, England). The sampling date, the standardized air volume, and other related information were recorded. The samples were maintained and transported individually in a polystyrene box at 4°C until analysis as described elsewhere (Li et al. 2018).

Previous studies applied outdoor and indoor airborne PM-bound arsenic from specific sampling sites for human exposure assessment (Cao et al. 2016). To better understand personal dynamic exposure to PM-bound arsenic via inhalation of air, including outdoor air and indoor air, PM_{10} air samples were collected by personal air samplers carried by the participants in this study, and personal exposure to $PM_{2.5}$-bound arsenic could be estimated by its partitioning ratio (64% in Taiwan and 63% in Greece) in $PM_{2.5}/PM_{10}$ according to the

previous study (Tsopelas et al. 2008; Ying et al. 2003). Thus, personal PM_{10} samples were collected by personal air samplers (LP-5, A. P. Buck Inc. Orlando, FL, USA) (Li et al. 2016) from 26 male and 28 female healthy residents aged between 18 and 65 in Wuhan; samples were collected in 3 consecutive days (2 L/min, 37 mm Whatman quartz fiber filter) from October to November 2016, and their spot urine samples ($n = 54$) were collected on the third day; urine samples were also collected from the participant's neighbors ($n = 66$, one urine sample from each participant).

Sample preparation and instrumental analysis

According to our previous reports, total arsenic in the outdoor air $PM_{2.5}$ samples, personal air PM_{10} samples (Li et al. 2018), and urine samples (Liu et al. 2018) were analyzed without modification.

Procedural blanks, duplicates, and matrix spikes were performed every 20 samples. The accuracy was assessed by reference materials (NIST SRM 1648A and 2668). The limit of quantification (LOQ) of arsenic was calculated with a signal/noise (S/N) ratio of at least 10, namely 0.10 μg/L. The LOD was set as S/N ≥ 3. As arsenic was not found in any kind of blank samples, the method detection limit (MDL) was calculated from an S/N ratio of 10 (Van den Eede et al. 2015; Wang et al. 2019). The MDLs of arsenic in the $PM_{2.5}$ and PM_{10} samples were 0.17 and 0.34 ng/m^3, respectively. The MDL of arsenic in urine samples was 0.20 μg/L. The recoveries of arsenic in matrix spiked samples were 92.3–104%.

Estimation of daily exposure

The estimated daily intake (EDI; ng/kg body weight (bw)/day) of arsenic through inhalation of PM_{10} was calculated with the following Eq. (1) (USEPA 2011), assuming a 100% absorption rate from inhalation to blood circulation.

$$\mathrm{EDI}_{\mathrm{inhalation}} = (C \times \mathrm{AIR})/\mathrm{BW} \quad (1)$$

where C is the arsenic concentration (median and 95th percentile) in personal air samples (ng/m^3), AIR is the air inhalation rate (m^3/day), and BW is the body weight (kg), as previously described (Li et al. 2018). Details about the parameters are shown in Table S2.

The urinary concentration of arsenic was used to estimate the total daily human exposure to bioavailable arsenic (Li et al. 2017) with the Eq. (2) below, as described elsewhere (Liao et al. 2012).

$$\mathrm{EDI}_{\mathrm{urine}} = C \times V/\mathrm{BW} \quad (2)$$

where C (μg/L) and V (L) represent the total urinary arsenic concentration (Caldwell et al. 2009; Liu et al. 2018) and urine volume, respectively.

Statistical analysis

Statistical analysis was conducted with SPSS 23.0. The residuals were not normally distributed (with Shapiro-Wilk normality test). After natural log-transformation, the residuals were normally distributed. The homoscedasticity of the variance was verified by Levene's test and the variance was equal. Then, T tests or one-way ANOVA was used to identify the differences between/among groups.

Results and discussion

Concentrations of arsenic in outdoor $PM_{2.5}$

Arsenic was detected in all collected $PM_{2.5}$ samples, with values ranging from 0.42 to 61.6 ng/m^3 (mean, 8.48 ng/m^3; median, 5.89 ng/m^3). The mean arsenic concentrations from Wuhan (Wuchang and Qingshan) in 2015 (10.7 ng/m^3; median, 7.92 ng/m^3) were significantly higher than those in 2016 (6.81 ng/m^3; median, 4.85 ng/m^3) and 2017 (7.81 ng/m^3; median, 5.25 ng/m^3), while there was no significant difference between 2016 and 2017 (Table 1). The decrease in arsenic in air from 2015 to 2016 and 2017 could be explained by the reduced steel production of the Wuhan Steel Company, which is one of the largest steel companies in China (Lv et al. 2006).

