Bioavailability and speciation of arsenic in urban street dusts from Baoding city, China

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ABSTRACT
Twenty-one street dust samples were collected from the urban sites in Baoding, China, to investigate the species and bioavailability of arsenic in them. The ecological risk and bioavailability of arsenic were evaluated using three models including Bioavailability Factor (BF), Contamination Factors (Cf) and Geoaccumulation Index model (Igeo). The species of arsenic in the dust samples were analyzed using an optimized BCR sequential extraction method. The total concentrations of As in the street dust samples ranged from 13.16 mg kg−1 to 67.26 mg kg−1. Geoaccumulation index (Igeo) of As ranged from 0.28 to 1.99. The speciation analysis indicated that As in the street dust samples were mainly in the residual fraction (F4), and the proportion ranged from 84.35% to 87.07%. Moreover, the ranges of the BF and Cf were 0.650–0.129 and 0.119–0.186, respectively. The results indicated that arsenic contained in the street dust samples was with low bioavailability.

Introduction
Arsenic (As) has been increasingly concerned by the researchers all over the world because of its high toxicity and wide distribution. Arsenism can lead to low bone marrow function, damage of liver, sense organ barrier and inflammation of peripheral. Previous studies have shown that As could be enriched in atmosphere particulate matters and inhalation has become an important approach to arsenic exposure [1,2]. Street dust is an important source of atmospheric particulate matters and it is well-established that street dust is an important indicator of heavy metals in urban environment [3]. Most of street dusts are fine particulate matters which are not only easily to be inhaled to the depth of respiratory system but also possess the stronger adsorbability to heavy metals and increase the health risk to human beings. Heavy metals enriched in street dusts had been well studied [4–9], but little research was done about the distribution of arsenic species in particulate matters. Arsenic in particulate matters has attracted many attentions and the reports indicated that arsenic pollution in atmosphere was serious in China [16,17]. Therefore, it is necessary to investigate the distribution of arsenic species and evaluate the bioavailability of arsenic in particulate matters in China.

In order to reflect the bioavailability and ecological risk of heavy metals more intuitively, several models such as Ecological Risk Index (RI) [18], Bioavailability Factor (BF) [19], Contamination Factors (Cf) [20] and Geoaccumulation Index model (Igeo) [21] have been proposed. Among them, the sequential extraction method is regarded as an effective approach [13,14]. Nemati et al. [15] investigated the fraction distributions of Cd, Cr, Cu, Fe, Mn, Pb, V and Zn in shrimp aquaculture sludge and different depths of sediments with sequential extraction. However, little research was done about the distribution of arsenic species in particulate matters. Arsenic in particulate matters has attracted many attentions and the reports indicated that arsenic pollution in atmosphere was serious in China [16,17]. Therefore, it is necessary to investigate the distribution of arsenic species and evaluate the bioavailability of arsenic in particulate matters in China.

Arsenic in dusts can be present in various chemical forms (named species). Different species of arsenic have different bioavailabilities and different environmental effects to human beings. In order to evaluate the environmental behaviors and health effects of arsenic in solid phase, several methods including sequential extraction, gastric (GP)/intestinal phase (IP) extraction and simulated lung fluid extraction have been proposed [11,12]. Among them, the sequential extraction method is regarded as an effective approach [13,14]. In this research, the sequential extraction, Bioavailability Factor (BF), Contamination Factors (Cf) and Geoaccumulation Index model (Igeo) were used to study and evaluate the bioavailability of arsenic in street dusts.
Baoding area located in the southeast of Beijing and Tianjin with more than 10 million of people and it is a typical medium-sized city with the rapid development in north China. In recent years, haze pollution is very serious in Hebei province, China, especially in Baoding. Coal consumption and rapid growth of vehicles in this city cause various air pollutants and the released pollutants including toxic elements can be accumulated in street dusts which are regarded as a major source of fine particles in air [23]. Considering the high toxicity of arsenic and the high concentration of arsenic in particulate matters in the north of China [16,17], to investigate arsenic in street dusts from Baoding will be very helpful for the complete understand of arsenic pollution in haze particles in the Beijing, Tianjin and Hebei region.

