Bioaccessibility and human health risk assessment of lead in soil from Daye City

Q Li¹, F Li²*, M S Xiao², Y Cai², L Xiong³, J B Huang², J T Fu²

¹Central and Southern China Municipal Engineering Design & Research Institute Co., Ltd., Wuhan 430010, China
²Research Center for Environment and Health, Zhongnan University of Economics and Law, Wuhan 430073, China
³People's Bank of China Huangshi City Center Branch, Huangshi 435000, China

*Corresponding author e-mail: lifei@zuel.edu.cn

Abstract. Lead (Pb) in soil from 4 sampling sites of Daye City was studied. Bioaccessibilities of Pb in soil were determined by the method of simplified bioaccessible extraction test (SBET). Since traditional health risk assessment was built on the basis of metal total content, the risk may be overestimated. Modified human health risk assessment model considering bioaccessibility was built in this study. Health risk of adults and children exposure to Pb based on total contents and bioaccessible contents were evaluated. The results showed that bioaccessible content of Pb in soil was much lower than its total content, and the average bioaccessible factor (BF) was only 25.37%. The hazard indexes (HIs) for adults and children calculated by two methods were all lower than 1. It indicated that there were no non-carcinogenic risks of Pb for human in Daye. By comparing with the results, the average bioaccessible HIs for adults and children were lower than the total one, which was due to the lower hazard quotient (HQ). Proportions of non-carcinogenic risk exposure to Pb via different pathways have also changed. Particularly, the most main risk exposure pathway for adults turned from the oral ingestion to the inhalation.

1. Introduction
Due to the strong influence of anthropic activities related to industry, urban soil widely contaminated by various heavy metals [1-2]. Heavy metals in soil do harm to human health via the oral ingestion, inhalation and dermal contact [3-5]. Lead (Pb) is a kind of poisonous heavy metal. Once Pb enters the human body, a part of Pb will be excreted by the body metabolism, and most part will be accumulated in the body [6]. It has great adverse for human body, especially for children [7]. It is of great sense to accurately qualified human health risk of human exposure to Pb.

Traditional health risk assessment method is established in the basis of total content [8-9]. However, the actual risk is overestimated, because traditional method ignores the fact that bioaccessible part of total contents would adversely impact human health [10-11]. Therefore, it is necessary to take into account bioaccessibility of heavy metals when evaluate human health risks. Simplified bioaccessible extraction test (SBET) is a widely used simulate test in vitro to simulate the release of Pb in soil in gastric juice, which can analyze the bioaccessibility of Pb [12].
Soil from part areas of Daye City was selected as the study object. The aims of this study were: (1) to measure the bioaccessibility of Pb in soil by method of SBET, (2) establish health risk assessment model considering bioaccessibility, and (3) to compare health risks based on total contents and bioaccessible contents.

2. Methods and materials

2.1. Study area
Daye City is located on the South Bank of the middle reaches of the Yangtze River and southeast of Hubei province. It is a famous hometown of bronze culture, with an area from 29°40′N to 30°15′N, 114°31′E to 115°20′E. Daye City is rich in resources, and there are 273 mineral deposit found in the city. There are about 17 kinds of metal minerals including Cu, Fe and Au.

2.2. Soil samples collection
Sediment samples were collected from four sampling sites (S1: Yangqiaocun, S2: Xiaganwan, S3: Shangrila community and S4: Yingcai road) in Daye City during Aug. 2015, referring to the NY/T1121.1-2006. The soil samples were collected in politertrafluoroethylene (PTFE) bags and then transferred rapidly to the laboratory in Wuhan. In the laboratory, surface sediments were air dried for 10 days in a cool ventilated place. Then, sediments were crushed into small pieces by using pestles and mortals. Next, sediment samples were sifted in 10 mesh nylon sieves to remove stones and plant residue. Finally, all the sediment samples were sifted in 100 mesh sieves and were kept in the plastic bottles prior to analyses.

2.3. Analytical methods

2.3.1. SBET method. SBET method was performed as follows: 50 mL of extraction fluid was added to 0.5 g of dry soil. The extraction fluid was 0.4 M glycine adjusted to pH 1.5 with concentrated HCl. This mixture was rotated end-over-end at 37 °C at 30 rpm for 1 h. Samples were centrifuged by a high speed centrifuge for 10 min and then filtered through a 0.45-μm cellulose acetate disk filter. The samples were stored in a refrigerator at 4 °C until analysis. Concentrations of Pb in the filtrate are determined by FAAS (AAS, ZEEnit700, Germany).