The mean concentration of $PM_{2.5}$-bound arsenic in 2017 at each sampling site was, in decreasing order, as follows: Jiang'an (9.56 ng/m^3) > Dongxihu (8.57) > Qingshan (8.30) = Hanyang (8.30) > Wuchang (7.31) (Table S3). The median concentration in 2017 was as follows: Jiang'an (6.33 ng/m^3) > Qingshan (5.82) = Hanyang (5.42) > **Dongxihu (4.87)** > Wuchang (4.47) (Table S3), and there were no significant differences among the studied sites. No significant difference was observed between Wuchang and Qingshan (these were the only 2 sites observed for 3 years) during each year (2015–2017).

The $PM_{2.5}$-bound arsenic concentrations could be attributed to coal combustion (such as thermal power plants in Wuhan), traffic exhaust, and other industries in Wuhan. No significant differences in air $PM_{2.5}$-bound arsenic concentrations were observed among the sampling sites, which was consistent with a previous conducted from 2003 to 2004 (Lv et al. 2006), and this result could be explained by the widespread arsenic pollution present in the urban areas of the entire city, regardless of whether the area is located in an industrial district, commercial area, traffic area, or relatively clean air area.

China witnessed a decline in its PM concentration during 2001–2010 (Jacquemin et al. 2015). A similar decline was observed for arsenic in air PM in Wuhan, China (Table 2), from 66–70 ng/m^3 in PM_{10} during 2003–2004 (Lv et al. 2006; Querol et al. 2006) and 34.7–49.4 ng/m^3 in $PM_{2.5}$

Table 1 Concentrations of arsenic (ng/m^3) in outdoor air $PM_{2.5}$ collected from 2 sites during 2015–2016 and 5 sites during 2017 in Wuhan

Year	Site	*N*	Mean	SD	GM	Median	5th	95th	Range
2015	2 in 1	156	10.7	9.38	7.47	7.92	7.00	9.04	0.76–61.6
2016	2 in 1	158	6.81	6.04	4.91	4.85	4.07	5.77	0.68–43.0
2017	2 in 1	166	7.81	7.30	5.40	5.25	4.47	6.21	0.42–38.7
2017	5 in 1	265	8.18	7.55	5.77	5.51	4.84	6.27	0.42–41.4
2015–2017	Total	579	8.48	7.85	5.92	5.89	5.40	6.58	0.42–61.6
*Sig.	/	/	/	/	/	1 vs. 2 1 vs. 3	/	/	/

GM, geometric mean

2 in 1 (Wuchang and Qingshan) or 5 in 1 (Wuchang, Qingshan, Dongxihu, Hanyang, and Jiang'an) indicates 2 or 5 sampling sites combined, respectively

*Significant differences ($p < 0.05$) were observed among 3 years (1 means 2015, 2 means 2016, and 3 means 2017). Significant differences were compared with ANOVA

during 2012–2013 (Zhang et al. 2015) to 8.48 ng/m^3 in $PM_{2.5}$ during 2015–2017.

Data on air arsenic concentrations from cities in China and other countries are summarized in Table 2 and Table S4. The highest air arsenic concentration from China was reported in Guangxi (a mining town, mean 21.8 μg/m^3) (Zhang et al. 2009), followed by Hunan (a school in an industrial city, median 380 ng/m^3 (Cao et al. 2016), and another place close to a battery plant, median 210 ng/m^3 (Cao et al. 2015)) (Table S4). The arsenic in the air PM of these extremely polluted areas was 10–1000 times higher than that recorded in the Chinese megacities of Beijing (mean 7.6–32.6 ng/m^3) (Tao et al. 2014; Wu et al. 2013a; Wu et al. 2013b; Wu et al. 2015), Shanghai (7.9–30.8) (Chen et al. 2008; Ming et al. 2015), Guangzhou (17.6) (Huang et al. 2014a; Huang et al. 2014b), and Shenzhen (28.6) (Duan and Tan 2013) (Table 2) and in some other capital or industrialized cities (Table S4).

