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RESEARCH ARTICLE

Assessing the concentration and potential health risk of heavy metals in China's main deciduous fruits

NIE Ji-yun[1], KUANG Li-xue[1], LI Zhi-xia[1], XU Wei-hua[2], WANG Cheng[3], CHEN Qiu-sheng[4], LI An[5], ZHAO Xu-bo[6], XIE Han-zhong[7], ZHAO Duo-yong[3], WU Yong-long[1], CHENG Yang[1]

[1] *Institute of Pomology, Chinese Academy of Agricultural Sciences/Laboratory of Quality & Safety Risk Assessment for Fruit (Xingcheng), Ministry of Agriculture, Xingcheng 125100, P.R.China*
[2] *Yantai Academy of Agricultural Sciences, Yantai 265500, P.R.China*
[3] *Institute of Quality Standards & Testing Technology for Agro-Products, Xinjiang Academy of Agricultural Sciences, Urumchi 830091, P.R.China*
[4] *Institute of Tianjin Agriculture Quality Standard and Testing Technology, Tianjin 300381, P.R.China*
[5] *Beijing Research Center for Agricultural Standards and Testing, Beijing 100097, P.R.China*
[6] *College of Food Science and Engineering, Northwest A&F University, Yangling 712100, P.R.China*
[7] *Zhengzhou Institute of Pomology, Chinese Academy of Agricultural Sciences, Zhengzhou 450009, P.R.China*

Abstract

To assess levels of contamination and human health risk, we analyzed the concentrations of the heavy metals lead (Pb), cadmium (Cd), chromium (Cr), and nickel (Ni) in China's main deciduous fruits — apple, pear, peach, grape, and jujube. The concentration order of the heavy metals was Ni>Cr>Pb>Cd. In 97.5% of the samples, heavy metal concentrations were within the maximum permissible limits. Among the fruits studied, the heavy metal concentrations in jujube and peach proved to be the highest, and those in grape proved to be the lowest. Only 2.2% of the samples were polluted by Ni, only 0.4% of the samples were polluted by Pb, and no samples were polluted by Cd or Cr. Compared with the other fruits, the combined heavy metal pollution was significantly higher ($P<0.05$) in peach and significantly lower ($P<0.05$) in grape. For the combined heavy metal pollution, 96.9% of the samples were at safe level, 2.32% at warning level, 0.65% at light level, and 0.13% at moderate level. In the fruits studied, the contribution of heavy metals to the daily intake rates (DIR) followed the order of Ni>Cr>Pb>Cd. The highest DIR came from apple, while the lowest DIR came from grape. For each of the heavy metals, the total DIR from five studied fruits corresponded to no more than 1.1% of the tolerable daily intake, indicating that no significant adverse health effects are expected from the heavy metals and the fruits studied. The target hazard quotients and the total target hazard quotients demonstrated that none of the analyzed heavy metals may pose risk to consumers through the fruits studied. The highest risk was posed by apple, followed in decreasing order by peach and pear, jujube, and grape. We suggest that the main deciduous fruits (apple, pear, peach, grape, and jujube) of China's

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Correspondence NIE Ji-yun, Tel: +86-429-3598178,
E-mail: jiyunnie@163.com

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main producing areas are safe to eat.

Keywords: deciduous fruits, heavy metals, health risk assessment, China

1. Introduction

Heavy metals such as lead (Pb), cadmium (Cd), chromium (Cr), and nickel (Ni) generally refer to metals and metalloids having densities greater than 5 g cm^{-3} (Oves *et al.* 2012). Barring occupational exposures, the main route of human exposure to heavy metals is through dietary intake (Sharma and Tripathi 2008). Once the heavy metals are dispersed into water, soil and air, they can accumulate in the crops (Hao *et al.* 2009; Hernández-Martínez and Navarro-Blasco 2012). Heavy metal pollution of food items is one of the most important aspects of food quality assurance (Wang *et al.* 2005; Radwan and Salama 2006; Khan *et al.* 2008). Fruits can accumulate high levels of heavy metals in their edible parts (Roba *et al.* 2016). Heavy metal pollution in fruits is arisen by many ways, such as irrigation water, industrial emissions, the harvesting process, storage and/or at the point of sale (Huang *et al.* 2014).

Heavy metals are harmful because of their non-biodegradable nature, long biological half-lives, and potential to accumulate in body (Arora *et al.* 2008). Prolonged consumption of unsafe concentrations of heavy metals through foodstuffs may lead to the chronic accumulation of heavy metals in the kidney and liver of humans, causing disruption of numerous biochemical processes, and leading to cardiovascular, nervous, kidney and bone diseases (Järup 2003; Sharma *et al.* 2009). Some heavy metals such as Cd, Cr, and Pb, are nonessential and can cause negative human health effects (Järup 2003; Ferré-Huguet *et al.* 2008; Martí-Cid *et al.* 2008a; Martorell *et al.* 2011). Other heavy metals, such as Ni, are micronutrients for human beings, but excessive intake may affect health (Powers *et al.* 2003). The consumption of foodstuff polluted with heavy metals may lead to accumulation of these contaminants in different tissues, causing both chronic and acute health outcomes (Järup 2003). It is therefore reasonable to hypothesize that the intake of fruits containing heavy metals is a potential health risk to consumers.