Materials and methods

Street dust sample collection

Twenty-one street dust samples were collected from 13 sites on the streets in Baoding city, 4 sites in the residential areas and 4 sites in the parks, in May, 2016. The details for the sampling locations were presented in Figure 1 and Table 1. The samples from North China Electric Power University (NCEPU) were sampled as the control samples because it is far from heavy traffic and industrial pollution. The street dust at each sampling site was brushed into a polyethylene bag using a plastic brush. The sampling locations on main roads were determined by grid method (2.5 × 2.5 km) and each dust sample was collected at crossroad near the center of each grid. The sampling sites were 50 cm away from the roadside and 50 m long from the sweeping section. The sampling locations in parks and residential areas were more than 300 m away from the main roads, and the samples were collected at the sites with little vehicles.

Sample pretreatment

Street dust samples were sieved through a 40 mesh (<420 μm) nylon sieve to remove debris, such as stones and leaves. Then, those samples were freeze-dried, sieved through a 200 mesh (<74 μm) sieve and stored in sealed polyethylene bags in a refrigerator.

Instruments and reagents

An atomic fluorescence spectrometer (Beijing Titan Instruments Co., Ltd, Beijing, China) was used to determine the concentrations of As in the extraction solution. The pH value of the solution was determined by a pH meter (PHS-3C, Shanghai Jingke Industrial Co., Ltd.). An ultracentrifuge (H3021D, Shanghai Zhixin Instrument Technology Co., Ltd.) and a horizontal rotator shaker (ZWY-200D, Shanghai Zhicheng Analytical Instrument Manufacturing Co., Ltd.) were employed in this experiment. All of the glass containers were soaked in 50% HNO₃ (v/v) and washed with distilled water before use. The extracts of the samples were decanted into the Polypropylene (PP) tubes after centrifugation and stored at 4 °C. All the reagents applied in this experiment were analytical grade or better.

pH measurement

The pH of the street dust samples were measured by maxing sample with deionized water at a ratio of 1:2.5 (sediment to water) (LY/T 1239-1999). The supernatant of
filtered through a 0.45 μm nylon filter and stored at 4 °C in a fridge for the final determination. The samples were treated in replicates.

Step two, reducible fraction (F2): The residue after extraction step one was subjected to this step by adding 20.0 mL of hydroxylamine hydrochloride (0.5 mol/L NH₄OH-HCl) adjusted by CH₃COOH to pH 2.0. The samples were shaken for 16 h. Then, the following treatments were same as the first step.

Step three, oxidable fraction (F3): 5.0 mL of 8.8 mol/L H₂O₂ were added in the centrifuge tube with the residue from step two and heated at 85 °C for 2 h to make it evaporate to a small volume (1.0–2.0 mL). After that, 20.0 mL of 1 mol/L CH₃COONH₄ (adjusted to pH 2 with HNO₃) was added and shaken for 16 h. Then, the following treatments were same as the first step.

Step four, residual fraction (F4): The residue from step three was transferred into a colorimetric tube (50.0 mL), and 4.0 mL of aqua regia was added for digestion with water-bath heating at 60 °C for 12 h. Then, it was cooled down to room temperature and diluted to 50.0 mL with deionized water. The following treatments were same as the first step.

Statistics

ArcGis was used to picture the sketch map of the sampling locations. The data were analyzed with Excel 2010 and Origin8.0. Statistical analysis was conducted with Excel 2010 and SPSS 17.0.

Results and discussion

pH measurement

The pH of street dust plays an important role for the species of heavy metals. The pH can affect the adsorption states of heavy metals on the surface of street dust and change the occurrence form of heavy metals [25]. The percentage of soluble arsenic was higher in alkaline soil because of the strong exchange captivity of OH-, which could increase the toxicity of arsenic [26]. Table 3 showed the pH values of the street dust samples.