Generally, the airborne arsenic concentrations in most areas of China were much higher than those in most cities of Europe and North America (Table 2). The lowest mean concentration of arsenic was reported in New Zealand, at only 5.6×10^{-3} ng/m^3 (Marx et al. 2014). The arsenic concentration (mean 8.49 ng/m^3) in this study was 10–20 times higher than that of most values reported in European countries, such as in Spain (Barcelona) (0.5 ng/m^3) (Rivas et al. 2014) in southern Europe, where the densities of population and motor vehicles were also high. The concentration of arsenic in this study was also higher than those reported in the US, including Washington, DC (mean ~ 1.0 ng/m^3) (Greene and Morris 2006) and Greater Houston, TX (Liu et al. 2016); and those in Europe, including Turkey (mean 2.23–5.55 ng/m^3) (Onat and Şahin 2014) and Greece (mean 1.42–2.39 ng/m^3) (Thomaidis et al. 2003); and those in Asia, including Visakhapatnam, India (2.5–5.3 ng/m^3) (Police et al. 2016).

Currently, the airborne arsenic concentrations reported in most cities of China (Duan and Tan 2013) is higher than the ambient air target value of 6.6 ng/m^3 recommended by the WHO, In addition to China, only a few other countries (regions) have reported high air arsenic concentrations so far, including Bor, Serbia (Serbula et al. 2010; Serbula et al. 2017), some parts of Poland (Widziewicz et al. 2016), and Pakistan (von Schneidemesser et al. 2010) (Table 2). Meanwhile, in China, the average concentrations of PM-bound arsenic in only a few cities/regions, such as Chengdu (5.9 ng/m^3) (Table S4), Lhasa (1.8 ng/m^3), Hong Kong (3.9–6.0 ng/m^3) (Duan and Tan 2013; Pun et al. 2014), and Taiwan (3.15–4.99 ng/m^3) (Fang et al. 2012; Ying et al. 2003) (Table 2), were lower than the target value. Further research on the distribution of arsenic in air PM from China and its health effects is urgently required.

Seasonal variation

The seasonal variation in the arsenic concentrations in air $PM_{2.5}$ is shown in Table 3 and Table S5. Summer had the lowest concentration of arsenic in air $PM_{2.5}$ in 2015, while there were no significant differences among spring, autumn, and winter. During 2016, significant differences were observed between spring and summer/autumn. Autumn had the lowest concentration in 2017. Data on arsenic in outdoor air $PM_{2.5}$ in Wuhan for each month during 2015–2017 are shown in Table S6.

Overall, no significant difference in air $PM_{2.5}$-bound arsenic was observed between winter and spring in the present study. This result could be explained by the unfavorable atmosphere for diffusion, sandstorm migration from North China (Zhang et al. 2015), or biomass burning during spring (Querol et al. 2006). The seasonal pattern showed that the arsenic in $PM_{2.5}$ in summer and autumn was lower than that in spring and winter, which was consistent with our previous study about nickel in air PM for most inland regions in mainland China (Li et al. 2018). The seasonal pattern could be

Table 2 Arsenic concentrations in air particulate matter from other published data