The pH and total concentration of Pb in soil from each sampling sites were analyzed in the previous study [13].

2.3.2. Formula for bioaccessible factor of heavy metal.

\[
BF = \frac{C_{\text{bioaccessible}}}{C_{\text{total}}} \times 100\%
\]  

(1)

Where BF is bioaccessible factor of heavy metal, unitless; \( C_{\text{bioaccessible}} \) is bioaccessible concentration of heavy metal in soil, mg·kg\(^{-1}\); \( C_{\text{total}} \) is total concentration of heavy metal in soil, mg·kg\(^{-1}\).

2.3.3. Quality control. To ensure reliability and accuracy of the analysis results, the quality assurance and quality control were assessed strictly by using blank samples, parallel samples and standard reference materials (GB07423). The analysis results were reliable when repeat sample analysis error was below 5%, and the analytical precision for replicate samples was within ± 10%. Accepted recoveries of standard samples ranged from 90% to 110%.

2.4. Human health risk assessment based on bioaccessibility

2.4.1. Exposure assessment. In this study, the exposure routes of human exposure to Pb in soil of Daye were ingestion, dermal contact and inhalation. Considering bioaccessibility of Pb, the human
exposure assessment (soil ingestion) was adjusted when calculating the \( \text{ADD}_{\text{Ing}} \). Exposure doses of human exposure to pollutants through these three exposure routes can be calculated as follows [14-15]:

1. Ingestion

\[
\text{ADD}_{\text{Ing}} = BF \times \frac{CS \times IR \times CF \times FI \times EF \times ED}{BW \times AT}
\]

Where, \( \text{ADD}_{\text{Ing}} \) is average daily doses in ingestion exposure pathway, \( \text{mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1} \); \( BF \) is bioaccessible factor, unitless, it is assumed for 1 when doing exposure assessment based on total content; \( CS \) is chemical concentration in soil, \( \text{mg} \cdot \text{kg}^{-1} \); \( IR \) is ingestion rate, \( \text{mg} \cdot \text{d}^{-1} \); \( CF \) is conversion factor, \( 10^{-6} \text{ kg} \cdot \text{mg}^{-1} \); \( EF \) is exposure frequency, \( \text{d} \cdot \text{a}^{-1} \); \( ED \) is exposure duration, \( \text{a} \); \( BW \) is body weight, \( \text{kg} \); \( AT \) is averaging time, \( \text{d} \).

2. Dermal contact

\[
\text{ADD}_{\text{Der}} = \frac{CS \times AF \times CF \times SA \times ABS \times EF \times ED}{BW \times AT}
\]

Where, \( \text{ADD}_{\text{Der}} \) is average daily doses in skin contact exposure pathway, \( \text{mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1} \); \( CS \) is chemical concentration in soil, \( \text{mg} \cdot \text{kg}^{-1} \); \( AF \) is soil to skin adherence factor, \( \text{mg} \cdot \text{cm}^{-2} \); \( CF \) is conversion factor, \( 10^{-6} \text{ kg} \cdot \text{mg}^{-1} \); \( SA \) is the skin area available for contact, \( \text{cm}^2 \); \( ABS \) is absorption factor, unitless; \( EF \) is exposure frequency, \( \text{d} \cdot \text{year}^{-1} \); \( ED \) is exposure duration, \( \text{a} \); \( BW \) is body weight, \( \text{kg} \); \( AT \) is averaging time, \( \text{d} \).

3. Inhalation

\[
\text{ADD}_{\text{Inh}} = \frac{CS \times PM10 \times DAIR \times PIAF \times FSPO \times CF \times EF \times ED}{BW \times AT}
\]

Where, \( \text{ADD}_{\text{Inh}} \) is average daily doses in inhalation exposure pathway, \( \text{mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1} \); \( CS \) is chemical concentration in soil, \( \text{mg} \cdot \text{kg}^{-1} \); \( PM10 \) is the concentration of respirable particulate pollution, \( \text{mg} \cdot \text{m}^{-3} \); \( DAIR \) is daily air inhalation rate, \( \text{m}^3 \cdot \text{d}^{-1} \); \( PIAF \) is retention fraction of inhaled particulates in body, unitless; \( FSPO \) is fraction of soil-borne particulates in air, unitless; \( CF \) is conversion factor \( 10^{-6} \text{ kg} \cdot \text{mg}^{-1} \); \( EF \) is exposure frequency, \( \text{d} \cdot \text{year}^{-1} \); \( ED \) is exposure duration, \( \text{a} \); \( BW \) is body weight, \( \text{kg} \); \( AT \) is averaging time, \( \text{d} \).