It was observed that the pH values of dust from the main streets (average: 8.16) were higher than that of the samples from the parks and communities at the same area (average: 7.26). All of the samples were in alkaline

### Table 1. Sampling locations.

| Sampling location | NO. | Coordinates         |
|------------------|-----|---------------------|
| Chaoyang street × Fuxing road | A1  | 115°472’N, 38°900’E |
| Chaoyang street × Wusi road    | A2  | 115°469’N, 38°677’E |
| Chaoyang street × Tianwei road | A3  | 115°475’N, 38°666’E |
| Yangguang street × Qiyi road   | A4  | 115°468’N, 38°891’E |
| Yangguang street × Baxing road | A5  | 115°482’N, 38°677’E |
| Hengxiang street × Fuxing road | A6  | 115°500’N, 38°903’E |
| Hengxiang street × Wusi road   | A7  | 115°496’N, 38°683’E |
| Hengxiang street × Tianwei road| A8  | 115°486’N, 38°682’E |
| Lianchi street × Yiji road     | A9  | 115°517’N, 38°888’E |
| Lianchi street × Dongfeng road  | A10 | 115°503’N, 38°675’E |
| Changcheng street × Fuxing road | A11 | 115°530’N, 38°897’E |
| Changcheng street × Wusi road  | A12 | 115°513’N, 38°680’E |
| Changcheng street × Tianwei road | A13 | 115°505’N, 38°681’E |
| Jingxiu park                     | B1  | 115°467’N, 38°889’E |
| Baoding Zoo                      | B2  | 115°488’N, 38°861’E |
| Dongfeng park                    | B3  | 115°516’N, 38°871’E |
| NCEPU                           | B4  | 115°522’N, 38°895’E |
| Xiulan community                 | C1  | 115°512’N, 38°900’E |
| Zhonghua community               | C2  | 115°511’N, 38°869’E |
| Xinyiida community               | C3  | 115°479’N, 38°898’E |
| Sillying community               | C4  | 115°481’N, 38°677’E |

### Table 2. The sequential extraction procedure used in this experiment.

| Extraction step | Fraction | Extraction procedure |
|-----------------|----------|----------------------|
| 1               | Acid-soluble (F1) | 20.0 mL 0.11 M CH₃COOH, 16 h |
| 2               | Reducible (F2) | 20.0 mL 0.5 M NH₄OH-HCl, pH 2.16 h |
| 3               | Oxidable (F3) | 5.0 mL 8.8 mol/L H₂O₂, 85 °C, pH 2, 16 h |
| 4               | Residual (F4) | 3.0 mL HCl, 1.0 mL HNO₃, 60 °C, 12 h |

### Speciation analysis of arsenic using sequential extraction method

A BCR sequential extraction procedure [24] was applied to investigate the distribution of arsenic in each fraction and obtain the bioavailability of arsenic in street dust samples from Baoding urban area. The extraction reagent and procedure was demonstrated in Table 2.

Step one, acid-soluble fraction (F1): 0.5 g of each street dust sample and 20.0 mL of acetic acid (CH₃COOH 0.11 mol/L) were added into a centrifuge tube (50 mL). The tubes were shaken at 500 rpm at room temperature for 16 h. Then, the extraction was centrifuged at 4000 rpm for 15 min, and the extraction was removed in a centrifuge tube. Finally, 10.0 mL of DI water was mixed with the residue to shake for another 10 min, and then it was re-centrifuged. The supernatant was poured into the former extract from the same step. The extract was
or weak alkaline. Our results were in agreement with the previous report suggesting that the pH of street dust was higher than soil at the same area due to the alkaline substances in aging road and building materials [26].