Fruits contain carbohydrates, proteins, vitamins, minerals, and fibers required for human health (Cherfi *et al.* 2014). They are important components of human diet both in terms of consumed quantities and nutritional value (Roba *et al.* 2016). In China, apple (*Malus spp*. Mill.), pear (*Pyrus spp*.), peach (*Prunus persica* L.), grape (*Vitis* L.), and jujube (*Ziziphus jujube* Mill.) are the most important deciduous fruits, accounting for 55% of the total fruit output and more than 80% of the total deciduous fruit output (CAYEC 2014). Although some studies have reported the heavy metal pollution and its health risk in fruits cultivated in China (Xiao *et al.* 2010; Sheng *et al.* 2014), as far as we know, there are few studies focusing on China's main deciduous fruits and their main producing areas. This study aimed to investigate the concentrations and pollution of the heavy metals Pb, Cd, Cr, and Ni in the above-mentioned deciduous fruits cultivated in China's main producing areas, and to assess the possible human health risk associated with consumption of these fruits by calculating the daily intake rates (DIR) and the target hazard quotients (THQ). The results of our study may provide some insight into heavy metal pollution for main deciduous fruits in China, and serve as a basis for comparison with other countries and other fruits.

2. Materials and methods

2.1. Sampling and preparation

A total of 775 deciduous fruit samples (Table 1) were collected at harvest time in 2014 from the main producing areas of China, including Liaoning, Shaanxi, Shandong, Hebei, Xinjiang, Jiangsu, Henan, and Anhui (Fig. 1). Production from these provinces account for 62.2, 57.6, 63.2, 57.8, and 74.9 of Chinese total output of apple, pear, peach, grape,

Table 1 Number of fruit samples from different provinces of China

Fruits	Anhui	Hebei	Henan	Jiangsu	Liaoning	Shandong	Shaanxi	Xinjiang	Total
Apple		42			55	60	55		212
Pear		50		10	49	25	15	46	195
Peach	10	25	20	10	20	32	20		137
Grape		20		15	15	22	10	55	137
Jujube		20				24	20	30	94
Total	10	157	20	35	139	163	120	131	775

and jujube, respectively (NBSC 2013). The numbers of sampling counties in related provinces were determined by the yield of the related fruits in each province. Normally, five representative orchards were chosen in each county, and one sample was collected from each orchard. Each sample (10 fruits for apple, pear and peach; five bunches for grape; and 1 kg for jujube) was randomly collected from five trees in a single orchard. All products were sampled by trained and authorized inspectors. Sample collection was performed according to national guideline (GB/T 8855-2008). All samples and sub-samples were washed with distilled water and homogenized using a food processor.

2.2. Determination of heavy metal concentrations

The determination of heavy metal concentrations references to related national standards (GB/T 5009.138-2003; GB 5009.12-2010; GB 5009.123-2014; GB 5009.15-2014). Each sample was digested according to the procedures listed in Table 2, and then transferred to a volumetric flask and diluted to 10–25 mL with deionized water. For the determination of concentrations of Pb, Cd, Cr and Ni, the two solution of each sample were analyzed by graphite furnace atomic absorption spectrometry, the instrument reference conditions were listed in Table 3. The limits of

Fig. 1 Location map of the study area showing the 118 sampling counties.

Table 2 Digestion procedures for four heavy metals (HMs) in fruits

HMs →	Weight sample →	Baked to nearly dry →	Add HNO_3 →	Soaked →	Add H_2O_2 →	Maintained →	Cooled
Ni	2–5 g	At 80°C	5 mL	≥12 h	7 mL	120ºC, 2–3 h	Naturally
Pb	1–2 g		2–4 mL	≥12 h	2–3 mL	120–140ºC, 3–4 h	Naturally
Cd	1–2 g		5 mL	≥12 h	2–3 mL	120–160ºC, 4–6 h	Naturally
Cr	0.3–1 g		5 mL			140–160ºC, 4–5 h	Naturally

Table 3 The instrument reference conditions for determination of four heavy metals (HMs)

