Journal of Hazardous Materials 297 (2015) 191–197

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

In vitro bioaccessibility of lead in surface dust and implications for human exposure: A comparative study between industrial area and urban district

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HIGHLIGHTS

- *In vitro* Pb bioaccessibility was assessed by PBET assay.
- Calcium may play an important role in reducing intestinal Pb bioaccessibility.
- Dust is an important contributor to the Pb exposure of children.
- Health risk of Pb in dust for children was serious.

ARTICLE INFO

Article history:
Received 6 December 2014
Received in revised form 20 April 2015
Accepted 25 April 2015
Available online 28 April 2015

Keywords:
Lead
Dust
Bioaccessibility
In vitro PBET

ABSTRACT

In this study, ground surface dust samples from two contrasting areas, a former zinc smelting area in Guizhou Province and a common urban district in Wuhan city, Hubei Province, China, were assessed for *in vitro* Pb bioaccessibility using a physiologically based extraction test (PBET). Extremely elevated concentrations of Pb (220–6348 mg/kg) and other trace metals were observed in the zinc smelting area. While moderate high metal concentrations (79–1544 mg/kg of Pb) in the urban dusts were attributed to various urban activities, coal combustion and traffic emissions. Lead bioaccessibility in the stomach-phase varied from 17.6 to 76.1% and no significant difference was found between industrial and urban dust samples. Compared with the stomach-phase, Pb bioaccessibility in the more alkaline intestinal-phase was considerably lower (1.2–21.8%). A significantly negative correlation was found between dust Ca concentrations and Pb bioaccessibility in the intestinal-phase, suggesting that Ca plays an important role in reducing the bioaccessible Pb in the intestinal-phase. The estimated Pb exposure based on gastric bioaccessible Pb was 13.9 and 1.8 μg/kg day for children living in the industrial and urban areas, respectively, accounting for 85% and 41% of their corresponding total Pb exposure.

1. Introduction

Lead (Pb) is a ubiquitous non-essential element in the environment and has numerous acute and chronic adverse effects on humans, especially children, by influencing their nervous system and reducing their intelligence [1]. Without strict pollution control measures, anthropogenic generated Pb contaminants can be released and transferred to various environmental settings, such as water, soil/dust and plant, resulting in significant degradation of the ecosystems [2–8].

Dust is a representative environmental media that can serve as a sink for contaminants that are deposited from the atmosphere. Meanwhile, dust is also a secondary source of contaminants in surface environments. Contaminants containing dust particles can be re-suspended into the atmosphere or be adhered onto human skins. Through inhalation, dermal contact and ingestion, dust/soil particles can enter the human bodies and subsequently result in elevated exposure to toxic contaminants [9–12]. In dusty environments, it has been estimated that adults could ingest up to 100 mg of soil per day. While for children, the daily intake of soil may be as high as 200 mg due to their common habits of hand-to-mouth activities

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http://dx.doi.org/10.1016/j.jhazmat.2015.04.074
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[13]. Dust has now been recognized as the most important contributor to the Pb exposure of children. Many studies had found close relationships between dust Pb concentrations and children's blood Pb levels (BLLs) [14–17].

The real uptake (*i.e.*, absorption) of Pb *via* dust ingestion is greatly dependent on the solubility (*i.e.*, bioaccessibility) of Pb in the gastrointestinal system [18–20]. Due to the complex, time consuming and cost prohibitive nature of vivo assays [21], the oral bioaccessibility of Pb in soil/dust matrix has been usually predicted by various *in vitro* methods, such as dilute HCl extraction [22] and physiologically based extraction test (PBET) [19,20,23]. The PBET introduces digestive enzyme and pepsin into extraction solutions and mimics sequentially the chemical conditions in the gut and intestine and thus may provide valuable mechanistic information on the Pb mobilization from dust in the gastrointestinal tract [20]. In addition, the *in vitro* bioaccessibility measurement has been proved to be related to *in vivo* animal bioavailability [18,23,24–26]. However, many of such tests were designed for soil samples [27], while the application of PBET for surface dust samples has been relatively limited [20].

The most important sources of Pb in China are industrial operations (especially Pb/Zn mining and smelting) and various urban emissions, such as coal combustion, vehicle emissions, and municipal waste disposal [2–5]. Therefore, information about bioaccessibility of Pb from these representative sources is critical in health assessment for most citizens. In the present study, ground surface dust samples were collected from both a former zinc smelting area and a common urban district to characterize the profile differences in Pb bioaccessibility between these two areas by applying an *in vitro* PBET method. Subsequently, the human health risk associated with dust ingestion was estimated.

2. Materials and methods

2.1. Sample collection

The dust samples were collected from two typical areas. One is an old industrial region located in southwestern China (104°10′–105°03′E, 26°46′–27°28′N) with a population of eight hundred in 2013. The zinc smelting activities have been operating in this region since 1950s and banned in 2004 with a cumulative yield of about 300 thousand tons [7,28]. Dust samples in this region were collected from both indoor and outdoor (living room, bed room, courtyard and primary school playground) grounds. The second studied region is a metropolis (Wuhan, 113°41′–115°05′E, 29°58′–31°22′N) located in central China with a population of ten million in 2013. The major industries in this city include ferrous smelters, coal-power plants, chemical plants, machine manufacturing, and auto industries. Dust samples in this city were collected from outdoor ground (courtyard and kindergarten playground). Thirty seven and twenty one ground surface dust samples were collected from the industrial area and urban district, respectively, using a brush and plastic spatula [5]. In addition, relatively uncontaminated ground dusts from suburbs of each studied areas were collected as control samples. All dust samples were stored in sealed polyethylene bags, labeled and then transported to the laboratory.