**Table 3.** pH of the street dust in Baoding urban area.

| Sampling location          | pH       | Sampling location          | pH       |
|----------------------------|----------|----------------------------|----------|
| Dongfeng park (DF)         | 6.86 ± 0.67| Changcheng Streets (CC)    | 8.76 ± 0.95 |
| Zhonghua community (ZH)    | 7.43 ± 0.53| Fuxing Load (FY)           | 8.68 ± 0.96 |
| NCEPU(HD)                  | 7.82 ± 0.22| Chaoyang Street (CY)       | 7.97 ± 0.67 |
| Xiulan community (XL)      | 6.47 ± 0.34| Tianwei Load (TW)          | 8.17 ± 0.66 |
| Jingxiu park (JX)          | 6.83 ± 0.15| Yangguang Street (YG)      | 7.82 ± 0.43 |
| Sillying community (SLY)   | 7.93 ± 0.48| Hengxiang Street (HX)      | 8.17 ± 0.67 |
| Baoding zoo (BZ)           | 7.44 ± 0.37| Lianchi Street (LC)        | 8.08 ± 0.02 |
| Xinyidai community (XYD)   | 6.94 ± 0.28| Wusi Load (WS)             | 7.60 ± 0.30 |
| Average                    | 7.26     | Average                    | 8.16     |

**Figure 2.** Contents of As in the street dust samples from different sites, (a), park and community samples, (b), street samples.

The total concentrations of As in street dust samples were higher than the environmental background values (9.20 mg kg⁻¹) [27] (Figure 2) and the concentration...
range was between 13.16 and 67.26 mg kg\(^{-1}\). The total As in the street dust samples from the main streets (average 27.37 mg kg\(^{-1}\)) was lower than that from parks and communities (average: 47.50 mg kg\(^{-1}\)). The concentration of arsenic in the LC sample (35.41 mg kg\(^{-1}\)) was the highest compared with the others. This phenomenon probably caused by the heaviest traffic on this street. The contents of total As in the samples from the parks were similar with the samples from the communities (all above 40.0 mg kg\(^{-1}\)). The highest value (54.97 mg kg\(^{-1}\)) was found at the ZH site.

According to the Environmental quality standard for soils (GB 15618-1995/2008), the secondary standard for As in residential area and commercial district was 50.0 mg kg\(^{-1}\), the secondary standard for Community were higher than the secondary standard. In the samples from NCEPU, Jingxiu Park and Zhonghua communities (average: 47.50 mg kg\(^{-1}\)) was the highest compared with the others. This phenomenon probably caused by the heaviest traffic on this street. The contents of total As in the samples from the parks were similar with the samples from the communities (all above 40.0 mg kg\(^{-1}\)). The highest value (54.97 mg kg\(^{-1}\)) was found at the ZH site.

**Speciation analysis of arsenic**

The street dust samples from NCEPU, Xiulan community and Dongfeng park were chosen to investigate the distribution of heavy metals in each fraction using BCR sequential extraction procedure. A soil sample from NCEPU was also treated with the same procedure as a control sample. The results were showed in Table 4.

| Speciation | NCEPU soil | NCEPU | Xiulan community | Dongfeng park |
|------------|------------|-------|------------------|--------------|
| F1         | ND*        | 1.34 ± 0.06 | 1.25 ± 0.15 | 2.21 ± 0.18 |
| F2         | 0.47 ± 0.13 | 0.95 ± 0.09 | 1.08 ± 0.26 | 1.32 ± 0.06 |
| F3         | 0.81 ± 0.23 | 1.59 ± 0.09 | 0.42 ± 0.27 | 2.15 ± 0.41 |
| F4         | 38.16 ± 6.37 | 25.98 ± 1.15 | 23.06 ± 0.39 | 30.58 ± 4.05 |
| SUM        | 39.44 ± 6.73 | 29.86 ± 1.39 | 25.81 ± 1.07 | 36.26 ± 4.70 |

*Not detectable.

was from 84.35% to 87.07%. However, acid-soluble (F1), reducible (F2) and oxidable (F3) accounted for small part of the total amount with the percentage range of 4.48% ~6.09%, 3.19%~4.09% and 5.33%~5.93%, respectively. In addition, the distribution of As in various fractions in each sample was showed in Figure 3.