Cities	$PM_{2.5}$ (ng/m^3)	PM_{10} (ng/m^3)	TSP (ng/m^3)	Sampling period	References
Wuhan*		66–70		Twice every other week from Sep 2003 to Sep 2004	Lv et al. 2006; Querol et al. 2006
Wuhan*	34.7–49.4			Every sixth day from Aug 2012 to Jul 2013	Zhang et al. 2015
Wuhan*	8.48			2015-2017	This study
Beijing, China*	16.0–32.6			2008–2012	Wu et al. 2013b
Beijing, China*	7.6–21.8			2008–2012	Wu et al. 2013a
Beijing, China*	11.6			Whole month for Apr, Jul, and Oct in 2009 and Jan in 2010	Tao et al. 2014
Shanghai, China*	30.8 (27–36)			13 sets of samples at each of the 4 sites during Apr 2004 to Apr 2005	Chen et al. 2008
Shanghai, China*	7.9–26.5			2013.01	Ming et al. 2015
Guangzhou, China*	17.6			7 samples during Jul to Aug, 2010	Huang et al. 2014a; Huang et al. 2014b
Shenzhen*	28.6			Jun 2010-Dec 2010	Duan and Tan 2013
Lhasa*		1.8		Sep 2007-Aug 2008	Cong et al. 2011
Hong Kong*	2.79–5.07	3.62–7.28		Nov 2000-Feb 2001	Ho et al. 2003
Hong Kong*		6		2001–2007	Pun et al. 2014
Southern Taiwan*	3.84	5.99		Aug and Dec 2000	Ying et al. 2003
Central Taiwan*			3.15–4.13	Sep 2009 to May 2010	Fang et al. 2012
Southern Alps, New Zealand*			$5.6e^{-3}$	16 Jan and 16 Feb 2009	Marx et al. 2014
Puerto Rico*	0.09–0.39			Nov 2000 to Sep 2001	Figueroa et al. 2006
Barcelona, Spain*	0.50			27 Jan to 22 Jun 2012 and 14 Sep 2012 to 22 Feb 2013	Rivas et al. 2014
Washington, DC, USA*	~ 1.0			23 Jun to 8 Aug and 20 Oct to 4 Dec in 2003	Greene and Morris 2006
Greater Houston, the US*	1.0			Jan 2008 to Dec 2013	Liu et al. 2016
Athens, Greece*	1.42–2.39			Mar 1995 to Mar 1996	Thomaidis et al. 2003
Istanbul, Turkey*			2.23–5.55	Jul 2008 to Aug 2010	Onat and Şahin 2014
Visakhapatnam, India*		2.5–5.3		Apr 2010 to Dec 2011	Police et al. 2016
Urban areas in Europe	0.5–3			-	European Communities 2001
Bor, Serbia*			12–323	1994–2008	Serbula et al. 2010
Bor, Serbia*			6.1–126	2009-2015	Serbula et al. 2017
Poland*	nf.-10.6			2001-2012	Widziewicz et al. 2016
Lahore, Pakistan*	18	22		Jan 2007 to Jan 2008	von Schneidemesser et al. 2010

nf., not found

*Mean concentration

explained by meteorological data such as higher air pressure, lower temperature, and lower humidity observed in the winter and spring, as shown in Table 3.

The chronic adverse health effects and PM have differed among studies; a lack of spatially resolved elemental composition data may be partly responsible for these differences (Beelen et al. 2015). Therefore, studies about the temporal trends and seasonal profiles of arsenic concentrations in outdoor air PM in most regions of China are needed to investigate the association between human exposure to air PM-bound arsenic and its adverse health effects and to establish efficient preventive measures and strategies.

Arsenic exposure through personal air PM_{10} inhalation

The contribution of personal inhalation of airborne PM to arsenic exposure was not yet well characterized. Outdoor air

Table 3 The seasonal variation in arsenic (ng/m^3) observed in outdoor air $PM_{2.5}$ in Wuhan, China, during 2015–2017 and related meteorological data