2.2. Sample analytical procedure

The dust samples were air-dried at room temperature, and passed through a 60 mesh sieve (250 μm) to remove rocks, plants, hair and other impurities. For the determination of Pb and other elements, 50 mg of sample (<250 μm fraction) were digested using 1 ml of HF and 1 ml of HNO_3 in the PTFE-lined stainless steel bombs heated to 190 °C for 24 h. Insoluble residues were dissolved using 6 ml of 40% v/v HNO_3 heated to 140 °C for 5 h. After cooling down, about 0.4 ml of the digest was transferred to a centrifuge tube, and added with 500 ng rhodium in 5% v/v HNO_3, then made up to approximate 10 ml with Milli-Q water (18.2 MΩ cm, Millipore Inc.). Rhodium was used as an internal standard to correct for matrix effects and instrumental drift [29].

Thirty seven dust samples (27 from industrial area and 10 from urban district) were randomly selected for *in vitro* bioaccessibility determination. The PBET employed in this study was adopted from Ruby et al. [18] with some modifications proposed by Tang et al. [30]. In brief, the assay included the gastric (pH 1.5, 1 h, 37 °C)

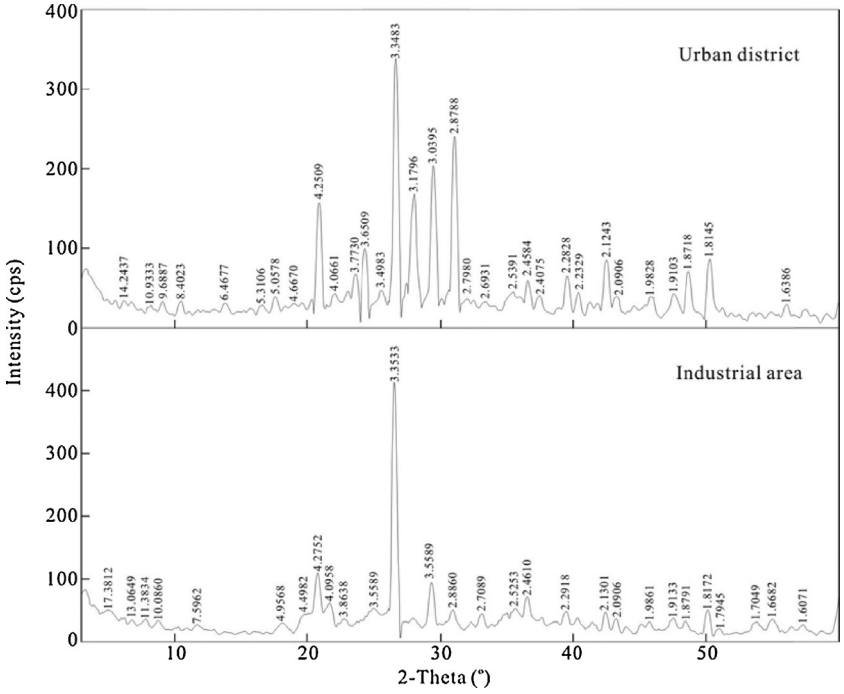

Fig. 1. X-ray diffraction pattern of selected dust samples.

and intestinal-phases (pH 7, 4 h, 37 °C). An aliquot of 2.0 g dust was mixed in 200 ml of synthetic digestive juice (composed of pepsin, sodium malate, sodium citrate, actic acid, acetic acid, bile salts, and pancreatin) and maintained in anaerobic conditions by purging argon gas.

The concentrations of trace metals in the total digested solution were determined using an inductively coupled plasma – mass spectrometry (ICP-MS, ELAN DRC-e, PerkinElmer Inc.). While the concentrations of Pb in PBET were measured using an atomic absorption spectroscopy (AAS, 5100PC, PerkinElmer Inc.). Major elements (Al, Ca, Fe, and P) were determined using an inductively coupled plasma-atomic emission spectrometer (ICP-AES). For quality assurance and quality control (QA/QC), the duplicates, method blanks and standard reference materials (soil and sediment SRM: GBW07305, GBW07405, and GBW(E)070009 from National Standard Material Center, China and NIST 2710 from National Institute of Standards and Technology, U.S.) were analyzed. Mean recoveries for studied elements (C(element, measured)/C(element, certified) × 100) in the four SRMs were between 85% and 113%. The duplicate samples showed the bias was less than 5%, indicating the samples are thoroughly homogenized [29].

Mineralogy of the randomly selected dust samples was characterized using a powder X-ray diffractometer (XRD) (X'Pert PRO DY2198, PANalytical Inc.). The diffraction pattern was recorded from 3° to 65°.

2.3. Pollution assessment

Pollution assessment for Pb and other trace metals was conducted using the geoaccumulation index (I_{geo}) introduced by Müller [31], which is calculated using the following equation:

$$I_{geo} = \log_2\left[\frac{C_{Sample}}{1.5 \times C_{Background}}\right] \quad (1)$$

where C_{Sample} is metal concentration in the dust, $C_{Background}$ is background value in the control samples. The factor 1.5 is introduced in this equation to minimize the effect of possible variations in the background values. According to Müller [31], the I_{geo} value is classified as: uncontaminated ($I_{geo} \leq 0$); uncontaminated to moderately contaminated ($0 < I_{geo} \leq 1$); moderately contaminated ($1 < I_{geo} \leq 2$); moderately to heavily contaminated ($2 < I_{geo} \leq 3$); heavily contaminated ($3 < I_{geo} \leq 4$); heavily to extremely contaminated ($4 < I_{geo} \leq 5$); extremely contaminated ($I_{geo} \geq 5$).