2.4.2. Non-carcinogenic risk assessment.

\[
HI = HQ_{\text{Ing}} + HQ_{\text{Der}} + HQ_{\text{Inh}} = \frac{\text{ADD}_{\text{Ing}}}{RfD_{\text{Ing}}} + \frac{\text{ADD}_{\text{Der}}}{RfD_{\text{Der}}} + \frac{\text{ADD}_{\text{Inh}}}{RfD_{\text{Inh}}}
\]

Where, \( HI \) is hazard index of non-carcinogens, unitless; \( HQ_{\text{Ing}}, HQ_{\text{Der}} \) and \( HQ_{\text{Inh}} \) are hazard quotients of ingestion, skin attach and inhalation, unitless; \( ADD \) is average daily dose, \( \text{mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1} \); and \( RfD \) is reference dose, \( \text{mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1} \). \( HQ \) is hazard quotient of non-carcinogens, according to the recommended value by USEPA, \( HQ \leq 1 \) indicates no risk, \( HQ > 1 \) indicates that risks do exist [16].

3. Discussion

3.1. Bioaccessibility of heavy metal in soil

Total contents, bio-accessible contents and corresponding bio-accessible factors of Pb in soil from Daye City were listed in the Table 1. The pH and total contents were determined in our previous study [13]. The pH in soil from Daye city were ranged from 7.24 to 8.18, it was shown that soil from Daye City were neutral and low alkaline soil. The ranges of total contents of Pb varied widely, which were from 41.93 to 104.47 mg·kg⁻¹. And the average content of Pb was 63.66 mg·kg⁻¹. The concentrations of Pb in all sampling sites were higher than background values of Hubei (26.7 mg·kg⁻¹). The bio-
accessible contents of Pb were much lower than total contents. Bioaccessible factors (BFs) were range from 17.41% to 30.83%, and average BF was 25.37%.

Table 1. Contents and bioaccessibilites of Pb in soil from Daye City

| Sampling sites | pH  | Total content (mg·kg⁻¹) | Bioaccessible content (mg·kg⁻¹) | Bioaccessible factor |
|---------------|-----|------------------------|-------------------------------|---------------------|
| S1            | 7.45| 48.21                  | 11.34                         | 23.52%              |
| S2            | 7.24| 104.47                 | 27.45                         | 26.28%              |
| S3            | 7.99| 41.93                  | 7.30                          | 17.41%              |
| S4            | 8.18| 60.01                  | 18.50                         | 30.83%              |
| Mean          | 7.72| 63.66                  | 16.15                         | 25.37%              |

3.2. Human health risk assessment

Human health risks exposure to Pb based on total content were calculated in previous study, which were shown as in Table 2. Because heavy metals could not be absorbed completely in human digestive system, the adverse effects of heavy metals on human would be overestimated, which would lead to an overestimate of human health risks. Therefore, bioaccessibility of Pb was taken into account when evaluated human health risks exposure to Pb in this study. The values of exposure factors were the same as factors listed in previous study [6], and bioaccessible factors were listed in Table 1. According to the arithmetic calculation from (2) to (5), the results of human health risks of Pb based on bioaccessibility were listed in Table 4.

For human health risk assessment based on total content, the hazard quotients based on total content (HQs) of human exposure to Pb under three routes were all lower than 1, and the HIs of Pb were also lower than 1. For adults, the average HQs were decreased in the order of HQ_{\text{ing}} (76.79%) > HQ_{\text{der}} (49.56%) > HQ_{\text{inh}} (21.17%). For children, the average HQs were decreased in the order of HQ_{\text{ing}} (97.76%) > HQ_{\text{der}} (1.82%) > HQ_{\text{inh}} (0.42%).