Year	Season	*N*	Mean	SD	GM	Median	5th	95th	Min	Max	Air pressure (kPa)	Temperature (°C)	Humidity (%)	Precipitation (mm)	Wind speed (m/s)	Sunlight hours (h)
2015	Winter	37	13.7	7.17	11.9	14.1	7.87	16.9	3.93	37.3	102.25	5.86	80.2	1.96	1.47	2.66
	Spring	40	12.1	12.8	7.71	7.77	4.85	9.69	1.39	61.6	101.19	17.0	79.2	4.87	1.77	4.61
	Summer	52	7.45	7.73	4.70	3.89	2.89	6.63	0.76	34.4	100.26	26.8	81.3	6.48	1.79	6.10
	Autumn	27	10.6	7.26	9.13	8.68	7.66	10.4	3.49	37.4	101.57	17.7	84.9	3.23	1.44	4.39
	Total	156	10.7	9.38	7.47	7.92	7.00	9.04	0.76	61.6	101.30	16.9	81.4	4.15	1.62	4.45
2016	Winter	41	6.92	6.89	5.48	5.23	4.27	5.89	2.07	43.0	102.40	5.78	78.7	1.60	1.72	3.58
	Spring	37	9.37	6.05	7.36	7.73	5.28	11.4	1.00	24.2	101.13	17.4	79.1	3.65	1.83	4.37
	Summer	56	5.34	4.97	3.84	3.30	2.52	4.33	0.75	23.7	100.21	27.7	80.6	14.0	1.76	6.11
	Autumn	24	6.14	5.85	3.87	*2.69*	1.91	7.32	0.68	19.9	101.47	18.0	80.4	2.32	1.72	3.57
	Total	158	6.81	6.04	4.91	4.85	4.09	5.72	0.68	43.0	101.30	17.3	79.7	5.71	1.76	4.41
2017	Winter	68	11.5	10.3	8.11	7.31	5.57	8.65	0.69	41.4	102.38	6.29	77.5	1.42	1.46	3.57
	Spring	67	11.2	7.31	8.90	9.09	6.58	12.1	2.19	33.9	101.23	17.3	78.0	4.73	1.75	4.95
	Summer	84	5.48	3.90	4.47	4.00	3.50	4.91	0.42	23.3	100.24	28.2	81.8	4.15	1.66	5.91
	Autumn	46	3.84	2.58	2.96	3.24	2.15	4.70	0.51	9.70	101.58	17.4	85.5	2.33	1.47	3.76
	Total	265	8.18	7.55	5.77	5.51	4.83	6.22	0.42	41.4	101.36	17.3	80.7	3.17	1.59	4.55

GM, geometric mean

Correlation coefficients between mean arsenic concentration during different seasons and the corresponding mean values of air pressure, temperature, humidity, precipitation, wind speed, and sunlight hours were 0.44, − 0.54, − 0.45, − 0.29, − 0.20, and − 0.38, respectively. Correlations were not significant ($p > 0.05$)

PM inhalation could be used for the calculation of human exposure. However, inhalation exposure will be underestimated based on the outdoor air concentration of arsenic and the outdoor exposure factor (the percentage of outdoor activity, 0.05–0.20) (Li et al. 2018; Rodes et al. 2010; USEPA 2011). To estimate the daily intake of arsenic through the inhalation pathway more accurately, air PM_{10} samples collected by personal air samplers were used in this study.

In autumn 2016, personal air PM_{10}-bound arsenic (median 10.8 ng/m^3, 95th percentile 13.8 ng/m^3) (Table 4 and Fig. 1a) and the values estimated for $PM_{2.5}$-bound arsenic (median **6.93** ng/m^3, 95th percentile 8.84 ng/m^3) (Table S7, based on a partitioning ratio of 64% for $PM_{2.5}/PM_{10}$) (Ying et al. 2003) were both higher than those detected in outdoor $PM_{2.5}$ (median 2.69 ng/m^3, 95th percentile 7.32 ng/m^3) (Table 3); these results suggested atmospheric arsenic easily penetrated indoors spaces, with small-sized PM, or there were other indoor sources of arsenic, such as smoking and fuel combustion.

The exposure doses could be influenced by factors such as gender, residential and working area, age, and inhalation rate. To estimate the EDI of arsenic through inhalation, the population was categorized into five age groups (Table S2) (USEPA 2011). The median and 95th percentile values of arsenic concentrations were used for the calculation of the EDIs.

The EDIs of PM_{10}-bound arsenic inhalation for adults are shown in Table 5. The median arsenic exposure doses from air PM_{10} samples collected by personal air samplers in Wuhan were calculated. The gender differences in body weight were considered; the results, in decreasing order, are ranked as follows: urban female (GM, 3.56 ng/kg bw/day) > urban male (2.97) > rural female (1.77) and rural male (1.76). Additionally, the highest median EDIs of infants were twice those of teenagers (Table S8).

There were uncertainties in the estimation of daily intake. First, the mean values of AIR were extracted from the US Environmental Protection Agency (USEPA) exposure handbook (USEPA 2011), which could be not sufficiently appropriate for China. Second, the sampling sites in this study included only urban and suburban rural areas and might not be representative of residents who live in remote rural areas. Finally, personal air PM_{10} samples were collected from adults; thus, the results might not represent the exposure dose of other age groups due to their different behavior patterns.