2.4. Health risk assessment

Risk assessment of children associated with exposure to Pb-contaminated dust by ingestion was carried out to evaluate the noncancer toxic risk of local inhabitants. The average daily intake (ADI) of Pb through unintentional dust ingestion was calculated as follow [32,33]:

$$ADI = \frac{C \times IngR \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (2)$$

where C is the total concentration of Pb in the dust; IngR is the ingestion rate of ground dust, which is assumed to be 200 mg/day [13]; EF is the exposure frequency which is assumed to be 350 day/year [13]; ED is the exposure duration, namely 6 years [13]; BW is the average body weight, with 18 kg for pre-school children [34]; and AT is the averaging time, equivalent to ED × 365 days.

If using the bioaccessible Pb to calculate the ADI, Eq. (2) was modified to be:

$$ADI = \frac{C \times IngR \times EF \times ED}{BW \times AT} \times B \times 10^{-6} \quad (3)$$

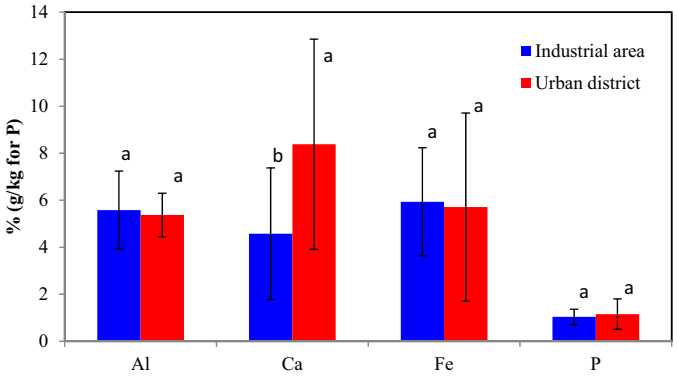

Fig. 2. The concentrations (mean ± SD) of Al, Ca, Fe, and P in the dust samples from industrial and urban areas. The different lower-case letters indicate significant difference ($p < 0.05$).

where B is the percentage of bioaccessible fraction of Pb in gastric or intestinal-phase.

The hazard quotient (HQ) was subsequently calculated as follows:

$$HQ = \frac{ADI}{RfD} \quad (4)$$

where RfD is a reference dose for Pb, which is assumed to be 3.5 μg/kg day [35]. The HQ assumes that there is a level of exposure (RfD), below which it is unlikely for sensitive populations to experience adverse health effects. If the exposure level exceeds this threshold (HQ > 1), potential adverse health effects may come into play. As a rule, the greater the value of HQ, the greater the level of concerns [13]. Recent research indicated that there is no evidence of a threshold for critical Pb induced effects influencing children [36], therefore, the ever used RfD for Pb (Provisional Tolerable Weekly Intake, PTWI) is no longer considered appropriate. But we still used it in our assessment of Pb risk since there is no alternative.

2.5. Statistical analysis

The data were statistically analyzed using the software SPSS v13.0 (SPSS Inc.). The correlation analysis between metals concentrations was conducted by a Pearson correlation, and the level of significance was set at $p < 0.05$ and $p < 0.01$ (two-tailed). An independent sample T test ($p < 0.05$) was used to assess the difference of observed results between industrial and urban areas and between the stomach and intestinal phases.

3. Results and discussions

3.1. Dust mineralogy and major element concentrations

The results of XRD analysis of the dust samples are shown in Fig. 1. The samples from the industrial areas were mainly composed of quartz, illite, calcite, montmorillonite, feldspar, and a small quantity of iron minerals. Comparatively, the dusts from the urban environment displayed a slightly more complicated mineral composition of quartz, montmorillonite, illite, amphibole, calcite, dolomite, feldspar, and iron minerals. The concentrations of selected major elements (Al, Ca, Fe, and P) are compared in Fig. 2. There were no statistical differences ($p > 0.05$) of the major element concentrations between the industrial area and urban district, except for Ca which exhibited significantly higher ($p < 0.05$) concentrations in the urban district than those in the industrial area. This may be due to the extensive use of cement and lime in urban construction.

Table 1
The concentrations of trace metals in the dust samples (mg/kg).

		Pb	Cd	Co	Cr	Cu	Ni	Zn
Industrial area (n = 37)								
	Mean	2182	26	25	105	371	67	3488
	SD	1676	22	9.0	36	413	32	2526
	Min	220	2.2	4.0	32	49	11	256
	Max	6348	124	42	195	2442	143	8245
Control sample (n = 3)	Mean	47	0.74	10	92	75	28	157
Urban district (n = 21)								
	Mean	281	1.6	14	130	138	33	835
	SD	316	1.2	7.4	98	114	11	630
	Min	79	0.475	8	64	46	17	243
	Max	1544	4.445	39	455	471	61	2459
Control sample (n = 3)	Mean	43	0.62	12	57	33	23	121