HMs	Wavelength	Slit	Lamp current	Drying	Ashing	Atomization	Background correction
Ni	232.0 nm	0.15 nm	4 mA	150°C, 20 s	1 050°C, 20 s	2 650°C, 4 s	Deuterium lamp or Zeeman
Pb	283.3 nm	0.2–1.0 nm	5–7 mA	120°C, 20 s	450°C, 15–20 s	1 700–2 300°C, 4–5 s	Deuterium lamp or Zeeman
Cd	228.8 nm	0.2–1.0 nm	2–10 mA	105°C, 20 s	400–700°C, 20–40 s	1 300–2 300°C, 3–5 s	Deuterium lamp or Zeeman
Cr	357.9 nm	0.2 nm	5–7 mA	85–120°C, 40–50 s	900°C, 20–30 s	2 700°C, 4–5 s	Deuterium lamp or Zeeman

detection (LOD) were 5, 0.1, 2 and 2.8 μg kg^{-1}, for Pb, Cd, Cr and Ni, respectively. Analytical reagent blanks, and a plant standard reference material [GBW10052 (GSB-30)] were prepared with each batch (20 samples) of digestion. They were then analyzed for the quality control of laboratory analyses. The results of the analyses were only accepted when the measured concentrations in the reference material were within one standard deviation of the certified values. A value of 1/2 LOD was assigned to all results below the LOD (Huang *et al.* 2014).

2.3. Calculation of pollution levels

To assess the pollution level of a single heavy metal in each sample, the single factor indexes (SFI) were used (Li *et al.* 2013; Ren 2013; Shen *et al.* 2013). The SFI were calculated according to eq. (1). If the SFI is more than 1, the sample was considered being polluted by the heavy metal *n*; otherwise, the sample was considered not being polluted by the heavy metal *n*.

$$P_n=C_n/S_n \tag{1}$$

P_n, the SFI value of the heavy metal *n*.

C_n, the concentration of the heavy metal *n*, mg kg^{-1}.

S_n, the assessment standard of the heavy metal *n*, mg kg^{-1} (Table 4).

Table 4 The assessment standards of the HMs for single factor index calculation

HMs	Assessment standards (mg kg^{-1})	References
Pb	0.1 for apple, pear and peach; 0.2 for grape and jujube	Nie and Dong 2014
Cd	0.05	Nie and Dong 2014
Cr	0.5	Li *et al*. 2012
Ni	0.3	Li *et al*. 2012

Table 5 Grading of combined pollution level of HMs

Grade	IPI	Combined pollution level
1	IPI≤0.7	Safe
2	0.7<IPI≤1	Warning
3	1<IPI≤2	Light
4	2<IPI≤3	Moderate
5	IPI>3	Heavy

IPI, integrated pollution indexes.

To assess the combined pollution of heavy metals in each sample, the integrated pollution indexes (IPI) were used (Li *et al.* 2013; Ren 2013; Shen *et al.* 2013). The IPI were calculated according to eq. (2), and graded according to Table 5.

$$P_{int}=\sqrt{(P_{max}{}^2+P_{ave}{}^2)/2} \tag{2}$$

P_{int}, the IPI value of heavy metals in a single sample.

P_{max}, the maximum level of heavy metal SFI in a single sample.

P_{ave}, the average level of heavy metal SFI in a single sample.

2.4. Calculation of daily intake rates

The DIR of heavy metals (mg kg^{-1} d^{-1}) were calculated according to eq. (3) (Singh *et al.* 2010; Cherfi *et al.* 2014; Li *et al.* 2014; Roba *et al.* 2016). The average adult body weight (B_W) was considered to be 63 kg (Jian *et al.* 2012). The ingestion rate of the fruits (I_R) was estimated by yield, exports, processing and storage loss (Nie *et al.* 2014, 2015; Li *et al.* 2015) (Table 6).

$$DIR=C\times I_R/B_W \tag{3}$$

C, the concentration of heavy metals in the fruits, mg kg^{-1}.

I_R, the ingestion rate of the fruits, kg d^{-1}.

B_W, the adult body weight, kg.

2.5. Calculation of target hazard quotient

To evaluate the potential hazardous exposure to heavy metals *via* consumption of the fruits by consumers, the target hazard quotients (THQ) (Cherfi *et al.* 2014; Roba *et al.* 2016) were calculated according to eq. (4). A THQ lower than 1 indicates that consumers may experience minor health effects; a total THQ (TTHQ) of two or more heavy metals greater than 1 indicates consumers are experiencing adverse health effects to some extent (Wang *et al.* 2005). The exposure duration (E_D) equals the life expectancy 74.8 years for adults in China (Shu *et al.* 2014). The oral reference dose (RfD) for Pb, Cd, Cr, and Ni are 0.0035, 0.001, 1.5, and 0.02 mg kg^{-1} d^{-1}, respectively (Khan *et al.* 2008; Mahmood and Malik 2013; USEPA 2015). The average exposure time (A_T) equals 365 d $yr^{-1}\times E_D$, and the exposure frequency (E_F) equals 365 d yr^{-1} (Roba *et al.* 2016).