Arsenic exposure via air particulate matter compared with total bioavailable daily intake based on urinary arsenic concentration

Generally, the elimination half-life of arsenic and its metabolites is approximately 2–4 days (Lauwerys and Hoet 2001), and urinary arsenic can reflect recent personal exposure. The total urinary arsenic concentration in this study (GM,

Table 4 Arsenic in air PM_{10} samples (ng/m^3) collected by personl air samplers from Wuhan, China, during autumn (October and November) 2016

Area	Gender	*N*	Mean	SD	GM	Median	5th	95th	Minimum	Maximum
Rural	Male	13	16.0	18.2	7.11	8.19	1.95	35.4	0.86	52.8
	Female	13	12.4	17.0	7.33	5.98	5.14	10.1	1.65	64.4
	Total	26	14.2	17.3	7.22	6.05	4.37	10.1	0.86	64.4
Urban	Male	13	16.7	10.3	14.6	13.8	10.6	17.6	7.76	44.0
	Female	15	13.7	7.17	12.4	12.0	9.83	16.6	6.68	35.5
	Total	28	15.1	8.72	13.4	12.2	10.8	15.7	6.68	44.0
Rural and urban	Male	26	16.3	14.5	10.2	11.6	8.21	16.8	0.86	52.8
	Female	28	13.1	12.5	9.71	10.4	6.57	13.1	1.65	64.4
	Total	54	14.6	13.4	9.94	10.8	8.20	13.8	0.86	64.4

GM, geometric mean

15.9 μg/L) was higher than that (GM, 1.69–4.45 μg/L) from the US NHANES during 2003–2014 (samples were limited to individuals who had urinary arsenobetaine concentrations below the MDL) (Welch et al. 2018). When urinary arsenic was stratified by considering rural-urban and gender differences, the distribution is (Fig. 1b and Table S9) as follows: rural male (median, 20.3 μg/L) > urban male (19.7) > urban female (17.6) > rural female (12.3). The results of a previous study on urinary arsenic in women who were pregnant and in their first trimester in urban Wuhan, China, (median, 18.1 μg/L)