3.2. Distribution of Pb and other trace metals in dust

The concentrations of Pb along with Cd, Co, Cr, Cu, Ni, and Zn in the surface dust samples from the industrial area and urban district are listed in Table 1. For the industrial area, the concentrations of trace metals in surface dust exhibited considerable variations, especially for Pb, Cd, Cu, and Zn which varied tens of folds from 220 to 6348 mg/kg, 2.2–124 mg/kg, 49–2442 mg/kg, and 256–8245 mg/kg, respectively. In comparison with data from the urban district, the mean concentrations of Pb (2182 mg/kg), Cd (26 mg/kg), and Zn (3488 mg/kg) from the industrial area were 7.8, 16.3, and 4.2 times higher, respectively. The correlation matrices between dust metal concentrations in each studied areas are shown in Table 2, and close correlations ($p < 0.01$, $r^2 = 0.392$–0.701, $n = 37$) among Pb, Cd, and Zn were noticed in samples from the industrial area. The simultaneously elevated concentrations of Pb, Cd, and Zn in surface dust were also observed in other zinc smelting industrial areas, such as Zhuzhou [29] and Huludao [37] in China and Avilés in Spain [38]. Therefore, the composition profiles of Pb, Cd, and Zn can be a representative fingerprint of zinc smelting activities. For the urban district, the concentrations of trace metals in the dust were comparable to those from other common cities, such as Shanghai [39], Guangzhou [4], HongKong [40], Seoul [41], London [42], and Oslo [43]. But the variations of metal concentrations were generally less pronounced than those of the industrial area. Lead and Zn, for instance, varied about a dozen folds from 79 to 1544 mg/kg and 243–2459 mg/kg, respectively. In addition, the relationships between metal concentrations in dust from the urban area were somewhat different from those of the industrial area (Table 2). For example, there were no significant correlations between Pb and other metals in the urban dusts, which may be attributed to a mixture of multi-sources of Pb within the urban environment, such as coal combustion and traffic emissions [44].

Table 2
Correlation matrix of trace metals in the dust samples.

	Pb	Cd	Co	Cr	Cu	Ni
Industrial area (n = 37)						
Cd	0.626**					
Co	0.350*	0.338*				
Cr	0.031	0.050	0.575**			
Cu	0.479**	0.824**	0.338*	0.082		
Ni	0.270	0.454**	0.653**	0.521**	0.678**	
Zn	0.837**	0.763**	0.486**	0.0962	0.529**	0.391*
Urban district (n = 21)						
Cd	0.192					
Co	0.368	0.594**				
Cr	0.113	0.281	0.598**			
Cu	0.252	0.718**	0.665**	0.703**		
Ni	0.204	0.512*	0.695**	0.741**	0.791**	
Zn	0.276	0.692**	0.479*	0.230	0.471*	0.382

* Significant level at $p < 0.05$ (two-tailed).
** Significant level at $p < 0.01$ (two-tailed).

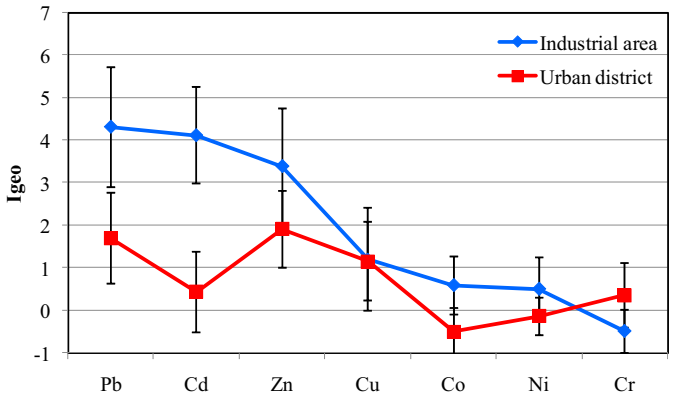

Fig. 3. Geoaccumulation index (I_{geo}) of trace metals in the dust samples from industrial and urban areas.

Table 3
Bioaccessible Pb in the stomach and intestinal phases.

	Stomach-phase		Intestinal-phase	
	(mg/kg)	(%)	(mg/kg)	(%)
Industrial area (n = 27)				
Mean	1309	41.5	207	5.8
SD	919	12.8	281	5.4
Min	218	19.2	14.5	1.2
Max	3466	76.1	1159	21.8
Urban district (n = 10)				
Mean	166	39.0	27.6	8.5
SD	219	17.0	26.6	6.4
Min	31.0	17.6	4.0	1.3
Max	772	64.5	91	17.7
Total (n = 37)				
Mean	1000	40.8	158	6.5
SD	942	13.9	253	5.7
Min	31.0	17.6	4.0	1.2
Max	3466	76.1	1159	21.8

In comparison with those of the control samples, the concentrations of trace metals in the dusts from both the industrial and urban areas were notably elevated (Table 1). The contamination status of trace metals can be clearly reflected by the geoaccumulation index (I_{geo}) (Fig. 3). For industrial area, Pb, Cd, and Zn were the principal metal pollutants, characterized by heavily to extremely contaminated (I_{geo} = 3∼5). While the dust samples from urban district were only moderately contaminated by Zn, Pb, and Cu (I_{geo} = 1∼2). Comparatively, the contamination of Co, Cr, and Ni in both the studied areas was moderate-to-low (I_{geo} = −1∼1).

3.3. Lead bioaccessibility

The bioaccessibility of Pb in selected dust samples is shown in Table 3. Bioaccessibility in this study is expressed as both absolute concentration in the stomach and intestinal phases and its relative percentage of the total concentration digested in concentrated strong acids.

3.3.1. Bioaccessibility in the stomach-phase

For the overall samples, the bioaccessibility of Pb in stomach-phase ranged from 17.6 to 76.1% with a mean of 40.8%. Pb bioaccessibility seems to depend on pollution sources and sample matrix. High gastric-phase Pb bioaccessibility has been reported for samples contaminated by lead shot due to the presence of highly soluble Pb oxides and carbonates [45]. In contrast, low Pb bioac-

Table 4
Comparison of estimated daily exposure to Pb through dust ingestion with dietary intake to children in the studies areas.