Table 6 Ingestion rates of five fruits in China

Fruits	Yield ($\times10^4$ t)	Exports ($\times10^4$ t)	Processing ($\times10^4$ t)	Storage loss ($\times10^4$ t)	Ingestion rate (kg d^{-1})
Apple	3968.3	99.5	850.0	992.1	0.0456
Pear	1730.1	38.1	173.0	432.5	0.0229
Peach	1192.4	3.7	155.0	477.0	0.0125
Grape	1155.0	10.5	231.0	462.0	0.0110
Jujube	634.0	0.8	63.4	158.5	0.0086

$$THQ=\frac{E_F\times E_D\times I_R\times C}{RfD\times B_W\times A_T} \tag{4}$$

E_F, the exposure frequency, d yr^{-1}.

E_D, the exposure duration, yr.

I_R, the ingestion rate of the fruits, kg d^{-1}.

C, the concentration of heavy metals in the fruits, mg kg^{-1}.

RfD, the oral reference dose, mg kg^{-1} d^{-1}.

B_W, the adult body weight, kg.

A_T, the average exposure time, d.

3. Results

3.1. Heavy metal accumulation

The concentrations of heavy metals varied between different deciduous fruit species and between different heavy metals. Among the fruits studied, jujube and peach contained the highest concentrations of heavy metals, while grape contained the lowest concentrations (Table 7). Among the analyzed heavy metals, Ni was more likely to accumulate than the other three heavy metals, and Cr and Pb are more likely to accumulate than Cd. Overall, the concentration order of heavy metals in the fruits studied was Ni>Cr>Pb>Cd. The present study revealed that the concentrations of Ni in the fruits were significantly higher ($P<0.05$) compared to Cr, Pb, and Cd; while the concentrations of Cd in apple, peach and jujube were significantly lower ($P<0.05$) compared to Pb, Cr, and Ni; and there was no significant difference between the concentrations of Pb and Cr in peach, apple, and grape.

Fruits proved to be potent heavy metal accumulators, considering that for some investigated fruit species there were samples with heavy metal concentration higher than permitted (Roba *et al.* 2016). In 2.2% of the analyzed samples (including 1.4% of apple samples, 1.5% of pear samples, 7.3% of peach samples, and 0.7% of grape samples), the levels of Ni were higher than its maximum permissible limit (0.3 mg kg^{-1}) (Li *et al.* 2012). In 0.4% of the analyzed samples (including 0.5% of apple samples and 1.5% of peach samples), the levels of Pb were higher than its permissible limit (0.1 mg kg^{-1}) (Nie and Dong 2014). The concentrations of Cd and Cr in all analyzed samples were within their respective maximum permissible limits (0.05 and 0.5 mg kg^{-1}) (Li *et al.* 2012; Nie and Dong 2014). The concentration of heavy metals in the fruits also displayed significant differences between samples, as revealed by the high coefficient of variation (CV) values (which ranged from 62.8 to 185.7%) in Table 7.

Table 7 Concentration ranges of HMs in the fruits studied (mg kg^{-1})

Fruits	Level	Pb	Cd	Cr	Ni
Apple	Median	0.0184	0.0006	0.0192	0.0623
	Mean	0.0233 b	0.0021 c	0.0250 b	0.0766 a
	UCL	0.0256	0.0026	0.0268	0.0840
	SD	0.0193	0.0039	0.0157	0.0605
	CV (%)	82.8	185.7	62.8	79.0
Pear	Median	0.0051	0.0009	0.0143	0.0696
	Mean	0.0090 c	0.0026 c	0.0186 b	0.0861 a
	UCL	0.0103	0.0032	0.0207	0.0980
	SD	0.0100	0.0040	0.0168	0.0860
	CV (%)	111.1	153.8	90.3	99.9
Peach	Median	0.0190	0.0029	0.0321	0.0739
	Mean	0.0277 b	0.0037 c	0.0322 b	0.1056 a
	UCL	0.0319	0.0042	0.0354	0.1252
	SD	0.0275	0.0031	0.0204	0.1180
	CV (%)	99.3	83.8	63.4	111.7
Grape	Median	0.0050	0.0005	0.0112	0.0170
	Mean	0.0117 b	0.0013 c	0.0153 b	0.0375 a
	UCL	0.0141	0.0016	0.0182	0.0484
	SD	0.0152	0.0018	0.0175	0.0604
	CV (%)	129.9	138.5	114.4	161.1
Jujube	Median	0.0159	0.0013	0.0207	0.0985
	Mean	0.0246 c	0.0029 d	0.0414 b	0.1035 a
	UCL	0.0287	0.0037	0.0514	0.1162
	SD	0.0224	0.0041	0.0524	0.0711
	CV (%)	91.1	141.4	126.6	68.7

UCL, the upper limit of 95% confidence interval for the mean. SD, standard deviation. CV, coefficient of variation. Different small letters in the same line indicated significant differences ($P<0.05$). The same as below.