For human health risk assessment based on bioaccessible content, the hazard quotients based on bioaccessible content (HQts) of human exposure to Pb under three routes were all lower than 1, and the HIs of Pb were also lower than 1. For adults, the average HQts were decreased in the order of HQ_{\text{der}} (45.66%) > HQ_{\text{ing}} (4.78%) > HQ_{\text{inh}} (49.56%). For children, the average HQts were decreased in the order of HQ_{\text{der}} (91.73%) > HQ_{\text{ing}} (6.74%) > HQ_{\text{inh}} (15.4%).

By comparison of average non-carcinogenic hazards, the bioaccessible hazard index (HI) for adults was lower than the total one, which was due to the lower hazard quotient (HQ). It was the same for children. Proportions of non-carcinogenic risk exposure to Pb via different pathways have also changed. For both adults and children, proportions of the bioaccessible non-carcinogenic risk via oral ingestion were decreased. The most main risk exposure pathway for adults turned from the oral ingestion (76.79%) to the inhalation (49.56%).

Table 2. Human health risk assessment of Pb based on total content [13]

| Health risk | Age      | S1       | S2       | S3       | S4       | Mean     |
|-------------|----------|----------|----------|----------|----------|----------|
| HQ_{\text{ing}} | adults   | 2.29E-02 | 4.97E-02 | 2.00E-02 | 2.86E-02 | 3.03E-02 |
|             | children | 1.72E-01 | 3.74E-01 | 1.50E-01 | 2.14E-01 | 2.28E-01 |
| HQ_{\text{der}} | adults   | 6.11E-04 | 1.32E-03 | 5.31E-04 | 7.60E-04 | 8.06E-04 |
|             | children | 3.21E-03 | 6.96E-03 | 2.80E-03 | 4.00E-03 | 4.24E-03 |
| HQ_{\text{inh}} | adults   | 6.33E-03 | 1.37E-02 | 5.51E-03 | 7.88E-03 | 8.36E-03 |
|             | children | 7.32E-04 | 1.59E-03 | 6.37E-04 | 9.11E-04 | 9.68E-04 |
| HI          | adults   | 2.98E-02 | 6.47E-02 | 2.60E-02 | 3.72E-02 | 3.95E-02 |
|             | children | 1.76E-01 | 3.83E-01 | 1.53E-01 | 2.19E-01 | 2.33E-01 |
based on total content and bioaccessible content were
of Wuhan Studies
- J 2016
posure pathway for adults turned from the oral
e
IOP Conf. Series: Earth and Environmental Science 108 (2018) 042116
doi:10.1088/1755-1315/108/4/042116

References
University of Economics and Law (2017Y1406).
(IWHS20172005) and the Innovative Education Program for Graduate Students of Zhongnan
Provincial Education Department (B2017601), the Open Fund from Institu-
This study was financially supported by the Science and Technology Research Project of Hubei

4. Conclusion
The total content of Pb in soil from part areas in Daye City was 63.66 mg·kg⁻¹. Bioaccessible content
of Pb in soil was much lower than the total one, and the average BF was only 25.37%. The
bioaccessibilities in 4 sampling sites were variable, which ranged from 17.41% to 30.83%. Human
health risk of adults and children exposure to Pb based on total content and bioaccessible content were
evaluated. HIs of adults and children calculated by two methods were all lower than 1. It indicated that
there were no no-carcinogenic risks of Pb for adults and children in Daye. By comparing with the
results, the average bioaccessible HIs for adults and children were lower than the total one, which was
due to the lower HQ. Proportions of non-carcinogenic risk exposure to Pb via different pathways have
also changed. Particularly, the most main risk exposure pathway for adults turned from the oral
ingestion to the inhalation.

Acknowledgements
This study was financially supported by the Science and Technology Research Project of Hubei
Provincial Education Department (B2017601), the Open Fund from Institute of Wuhan Studies
(IWHS20172005) and the Innovative Education Program for Graduate Students of Zhongnan
University of Economics and Law (2017Y1406).