	ADI (μg/kg day)				HQ			
	Dust digestion			Diet[a]	Dust digestion			Diet
	Total	Stomach	Intestinal		Total	Stomach	Intestinal	
Industrial area	27.8	13.8	2.20	2.54	7.94	3.95	0.63	0.73
Urban district	4.27	1.76	0.29	2.54	1.22	0.51	0.08	0.73

[a] The concentrations of Pb in crops from the industrial area were similar to the average values in China [58,59], thus the average daily Pb intake of the Chinese was used in both the industrial and urban areas [60].

cessibility was usually found in mine site soils due to the limited solubility of Pb species such as Pb sulphides, Pb phosphates, ferromanganese Pb oxides and iron Pb sulphates [18,46]. In this study, the Pb bioaccessibility in the dust samples from the Zn smelting area and urban district was almost the same despite the significant difference in their total Pb concentrations (Table 1 and 3), which may be attributed to the similar dust matrix and Pb species within these samples. Our data have shown that the dust samples between the two studied areas had similar mineral compositions and major element concentrations (Figs. 1 and 2). X-ray absorption near-edge structure (XANES) and extended X-ray absorption fine structure (EXAFS) analysis showed that Pb in the urban dust existed as $PbCrO_4$ and Pb-sorbed goethite, as well as soluble Pb oxides and carbonates [47]. Lead oxides and carbonates are usually found in smelting originated contaminants [48,49]. Furthermore, a chemical sequential extraction analysis revealed that Pb pollutant from both the industrial area [50,51] and the urban environment [52] was liable to be bound with carbonate and iron/manganese oxide fractions. Therefore, a substantial portion of the bioaccessible Pb in both the industrial and urban dust might be derived from the dissolution of Pb oxides or/and carbonates.

3.3.2. *Bioaccessibility in the intestinal-phase*

Compared with that in the gastric-phase, bioaccessible Pb in the intestine (1.2–21.8%) was significantly lower ($p < 0.01$). The mean bioaccessible Pb in the urban dust was slightly higher than that in the industrial samples, but the difference was not significant ($p > 0.05$). A reduction in Pb bioaccessibility from stomach to intestine has been reported previously in both soil and dust [20,23,53,54]. A number of studies found that co-precipitation with Fe and sorption to the amorphous Fe by surface complexation could reduce Pb bioaccessibility in the intestinal-phase [23,26,45,55]. However, Fe precipitation in the intestinal-phase of PBET assay was inhibited by citrate-complexed Fe, which has a high stability constant [23]. Therefore, the reduction in Pb bioaccessibility from stomach to intestine in this PBET assay seems to be controlled by other factors. In this study, a significantly negative correlation was found between dust Ca concentrations and Pb bioaccessibility in the intestinal-phase (Fig. 4), suggesting that calcium minerals may play an important role in reducing the bioaccessible Pb in the intestinal-phase. A follow-up XANES study is needed to investigate this inhibition mechanism in intestinal extraction.

3.4. *Implications for human exposure associated with dust ingestion*

In this study, the sum of Pb exposure through dust ingestion and dietary intake was used as an approximate estimation of total Pb exposure to children, because they are the most important sources of Pb exposure and generally account for above 90% of the total exposure inventory [56]. The daily exposure of Pb through ingestion of dust and dietary intake for reference is shown in Table 4. The ADI values calculated from total Pb concentrations were obviously higher than those from bioaccessible Pb and yielded HQ levels of

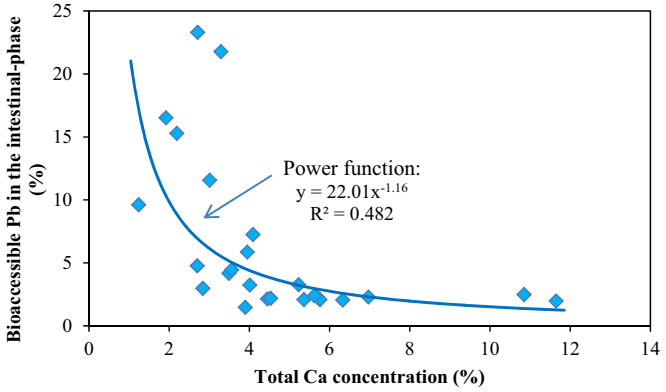

Fig. 4. Relationship between bioaccessible Pb in the intestinal-phase and total Ca concentrations in the dust.

7.94 and 1.22 for industrial area and urban district, respectively. In this sense, however, the estimate would be the upper bound. Compared with the total concentrations, the bioaccessible Pb can produce more radical results in the exposure assessment because Pb bioaccessibility usually has a close correlation with *in vivo* Pb relative bioavailability [23,26]. If gastric bioaccessible Pb was used for estimation, the ADI of children living in the industrial and urban areas would be 13.8 and 1.77 μg/kg day, respectively, accounting for 85% and 41% of their corresponding total Pb exposure (sum of stomach absorption of dust and dietary absorption). This suggested that dust is an important contributor to the total Pb exposure. In addition, for children living in both the studied areas, the total exposure to Pb (dust + diet) has all exceeded the RfD (HQ > 1). Our findings suggest that children in industrial and urban areas of China experience environmental Pb exposes of concern. However, if intestinal bioaccessible Pb was taken into account, the ADI and the subsequent HQ values would decline significantly (Table 4). Despite the fact that the absorption of Pb in humans takes place primarily in the small intestine [45], the intestinal-phase extracted Pb has rarely been related to its relative bioavailablility determined in *in vivo* assays. This appears to be due to the complex, non-equilibrium chemical status of Pb in the small intestines [19,23]. Consequently, dissolution of Pb in the gastric-phase appears to be a better predictor [19,23,57]. Therefore, the gastric bioaccessible Pb may be considered as an appropriate proxy for a conservative assessment of human health risk.