3.2. Heavy metal pollution

Table 8 presents the single factor indexes for heavy metals in the fruits studied. The trends of the single factor indexes for heavy metals in the analyzed fruits were in the order of Ni>Pb>Cr and Cd. We found that the single factor index for Ni was significantly higher ($P<0.05$) compared to Pb, Cr and Cd, and that the single factor index for Pb was significantly higher ($P<0.05$) compared to Cr and Cd, whereas there were no significant differences between the single factor indexes for Cr and Cd. Overall, the single factor indexes for heavy metals were generally far below 1; the indexes in peach were the highest, while those in grape were the lowest. We found that 2.2% of the samples (including 1.4% of apple samples, 1.5% of pear samples, 7.3% of peach samples, and 0.7% of grape samples) were polluted by Ni, and 0.4% of the samples (including 0.5% of apple samples and 1.5% of peach samples) were polluted by Pb, with single factor indexes beyond 1. Whereas the single factor indexes for Cd and Cr in the fruits studied were found all below 1, indicating no samples polluted by Cd or Cr.

The IPI for heavy metals in the fruits studied were given in Table 9. The heavy metal pollution in 96.9% of the samples were at safe level (IPI≤0.7), 2.32% of the samples were polluted at warning level (0.7<IPI≤1), 0.65% of the samples were polluted at light level (1<IPI ≤2), 0.13% of the samples were polluted at moderate level (2<IPI≤3), and no samples were polluted at heavy level (IPI>3). We found that the integrated pollution indexes for heavy metals in peach were significantly higher ($P<0.05$) than those in the other four fruits (jujube, apple, pear, and grape), while those in grape were significantly lower ($P<0.05$) compared to those in the other four fruits (peach, jujube, apple, and pear). Overall, the pollution level of heavy metals in peach were higher than levels in other fruits, and 7.30, 1.46, and 0.73% of peach samples were polluted at warning level, light level,

Table 8 The single factor indexes of the analyzed HMs in the fruits studied

Fruits	Level	Pb	Cd	Cr	Ni
Apple	Median	0.184	0.012	0.038	0.208
	Mean	0.233 a	0.042 b	0.050 b	0.256 a
	UCL	0.256	0.052	0.054	0.280
	SD	0.193	0.078	0.031	0.202
	CV (%)	82.8	185.7	62.0	78.9
Pear	Median	0.051	0.018	0.029	0.232
	Mean	0.090 b	0.052 c	0.037 c	0.287 a
	UCL	0.103	0.063	0.041	0.327
	SD	0.100	0.080	0.034	0.287
	CV (%)	111.1	153.8	91.9	100.0
Peach	Median	0.190	0.058	0.064	0.246
	Mean	0.277 b	0.074 c	0.064 c	0.352 a
	UCL	0.319	0.084	0.071	0.417
	SD	0.275	0.062	0.041	0.393
	CV (%)	99.3	83.8	64.1	111.6
Grape	Median	0.025	0.010	0.022	0.057
	Mean	0.059 b	0.025 c	0.031 c	0.125 a
	UCL	0.071	0.031	0.036	0.161
	SD	0.076	0.036	0.035	0.202
	CV (%)	128.8	144.0	112.9	161.6
Jujube	Median	0.080	0.025	0.041	0.328
	Mean	0.123 b	0.059 c	0.083 bc	0.345 a
	UCL	0.144	0.074	0.103	0.387
	SD	0.112	0.081	0.105	0.237
	CV (%)	91.1	137.3	126.5	68.7

Table 9 Sample proportions with different levels of integrated pollution index

Fruits	Median	UCL	Mean	SD	CV (%)	IPI≤0.7	0.7<IPI≤1	1.0<IPI≤2	2<IPI≤3	IPI>3
Peach	0.281	0.396	0.348 a	0.298	85.6	90.51%	7.30%	1.46%	0.73%	0.00%
Jujube	0.265	0.321	0.292 b	0.157	53.8	96.81%	3.19%	0.00%	0.00%	0.00%
Apple	0.241	0.289	0.270 bc	0.157	58.1	98.11%	1.89%	0.00%	0.00%	0.00%
Pear	0.188	0.256	0.227 c	0.211	93.0	98.46%	0.51%	1.03%	0.00%	0.00%
Grape	0.061	0.139	0.112 d	0.149	133.0	99.27%	0.00%	0.73%	0.00%	0.00%