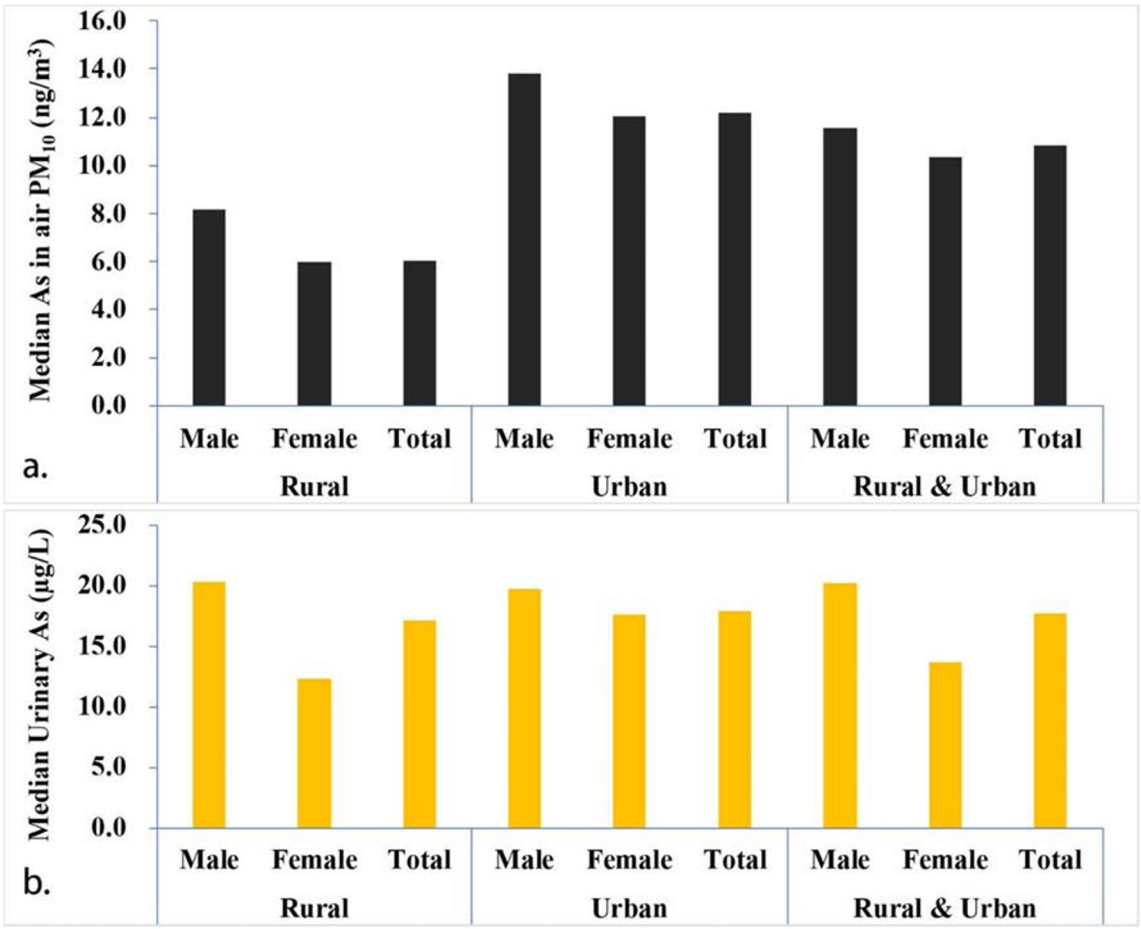

Fig. 1 Arsenic in air PM_{10} samples (ng/m^3) collected by personal air samplers (**a**) and in urine (μg/L) (**b**) for residents from rural and urban areas in Wuhan, China

Table 5 Estimated daily intakes (EDIs, ng/kg bw/day) of arsenic through inhalation of air and urinary arsenic concentrations for male and female adults in rural and urban areasof Wuhan, China, based on the median and 95th percentile concentrations

Pathway	Area	Gender	EDI, median	EDI, 95th percentile
Personal air inhalation of adults	Rural	Male	1.76	7.60
		Female	1.77	3.00
	Urban	Male	2.97	3.78
		Female	3.56	4.91
	Rural and urban	Male	2.49	3.62
		Female	3.07	3.89
Urine of adults	Rural	Male	417	546
		Female	348	557
	Urban	Male	405	696
		Female	497	775
	Rural and urban	Male	416	488
		Female	386	545

(Liu et al. 2018) were consistent with the values observed (17.6 μg/L) for urban females in the present study. The median urinary total arsenic (17.7 μg/L) in this study was close to the 95th percentile of inorganic-related arsenic in the US population (18.9 μg/L) (Caldwell et al. 2009), and the 95th percentile for the highest subgroup (urban male, 33.9 μg/L) in this study was close to the occupational biological effect index (35 μg/L) provided by the American Conference of Governmental Industrial Hygienists (ACGIH 2001).

The withdrawn provisional tolerable daily intake of inorganic arsenic had been previously recommended by WHO as 2.1 μg/kg bw/day (Halder et al. 2013), and the total daily intake based on urinary arsenic (EDI_{urine}) observed in this study (Table 5) was lower than this value, but higher than that recommended by USEPA (0.3 μg/kg bw/day) (USEPA 2018). The median values of total EDI_{urine} of arsenic, in decreasing order, were as follows: urban female (497 ng/kg bw/day) > rural male (417) > urban male (405) > rural female (348). Compared with the total EDI_{urine}, air exposure accounted for less than 1% (Table 5), which was consistent with a previous document (European Communities 2001).

However, the comparison should be interpreted with caution. When total urinary arsenic is used for the estimation of total daily intake, the daily intake could be underestimated as only approximately 66% (54–74%) of the total dose in human was excreted in urine (Buchet et al. 1981; Hays et al. 2010), although urinary excreted arsenic may represent the bioavailability of arsenic in diet to a large extent—a critical parameter to better estimate arsenic exposure (Li et al. 2017). Additionally, inorganic-related arsenic metabolites and seafood-related harmless arsenic metabolite arsenobetaine should be quantified separately in central China in future studies to better characterize the harmful part of human exposure to arsenic, since data are only available from Nanjing, China, (Wang et al. 2016; Zhang et al. 2014) and Inner Mongolia, China, (Liu et al. 2017) so far.