**Ecological risk assessment**

**Geoaccumulation Index model** \((I_{geo})\) was applied to evaluate the ecological risk of street dust in this study. It was proposed by Muller firstly and the factor was calculated using following Equation (1) [21]:

\[
I_{geo} = \log_{2} \left[ \frac{C_n}{k \times B_{E_n}} \right]
\]

where \(C_n\) is the measured concentration of the elemental in samples(mg kg\(^{-1}\)), \(B_{E_n}\) is environmental background value(mg kg\(^{-1}\)), \(k\) is correction factor (generally 1.5). The environmental background value of As was 9.2 mg kg\(^{-1}\) [27]. The \(I_{geo}\) values were calculated using Equation (1) to assess the pollution level at each sampling location according to the grades of geoaccumulation index (Table 5).

It was found that the geoaccumulation index \((I_{geo})\) ranged from 0.28 to 1.99 (Table 6). As contamination in Zhonghua community was most serious, for which the \(I_{geo}\) value was 1.99. The \(I_{geo}\) level was regarded as grade III, which meant moderate contamination. Furthermore, the \(I_{geo}\) values of the samples from the main streets were calculated by the average of As contents for each sampling location in the street, and the results were 1.31 (Chaoyang street), 0.84 (Yangguang street), 0.56 (Hengxiang street), 1.18 (Lianchi street), 0.57 (Changcheng street), 0.83 (Fuxing road), 0.91 (Wusi road), 0.82 (Qiyi road) and 0.55 (Tianwei road). All of the \(I_{geo}\) levels of sampling location were in gradell, which meant that the sampling sites were uncontaminated. The \(I_{geo}\) value of the samples from Chaoyang street and Lianchi street were in grade III, which meant moderate contamination.

**Bioavailability assessment**

There are several models to evaluate the bioavailability of heavy metals in soil or dust. In this study, Bioavailability Factor (BF) and Contamination Factors \((C_f)\) were applied to assess the bioavailability of As in the samples. The index values were calculated by the following Equation (2) and (3) [31].
where \( C_{\text{bio}} \) is the concentration of heavy metal in the bioavailable fractions (mg kg\(^{-1}\)), \( C_{\text{total}} \) is the total concentration of heavy metal (mg kg\(^{-1}\)). In our study, F1, F2 and F3 were regarded as the bioavailable fractions.

\[
BF = \frac{C_{\text{bio}}}{C_{\text{total}}}
\]

(2)

where \( F_1, F_2, F_3 \) and \( F_4 \) refer to the concentration of As in \( F_1, F_2, F_3 \) and Residue from the sequential extraction procedure, respectively (mg kg\(^{-1}\)). The BF and \( C_f \) of samples were calculated and presented in Table 7.

It was found that the BF and \( C_f \) of the samples collected in Dongfeng Park were higher than the others. In contrast, BF and \( C_f \) in the samples from Xiulan community were the lowest. In the other words, the dust from Dongfeng Park was with higher bioavailability. The bioavailability of As in the dust from Xiulan community was relatively lower. The BF and \( C_f \) for all of the street dust samples are about 3~5 times higher than the soil from NCEPU.

The relationship between pH and species distribution

The correlation between pH and species of As in the street dust samples was analyzed by SPSS17.0, and presented in Table 8.

It was observed that there was no significant correlation ship between pH and the species. This result indicated that pH could not affect the species of As in dust. The distribution of As in various fractions were probably affected by other factors [32,33].

Figure 3. Fraction distributions of As in the street dust samples.