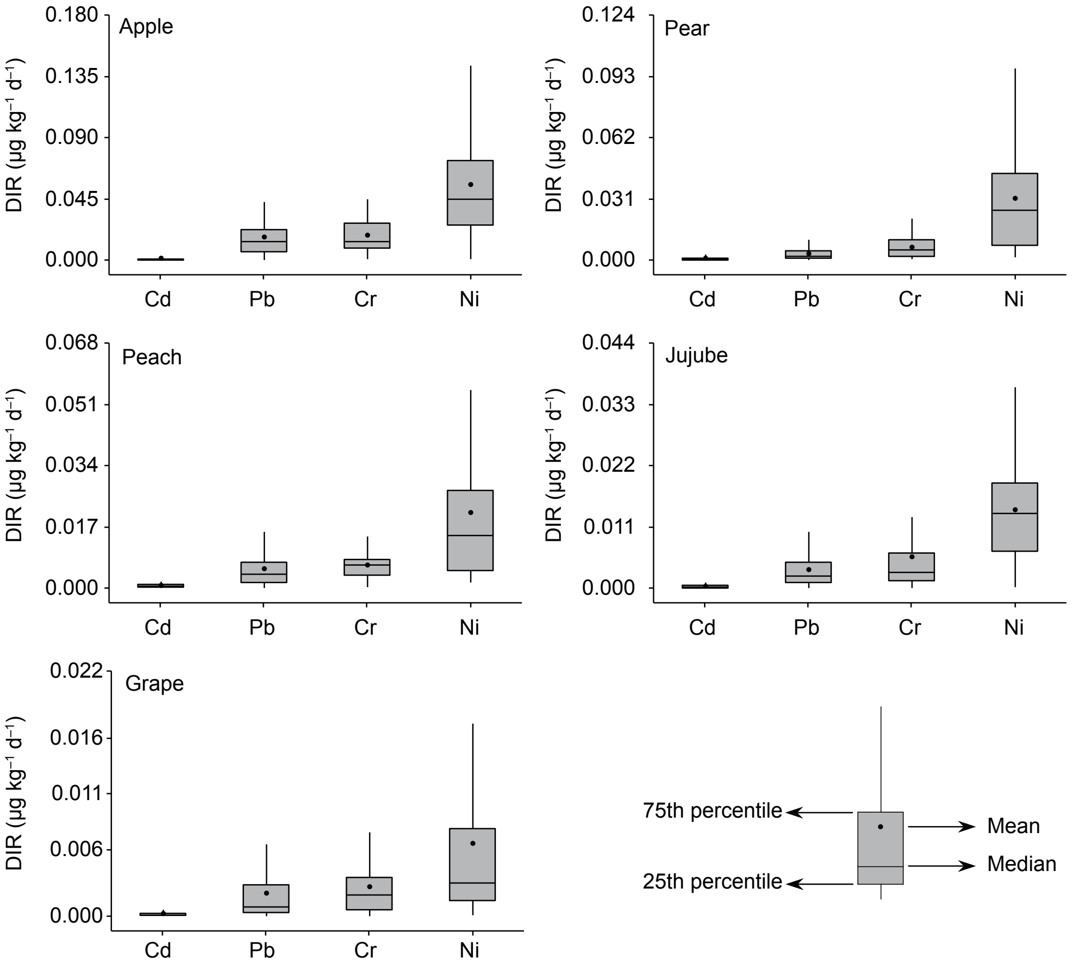

Fig. 2 The estimated daily intake rates (DIR) of heavy metals caused by consumption of the fruits studied.

and moderate level, respectively. The integrated pollution indexes for heavy metals in peach were significantly higher ($P<0.05$) than in other fruits, while those in grape were significantly lower ($P<0.05$) compared to other fruits, and there were no significant difference in integrated pollution indexes for heavy metals between jujube and apple, or between apple and pear.

3.3. Health risk assessment

Fig. 2 displays the estimated daily intake rates (DIR) of heavy metals caused by consumption of the fruits studied. The contribution of heavy metals to DIR followed the order of Ni>Cr>Pb>Cd. The highest DIR of heavy metals were caused by consumption of apple, while the lowest DIR were caused by consumption of grape. Generally, the total daily intake rates of Pb, Ni, Cd, and Cr caused by consumption of the fruits studied corresponded to 0.8165–1.0826%, 0.4963–0.6622%, 0.2971–0.4680% and 0.0023–0.0030%, respectively, of the tolerable daily intakes for Pb (0.0035 mg kg^{-1} d^{-1}), Ni (0.02 mg kg^{-1} d^{-1}), Cd (0.001 mg kg^{-1} d^{-1}), and Cr (1.5 mg kg^{-1} d^{-1}) (Khan *et al.* 2008; Mahmood and Malik 2013; USEPA 2015). Consequently, no significant adverse effects on consumer health are expected from the ingestion of these heavy metals *via* consumption of the fruits studied.

To assess the health risk associated with heavy metal pollution of the fruits studied, we estimated the THQ (Fig. 3). The THQ of heavy metals followed the order of Ni>Pb>Cd>Cr in pear, and the order of Pb>Ni>Cd>Cr in the other four fruits. Among the analyzed heavy metals in these fruits, Cr had the lowest potential health risk, which may be ascribed to its higher RfD (Li *et al.* 2012; Cherfi *et al.* 2014). The TTHQ caused by consumption of these fruits was in the order of apple (0.0084–0.098)>peach (0.0029–0.0038) and pear (0.0030–0.0039)>jujube (0.0018–0.0023)>grape (0.0009–0.0013). The THQ and TTHQ values we obtained demonstrated that none of the analyzed heavy metals may pose risk to consumers through consumption of these fruits. The THQ for all of the analyzed heavy metals and the TTHQ for all of the studied fruits were far lower than 1, suggesting