In most areas around the world, rice and water are reported to be the main sources of ingested arsenic, especially for inhabitants of Asia (Davis et al. 2017; Mantha et al. 2016). According to a previous report about arsenic exposure in children from a hometown with nonferrous metals, arsenic exposure via diet was the predominant exposure pathway of residents, accounted for approximately 82% of the total EDI; the second major exposure pathway was via soil and dust, accounted for approximately 12%. The PM-bound arsenic accounted for approximately 3% (Cao et al. 2016), which was higher than the ratio observed in this study. The differences might be attributed to the fact that Cao et al.'s study was conducted in an area polluted by arsenic severely.

Coal combustion and its related air PM components have been suggested to be related to lung adenocarcinoma, which is the predominant type of lung cancer in some specific areas, such as Xuanwei, Yunnan, China (Wu et al. 2016). Epidemiological evidence suggests that the arsenic concentration in air is a reliable predictor of lung/respiratory cancer risk (Lewis et al. 2015). Thus, although the exposure of arsenic via inhalable PM was minor or even negligible compared with the daily intake via oral ingestion, the problem caused by inhalation of inorganic arsenic in areas where airborne arsenic exceeds the standard, requires more attention.

Previously, studies on the health effects of PM-related air pollution in China mainly focused on PM_{10} or $PM_{2.5}$ (Fang et al. 2016). Studies are transitioning from $PM_{2.5}$ to select toxic components such as PAHs or arsenic. The associations between arsenic in air PM and lung cancer are expected to be analyzed in developed countries (Raaschou-Nielsen et al. 2016), while more attention is required on these issues in developing countries, such as China, Serbia, Pakistan, and Vietnam. In particular, the regional and gender differences in PM-bound arsenic exposure via inhalation observed in this study may provide some clues to the associations between arsenic inhalation exposure and adverse respiratory effects.

It is difficult to determine whether or not a threshold exists for the carcinogenicity of arsenic because the current available data were insufficient to justify the assumption of a threshold (European Commission 2001). Developing countries with arsenic air pollution, such as China, are appropriate sites to investigate the potential causality.

The results of this study will be helpful for relevant policy-making, and enhance public awareness of the health risks, as lung cancer appears to be a critical problem following chronic inhalation exposure to PM-bound arsenic (European Communities 2001). Although the atmospheric arsenic concentration within a year in some cities had been previously reported in China (Duan and Tan 2013), studies on the dynamic long-term (several years or longer) investigation of arsenic in $PM_{2.5}$ or PM_{10} and its health effects are needed in the future. This study implicated the importance of PM abatement technology (Campa et al. 2018; Minguillón et al. 2009) and the emergence of clean energy such as wind, water, and sunlight instead of coal combustion.

Conclusion

Tested PM-bound arsenic levels in this study were higher than those found in cities from most other countries. Clean energy instead of coal combustion is needed to lower the arsenic emissions in China. Infants are vulnerable to high levels of PM-bound arsenic. High concentrations of arsenic were observed in outdoor air $PM_{2.5}$ during 2015–2017, with a slight decreasing trend. The concentrations of arsenic in air $PM_{2.5}$ samples in winter and spring were higher than those in summer and autumn. Inhaled PM-bound arsenic in Wuhan exceeded the recommended target values for a long time; its lung accumulation and associations with diseases such as lung adenocarcinoma require more attention, although the contribution of inhaled air PM-bound arsenic to the total arsenic intake is minor.

This study described personal exposure via the inhalation of PM-bound arsenic in air and its contribution to the total daily intake based on excreted urinary arsenic. The sources of high levels of arsenic in air and its distribution throughout China, including remote rural areas and other developing countries, need further investigation.

Funding information This work was supported by the National Key Research and Development Program of China (2016YFC0206200), the Natural Science Foundation of Hubei (2016CFB274), the Special Project of Chinese Ministry of Environmental Protection (2111101-03 and 2111101-04), and the Health and Family Planning Commission of Wuhan Municipality (WG18A05).

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