4. Conclusion

The bioaccessibility of Pb in contaminated dust samples from industrial and urban areas was estimated using an *in vitro* PBET assay. No significant differences of gastrointestinal bioaccessibile Pb were found in the dust samples between the smelting industrial area and urban district, although their total Pb concentrations differed greatly. Compared with the stomach-phase, the intestinal-phase had a considerably lower bioaccessibility of Pb. A significantly negative correlation was found between dust Ca

concentrations and Pb bioaccessibility in the intestinal-phase, suggesting that Ca compounds may play an important role in reducing bioaccessible Pb in the intestinal phase. The estimated exposure based on gastric bioaccessible Pb along with dietary intake for reference suggested that dust is an important contributor to the total Pb exposure of children living in either of the studied areas. Therefore, our research provided solid evidence to highlight the importance of reducing children's risk of exposure to Pb in dust.

Acknowledgements

We appreciate the four anonymous referees for their valuable suggestions and questions which led to significant improvements of the previous manuscript. The study was supported by the National Natural Science Foundation of China (No. 41273003) and the Fundamental Research Funds for the Central Universities (CUG130502), China University of Geosciences (Wuhan).

References

[1] B.P. Lanphear, R. Hornung, J. Khoury, K. Yolton, P. Baghurst, D.C. Bellinger, R.L. Canfield, K.N. Dietrich, R.L. Bornschein, T. Greene, S.J. Rothenberg, H.L. Needleman, L. Schnaas, G. Wasserman, J. Graziano, R. Roberts, Low level environmental lead exposure and children's intellectual function: an international pooled analysis, Environ. Health Perspect. 113 (2005) 894–899.
[2] X.D. Li, C.S. Poon, P.S. Liu, Heavy metal contamination of urban soils and street dusts in Hong Kong, Appl. Geochem. 16 (2001) 1361–1368.
[3] T. Chen, Y. Zheng, M. Lei, Z. Huang, H. Wu, H. Chen, K. Fan, K. Yu, X. Wu, Q. Tian, Assessment of heavy metal pollution in surface soils of urban parks in Beijing, China, Chemosphere 60 (2005) 542–551.
[4] N.S. Duzgoren-Aydin, C.S.C. Wong, A. Aydin, Z. Song, M. You, X.D. Li, Heavy metal contamination and distribution in the urban environment of Guangzhou, SE China, Environ. Geochem. Health 28 (2006) 375–391.
[5] X.Y. Bi, S. Liang, X.D. Li, A novel in situ method for sampling urban soil dust: Particle size distribution, trace metal concentrations, and stable lead isotopes, Environ. Pollut. 177 (2013) 48–57.
[6] L.Y. Cai, Z.X. Wang, Y.Y. Wang, Z.H. Yang, H.Y. Wang, X. Wu, Ingestion risks of metals in groundwater based on TIN model and dose-response assessment – a case study in the Xiangjiang watershed, central-south China, Sci. Total Environ. 408 (2010) 3118–3124.
[7] X.Y. Bi, X.B. Feng, Y.G. Yang, G.L. Qiu, G.H. Li, F.L. Li, T.Z. Liu, Z.Y. Fu, Z.S. Jin, Environmental contamination of heavy metals from zinc smelting areas in Hezhang County, western Guizhou, China, Environ. Int. 32 (2006) 883–890.
[8] X.Y. Bi, S.Y. Liang, X.D. Li, Trace metals in soil dust, and tree leaves of the urban environment, Guangzhou, China, Chin. Sci. Bull. 58 (2013) 222–230.
[9] K. Ljung, O. Selinus, E. Otabbong, M. Berglund, Metal and arsenic distribution in soil particle sizes relevant to soil ingestion by children, Appl. Geochem. 21 (2006) 1613–1624.
[10] N. Yamamoto, Y. Takahashi, J. Yoshinaga, A. Tanaka, Y. Shibata, Size distributions of soil particles adhered to children's hands, Arch. Environ. Contam. Toxicol. 51 (2006) 157–163.
[11] F. Amato, M. Pandolfi, M. Viana, X. Querol, A. Alastuey, T. Moreno, Spatial and chemical patterns of PM_{10} in road dust deposited in urban environment, Atmos. Environ. 43 (2009) 1650–1659.
[12] A. Peña-Fernández, M.J. González-Muñoz, M.C. Lobo-Bedmar, Establishing the importance of human health risk assessment for metals and metalloids in urban environments, Environ. Int. 72 (2014) 176–185.
[13] US-EPA, Supplemental Guidance for Developing Soil Screening Levels for Superfund Sites. OSWER9355. 4–24. Office of Solid Waste and Emergency Response (2001).
[14] D.J. Paustenbach, B.L. Finley, T.F. Long, The critical role of house dust in understanding the hazards posed by contaminated soils, Int. J. Toxicol. 16 (1997) 339–362.
[15] B.P. Lanphear, T.D. Matte, J. Rogers, R.P. Clickner, B. Dietz, R.L. Bornschein, et al., The contribution of lead–contaminated house dust and residential soil to children's blood lead levels. A pooled analysis of 12 epidemiologic studies, Environ. Res. 79 (1998) 51–68.
[16] M.A.S. Laidlaw, M.P. Taylor, Potential for childhood lead poisoning in the inner cities of Australia due to exposure to lead in soil dust, Environ. Pollut. 159 (2011) 1–9.
[17] B. Gulson, K. Mizon, A. Taylor, M. Korsch, J.M. Davis, H. Louie, M. Wu, L. Gomez, L. Antin, Pathways of Pb and Mn observed in a 5-year longitudinal investigation in young children and environmental measures from an urban setting, Environ. Pollut. 191 (2014) 38–49.
[18] M.V. Ruby, A. Davis, T.E. Link, R. Schoof, R.L. Chaney, G.B. Freeman, P. Bergstrom, Development of an *in vitro* screening test to evaluate the *in vivo* bioaccessibility of ingested mine-waste lead, Environ. Sci. Technol. 27 (1993) 2870–2877.
[19] M.V. Ruby, A. Davis, R. Schoof, S. Eberle, C.M. Sellstone, Estimation of lead and arsenic bioavailability using a physiologically based extraction test, Environ. Sci. Technol. 30 (1996) 422–430.