Table 5. Grades of geoaccumulation index.

| I_{geo} | Grades | Pollution situation |
|---------|--------|---------------------|
| 0       | I      | Uncontaminated       |
| 0−1     | II     | Uncontaminated to moderately contaminated |
| 1−2     | III    | Moderately contaminated |
| 2−3     | IV     | Moderately to heavily contaminated |
| 3−4     | V      | Heavily contaminated |
| 4−5     | VI     | Heavily extremely contaminated |
| 5−10    | VII    | Extremely contaminated |

Table 6. Geoaccumulation index and the classification of arsenic in the street dust.

| Sampling location       | \( I_{geo} \) | Grades |
|-------------------------|--------------|--------|
| Chaoyang street × Fuxing road(CF) | 1.54         | III    |
| Chaoyang street × Wusi road(CW) | 1.74         | III    |
| Chaoyang street × Tianwei road(CT) | 0.84         | II     |
| Yangguang street × Qiyi road(YQ) | 0.85         | II     |
| Yangguang street × Baihua road(YB) | 1.72       | III    |
| Hengxiang street × Fuxing road(HF) | 0.63         | II     |
| Hengxiang street × Wusi road(HW) | 1.55         | III    |
| Hengxiang street × Tianwei road(HT) | 0.18         | II     |
| Lianchi street × Qiyi road(LQ) | 0.79         | II     |
| Lianchi street × Dongfeng road(LD) | 1.76        | III    |
| Changcheng street × Fuxing road(CF) | 0.60         | II     |
| Changcheng street × Wusi road(CW) | 0.28         | II     |
| Changcheng street × Tianwei road(CT) | 1.11        | III    |
| Jinxing park            | 1.67         | III    |
| Baoding Zoo             | 1.95         | III    |
| Dongfeng park           | 1.90         | III    |
| NCEPU                   | 1.62         | III    |
| Xiulan community        | 1.85         | III    |
| Zhonghua community      | 1.66         | III    |
| Xinyida A community     | 1.85         | III    |
| Siliying community      | 1.99         | III    |

Table 7. Bioavailability Factor (BF) and Contamination Factors (\( C_f \)) of each sample.

| Sample | NCEPU soil BF | NCEPU Cf | Xiulan community BF | NCEPU CF | Dongfeng park BF | NCEPU CF |
|--------|---------------|----------|----------------------|----------|------------------|----------|
| BF     | 0.026          | 0.075    | 0.065                | 0.129    | 0.034            | 0.149    | 0.119          | 0.186    |
indicated that the arsenic in street dust was with low bioavailability and it was hard to transport and transform during the sequential extraction steps.

**Mass balance**

In order to ensure the reliability of the optimized BCR sequential extraction in this research, the standard reference material (GBW-07405) was sequentially extracted, and three replicates were simultaneously conducted. The recovery of As was calculated according to the following Equation (4) [34].

\[
\text{Recovery} (%) = \left( \frac{(F_1 + F_2 + F_3 + F_4)}{C_{total}} \right) \times 100\%
\]

where \(F_1, F_2, F_3\) and \(F_4\) refer to the concentration of As in F1, F2, F3 and Residue fraction from the sequential extraction procedure, respectively (mg kg\(^{-1}\)). \(C_{total}\) is the total concentration of heavy metal (mg kg\(^{-1}\)). The recovery for As in the sequential extraction ranged from 89.75% to 103.58%, which indicated that there was no obvious loss of arsenic during the sequential extraction steps.

**Conclusions**

The bioavailability and speciation of arsenic in urban street dust from Baoding were evaluated using the sequential extraction method. Speciation analysis demonstrated that most of arsenic was present in the residual fraction. Furthermore, the BF and \(C_r\) of street dust samples were calculated and the range of them were 0.650–0.129 and 0.119–0.186, respectively, which indicated that the arsenic in street dust was with low bioavailability and it was hard to transport and transform in the ecosystem.

**Disclosure statement**

No potential conflict of interest was reported by the authors.

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**Table 8. Pearson correlation in pH and species distribution of street dust**.

| pH | F1 (%) | F2 (%) | F3 (%) | Sum (F1 + F2 + F3) (%) | Total |
|----|--------|--------|--------|------------------------|-------|
| pH 1 | −0.130 | −0.313 | −0.203 | −0.086 | 0.896 |

*Significance, p<0.1 level (bilateral)
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