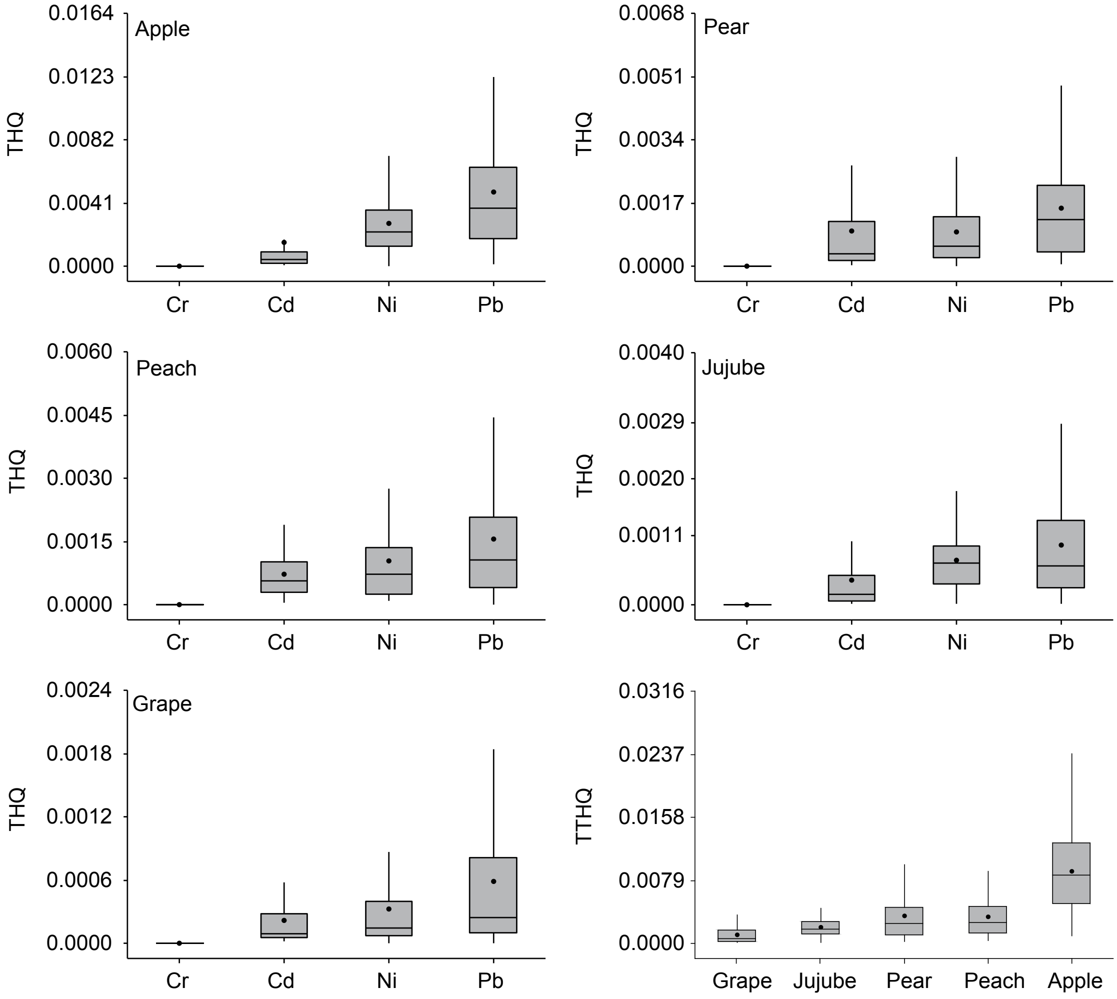

Fig. 3 The estimated target hazard quotients (THQ) of heavy metals caused by consumption of the fruits studied.

that consumers may experience little health effect (Wang *et al.* 2005).

4. Discussion

In the present study, almost all concentrations of the analyzed heavy metals in the fruits studied were within the maximum permissible limits, and several or dozens of times lower than those reported by Zhen (2008), Xiao *et al.* (2010) and Sheng *et al.* (2014) in China, Orisakwe *et al.* (2012) in Nigeria, Türkdoğan *et al.* (2002) in Turkey, Roba *et al.* (2016) in Romania and Khan *et al.* (2013) in Pakistan. Almost all the SFIs, the IPIs, the DIRs, and the THQs were accordingly far lower than those reported by above researchers, especially than those reported sparsely in China. Compared to that in vegetables (Zhang *et al.* 2010; Cherfi *et al.* 2014), the accumulation of heavy metals in the fruits studied was low, probably because a large proportion of heavy metals absorbed by trees were stored in other organs, especially leaves (Roba *et al.* 2016).