[20] A. Turner, K.H. Ip, Bioaccessibility of metals in dust from the indoor environment: application of a physiologically based extraction test, Environ. Sci. Technol. 41 (22) (2007) 7851–7856.
[21] S.W. Casteel, C.P. Weis, G.M. Henningsen, W.J. Brattin, Estimation of relative bioavailability of lead in soil and soil-like materials using young swine, Environ. Health Perspect. 114 (2006) 1162–1171.
[22] C.R. Thums, M.E. Farago, I. Thornton, Bioavailability of trace elements in brownfield soils in an urban area in the UK, Environ. Geochem. Health 30 (2008) 549–563.
[23] H.B. Li, X.Y. Cui, K. Li, J. Li, A.L. Juhasz, L.Q. Ma, Assessment of *in vitro* lead bioaccessibility in house dust and its relationship to *in vivo* lead relative bioavailability, Environ. Sci. Technol. 48 (2014) 8548–8555.
[24] D. Bannon, J.W. Drexler, G.M. Fent, S.W. Casteel, P.J. Hunter, W.J. Brattin, M.A. Major, Evaluation of small arms range soils for metal contamination and lead bioavailability, Environ. Sci. Technol. 43 (2009) 9071–9076.
[25] A.L. Juhasz, J. Weber, E. Smith, R. Naidu, B. Marschner, M. Rees, A. Rofe, T. Kuchel, L. Sansom, Evaluation of SBRC-gastric and SBRC-intestinal methods for the prediction of *in vivo* relative lead bioavailability in contaminated soils, Environ. Sci. Technol. 243 (2009) 4503–4509.
[26] E. Smith, I.M. Kempson, A.L. Juhasz, J. Weber, A. Rofe, D. Gancarz, R. Naidu, R.G. McLaren, M. Gräfe, *In vivo-in vitro* and XANES spectroscopy assessments of lead bioavailability in contaminated periurban soils, Environ. Sci. Technol. 45 (2011) 6145–6152.
[27] M.H. Zia, E.E. Codling, K.G. Scheckel, R.L. Chaney, *In vitro* and *in vivo* approaches for the measurement of oral bioavailability of lead (Pb) in contaminated soils: a review, Environ. Pollut. 159 (2011) 2320–2327.
[28] X.Y. Bi, X.B. Feng, Y.G. Yang, X.D. Li, G.P.Y. Shin, F.L. Li, G.L. Qiu, G.H. Li, T.Z. Liu, Z.Y. Fu, Allocation and source attribution of lead and cadmium in maize (Zea mays L.) impacted by smelting emissions, Environ. Pollut. 157 (2009) 834–839.
[29] Z.G. Li, X.B. Feng, G.H. Li, X.Y. Bi, J.M. Zhu, H.B. Qin, Z.H. Dai, J.L. Liu, Q.H. Li, G.Y. Sun, Distribution, sources and pollution status of 17 trace metal/metalloids in the street dust of a heavily industrialized city of central China, Environ. Pollut. 182 (2013) 408–416.
[30] X.Y. Tang, Y.G. Zhu, Y.S. Cui, J. Duan, L. Tang, The effect of ageing on the bioaccessibility and fractionation of cadmium in some typical soils of China, Environ. Int. 32 (2006) 682–689.
[31] G. Müller, Index of geoaccumulation in sediments of the Rhine River, Geojournal 2 (1969) 108–118.
[32] US-EPA, Human Health Evaluation Manual, Risk Assessment Guidance for Superfund, vol. I, Office of Soild Waste and Emergency Response, 1989 EPA/540/1-89/002.
[33] US-EPA, Soil Screening Guidance: Technical Background Document, Office of Soild Waste and Emergency Response, 1996 EPA/540/R-95/128.
[34] MOH (Ministry of Health, China). China Health Statistical Year book in 2010. Peking Union Medical College press, Beijing (2011).
[35] JECFA, WHO, Food Additives Series 32 (1993): 253–263. Geneva, Switzerland: World Health Organization.
[36] EFSA, Panel on Contaminants in the food chain (CONTAM); scientific opinion on lead in food, EFSA J. 8 (4) (2010) 1570.
[37] N. Zheng, J. Liu, Q. Wang, Z. Liang, Health risk assessment of heavy metal exposure to street dust in the zinc smelting district, Northeast of China, Sci. Total. Environ. 408 (2010) 726–733.
[38] A. Ordóñez, J. Loredo, E. De Miguel, S. Charleswort, Distribution of heavy metals in the street dusts and soils of an industrial city in northern Spain, Arch. Environ. Contam. Toxicol. 44 (2003) 160–170.
[39] G. Shi, Z. Chen, C. Bi, L. Wang, J. Teng, Y. Li, S. Xu, A comparative study of health risk of potentially toxic metals in urban and suburban road dust in the most populated city of China, Atmos. Environ. 45 (2011) 764–771.
[40] P.A. Tanner, H.L. Ma, P.K.N. Yu, Fingerprinting metals in urban street dust of Beijing Shanghai, and Hong Kong, Environ. Sci. Technol. 42 (2008) 7111–7117.
[41] H.T. Chon, K.W. Kim, J.Y. Kim, Metal contamination of soils and dusts in Seoul metropolitan city, Korea, Environ. Geochem. Health 17 (1995) 139–146.
[42] M.J.R. Schwar, J.S. Moorcroft, D.P.H. Laxen, M. Thompson, C. Armorgie, Baseline metal-in-dust concentrations in Greater London, Sci. Total Environ. 68 (1988) 25–43.
[43] E. De Miguel, J.F. Llamas, E. Chacón, T. Berg, S. Larssen, O. Røyset, M. Vadset, Origin and patterns of distribution of trace elements in street dust: unleaded petrol and urban lead, Atmos. Environ. 31 (1997) 2733–2740.
[44] Z.M. Zhu, G.Y. Sun, X.Y. Bi, Z.G. Li, G.H. Yu, Identification of trace metal pollution in urban dust from kindergartens using magnetic, geochemical and lead isotope analyses, Atmos. Environ. 77 (2013) 9–15.
[45] E. Smith, J. Weber, R. Naidu, R.G. McLaren, A.L. Juhasz, Assessment of lead bioaccessibility in peri-urban contaminated soils, J. Hazard. Mater. 186 (2011) 300–305.
[46] J.L. Schroder, N.T. Basta, S.W. Casteel, T.J. Evans, M.E. Payton, J. Si, Validation of the *in vitro* gastrointestinal (IVG) method to estimate relative bioavailable lead in contaminated soils, J. Environ. Qual. 33 (2004) 513–521.
[47] J.E.S. Barrett, K.G. Taylor, K.A. Hudson-Edwards, J.M. Charnock, Solid-phase speciation of Pb in urban road dust sediment: a XANES and EXAFS study, Environ. Sci. Technol. 44 (2010) 2940–2946.