Table 3. Human health risk assessment of Pb based on bioaccessibility

| Health risk | Age       | S1    | S2    | S3    | S4    | Mean   |
|-------------|-----------|-------|-------|-------|-------|--------|
| HQ          | adults    | 5.40E-03 | 1.31E-02 | 3.48E-03 | 8.81E-03 | 7.70E-03 |
|             | children  | 4.05E-02 | 9.84E-02 | 2.61E-02 | 6.61E-02 | 5.78E-02 |
| HQDer       | adults    | 6.11E-04 | 1.32E-03 | 5.31E-04 | 7.60E-04 | 8.06E-04 |
|             | children  | 3.21E-03 | 6.96E-03 | 2.80E-03 | 4.00E-03 | 4.24E-03 |
| HQinh       | adults    | 6.33E-03 | 1.37E-02 | 5.51E-03 | 7.88E-03 | 8.36E-03 |
|             | children  | 7.32E-04 | 1.59E-03 | 6.37E-04 | 9.11E-04 | 9.68E-04 |
| HI          | adults    | 1.23E-02 | 2.81E-02 | 9.52E-03 | 1.75E-02 | 1.69E-02 |
|             | children  | 4.44E-02 | 1.07E-01 | 2.95E-02 | 7.10E-02 | 6.30E-02 |

4. Conclusion

The total content of Pb in soil from part areas in Daye City was 63.66 mg·kg⁻¹. Bioaccessible content of Pb in soil was much lower than the total one, and the average BF was only 25.37%. The bioaccessibilities in 4 sampling sites were variable, which ranged from 17.41% to 30.83%. Human health risk of adults and children exposure to Pb based on total content and bioaccessible content were evaluated. HIs of adults and children calculated by two methods were all lower than 1. It indicated that there were no no-carcinogenic risks of Pb for adults and children in Daye. By comparing with the results, the average bioaccessible HIs for adults and children were lower than the total one, which was due to the lower HQ. Proportions of non-carcinogenic risk exposure to Pb via different pathways have also changed. Particularly, the most main risk exposure pathway for adults turned from the oral ingestion to the inhalation.

Acknowledgements
This study was financially supported by the Science and Technology Research Project of Hubei Provincial Education Department (B2017601), the Open Fund from Institute of Wuhan Studies (IWHS20172005) and the Innovative Education Program for Graduate Students of Zhongnan University of Economics and Law (2017Y1406).

References
[1] Li F, Zhang J D, Huang J H, Huang D W, Yang J, Song Y W, Zeng G M 2016 Environ. Sci. Pollut. R. 23 13100-13.
[2] Zahida K and Bilal A Q 2014 Hum. Ecol Risk. Assess. 20(3) 658-667.
[3] Li F, Zhang J D., Jiang W, Liu CY, Zhang, Z M, Zhang C D and Zeng G M 2016 Environ. Geochem. Health. 2 1-12.
[4] Tang J, Chen C Y, HaiYi, L I, Zhang, T Q, and Xiao R 2011 Sci. Geo. Sin. 30 117-122.
[5] Li F, Huang J H, Zeng G M, Liu W C, Huang X L, Huang B, Gu Y L, Shi L X, He X X and He Y 2015 Environ. Sci. Pollut. R. 22 12261-12275.
[6] Nadeem U, Fazal H, Farman U, Ayaz A, Nasir A and Amin U J 2017 Int. J. Agron. Agri. R. 10 24-32.
[7] Zheng L B, Chen W P, Jiao W T, Huang J L, and Wei F X 2013 Environ. Sci. 34(9) 3669.
[8] Li F, Qiu Z Z, Zhang J D, Liu C Y, Cai Y and Xiao M S 2017 Inter. J. Env. Res. Pub. Heal. 14 1011.
[9] Huang J H, Liu W C, Zeng G M, Li F, Huang X L, Gu X L, Shi L X, Shi Y H and Wan J 2016 Ecotoxicol. Environ. Saf. 129 199–209.
[10] Luo X S, Ding J, Xu B, Wang Y J, Li H B, and Yu S 2012 Sci. Total. Environ. 424 88.
[11] Jiang L, Peng C, Zhong M S, Yao Y J, Xia T X, Jia X Y and Han D 2014 Res. Environ. Sci. 27 406-408.
[12] Standard operating procedure for an in vitro bioaccessibility assay for lead in soil, United States Environmental Protection Agency, 2008.
[13] Xiao M S, Li F, Zhang J D, Lin S Y, Zhuang Z Y and Wu Z X 2017 IOP Conf. Series: Earth and Environ. Sci. 64 012066.

[14] Ruby M V, Schoof R, Brattin W, Goldade M, Post G and Harnois M 2016 Environ. Sci. Tec. 33 3697-3705.

[15] Li F, Xiao M, Zhang J, Yang J, and Zhu, L 2017 AMME. 1820 040011.

[16] Risk Assessment Guidance for Superfund (RAGS): Human health evaluation manual (Part A), Office of Emergency and Remedial Response, 1989.