The concentrations of heavy metals varied between different fruits (Table 7) because of their different absorption capacity and the regional soil and atmospheric degree of pollution (Roba *et al.* 2016). Apart from the fruit species, other factors may also affect the accumulation of heavy metals. Previous studies demonstrated that the higher the concentration of heavy metals in soil, the higher will be its probability in crops (Mapanda *et al.* 2007). Uptake of Pb is regulated by pH, particle size and cation exchange capacity of soil, as well as by root exudation and other physio-chemical parameters (Lokeshwari and Chandrappa 2006). The high levels of Pb in the fruits might be a consequence of roads traffic and lead emission from petrol (Zhen 2008; Cherfi *et al.* 2014). As for Cd, apart from the industrial pollution (Xiao *et al.* 2010; Khillare *et al.* 2012), phosphoric fertilizer is the main source of Cd in agricultural soils (Demirezen and Aksoy 2006).

To assess the human health risks of heavy metal pollution, it is essential to estimate the level of exposure by quantifying the exposure routes of a pollutant to the target organisms. There are various exposure pathways of pollutants to human, such as food chain, dermal contact and inhalation (Khan *et al.* 2013). Compared to oral intake, all other pathways are negligible (Khan *et al.* 2008). Food consumption is identified as the major pathway for human exposure to environmental contaminants, accounting for >90% of intake as compared to inhalation or dermal routes (Ferré-Huguet *et al.* 2008; Martí-Cid *et al.* 2008a, b; Martorell *et al.* 2011). Therefore, to evaluate potential human health risks, both DIR and THQ of heavy metals were calculated and summarized. THQ equals the multiplication of health risk indexes (HRI) with E_D (the exposure duration, equals the life expectancy for adults). So HRI were also often used to estimate the health risk of heavy metals through food consumption (Khan *et al.* 2008, 2013; Singh *et al.* 2010; Li *et al.* 2012; Mahmood and Malik 2013).

In China, few published data on heavy metal contamination in fruits are available, and literatures did not focus on main producing areas but on some special regions or sites. Zhen (2008) reported that fruits at sides of Shenyang-Dalian expressway were polluted by Pb and Cd with the mean concentrations of 0.082 and 0.010 mg kg^{-1} in apple (3.5 and 4.8 times as our study), and 0.102 and 0.013 mg kg^{-1} in grape (8.7 and 10 times as our study). Soil in orchards around Molybdenum mining area in Huludao was seriously polluted by heavy metals (Pb, Cd, and Cr), as led to heavy metal pollution to fruits with concentrations of 6.7–28.5 times in apple, and 17.9–25.3 times in pear than our study (Xiao *et al.* 2010). Survey (Sheng *et al.* 2014) indicated that the mean content of heavy metals (Pb, Cd, and Cr) in fruits (apple, pear, and peach) marketed in Bengbu City of China were 3.4–22.7 times (except Pb in apple) as our study, and the health risk of heavy metals from these fruits was mainly caused by Cr. Survey was also conducted in fruits (apple, pear, and grape) marketed in Xuzhou City of China (Sun *et al.* 2009), and the results showed that the mean heavy metal content were 2.9–16.2 times as our study, the pollution degree of grape exceeded the guard line, and more attention should be paid to Cd.

Study in apple and pear in Swat District of northern Pakistan (Khan *et al.* 2013) showed that the concentrations of Cd and Cr were 38.1 and 9.6 times (apple), and 34.6 and 12.4 times (pear) as our study, especially Cd was found higher in concentration than the limit (0.05 mg kg^{-1}) in 95% samples, and would pose a high level potential health risk to the consumers. It is therefore suggested that fruits from the contaminated locations should not be consumed without proper treatment. Fruits cultivated in mining areas were prone to be contaminated with heavy metals (Xiao *et al.* 2010; Roba *et al.* 2016). In Baia Mare mining area (Romania) strongly contaminated by heavy metals (especially Pb and Cd), except Cd in apple, the concentrations of Pb and Cd in fruits sampled in rural areas were over 33 times (Pb) and 6 times (Cd) higher (Roba *et al.* 2016) than those in our study.

5. Conclusion

The concentrations of Pb, Cd, Cr and Ni in most (97.5%) of the analyzed deciduous fruit samples were within the maximum permissible limits. Heavy metal concentrations varied among the fruits, they were the highest in jujube and peach, and the lowest in grape. The magnitude of heavy metals in all fruits was Ni>Cr>Pb>Cd. Compared to other fruits, the combined pollution of heavy metals was significantly higher in peach and significantly lower in grape. And there was no significant difference in combined pollution of heavy metals between jujube and apple, or apple and pear. In most (96.9%) of the analyzed samples, the heavy metal pollution was at safe level (IPI≤0.7). By calculating the daily intake rates (DIR) and the target hazard quotients (THQ), we concluded that consumption of the fruits studied posed little potential health risk. We therefore suggest that the main deciduous fruits (apple, pear, peach, grape, and jujube) of China's main producing areas are safe to eat.

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