[48] R.A. Schoof, M.K. Butcher, C. Sellstone, R.W. Ball, J.R. Fricke, V. Keller, B. Keehn, An assessment of lead absorption from soil affected by smelter emissions, Environ. Geochem. Health 17 (1995) 189–199.
[49] J. Yang, D.E. Mosby, S. Casteel, R.W. Blanchar, Lead immobilization using phosphoric acid in a smelter-contaminated urban soil, Environ. Sci. Technol. 35 (2001) 3553–3559.
[50] P. Wu, C.Q. Liu, Y.G. Yang, G.P. Zhang, Environmental impacts and geochemical partitioning of heavy metals (Pb Zn) in the historical Zn smelting wastes, Geochimica 32 (2003) 139–145 (in Chinese).
[51] Y.G. Yang, C.Q. Liu, P. Wu, G.P. Zhang, Soil heavy metal accumulation induced by local smelting and its microbial environmental effects in Hezhang County, Guizhou Province, Geochimica 32 (2003) 131–138 (in Chinese).
[52] R.C. Hu, L. Qiu, Speciation of heavy metal contaminants in top soils and atmospheric deposited dust from urban area of Wuhan, China, Resour. Environ. Eng. 22 (2008) 31–35 (in Chinese).
[53] M.V. Ruby, A. Davis, J.H. Kempton, J.W. Drexler, P.D. Bergstrom, Lead bioavailability: dissolution kinetics under simulated gastric conditions, Environ. Sci. Technol. 26 (1992) 1242–1248.
[54] D.P. Oliver, M.J. McLaughlin, R. Naidu, L.H. Smith, E.J. Maynard, I.C. Calder, Measuring Pb bioavailability from household dusts using an *in vitro* model, Environ. Sci. Technol. 33 (1999) 4434–4439.
[55] S.E. O'Reilly Jr, M.F. Hochella, Lead sorption efficiencies of natural and synthetic Mn and Fe-oxides. Geochim, Cosmochim. Acta 67 (2003) 4471–4487.
[56] IPCS, Environmental health criteria 85: Inorganic Lead, Geneva WHO, (1993) 49–50.
[57] US-EPA, Guidance for evaluating the oral bioavailability of metals in soils for use in human risk assessment (OSWER 9285.7-80), (2007).
[58] K.P. He, L.J. Zhang, C.M. Shi, X.W. Zhu, K.Y. Wang, Assessment of lead contamination and exposure of residents and crops in traditional artisanal zinc smelting areas, J. Environ. Occup. Med. 34 (4) (2013) 263–267 (in Chinese).
[59] Y.E. Shang, Z.Q. Yu, F.C. Meng, J. Guo, Research and analysis about Chinese dietary intake of lead, J. Beijing Technol. Bus. Univ. (Nat. Sci. Ed.) 30 (3) (2012) 81–84 (in Chinese).
[60] X.W. Li, Q. Liu, L.P. Liu, Y.N. Wu, Application of the data from China total diet study to assess the distribution of lead exposure in different age-gender population groups, J. Hyg. Res. 41 (3) (2012) 379–384 (in Chinese).