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Trace Elements in the Environment

Lead smelting alters wheat flour heavy metal concentrations and health risks

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Abstract

Wheat (*Triticum aestivum* L.) flour consumption may be a major source of human metal intake, especially when wheat is cultivated in metal-contaminated soils. This work investigated Cd, Cu, Pb, and Zn distribution in whole wheat flour, wheat flour, and wheat bran when grown in an area polluted by Pb smelting. Wheat product heavy metal concentrations were analyzed, and the (non)carcinogenic risks were assessed. Mean Cd, Cu, Pb, and Zn concentrations in whole wheat flour were 0.38, 3.83, 0.48, and 29.3 mg kg^{-1}, respectively; those in flour were only slightly reduced. The ratios between noncarcinogenic average daily dose of whole wheat flour and wheat flour consumption ranged from 1.06 to 3.76, with Pb having the greatest values compared with other metals. For children, the average hazard quotients (HQs) of whole wheat flour consumption of Cd, Cu, Pb, and Zn were 4.19, 1.06, 1.53, and 1.07; those for wheat flour consumption were 3.81, 0.68, 0.70, and 0.98, respectively. The HQs of adults were less than those of children. Overall results indicated that consumption of wheat products may lead to health concerns in the heavy metal contaminated area, yet when wheat flour rather than whole wheat flour is consumed, only the human health risk from Pb ingestion is reduced. Altering or removing human edible crops in the most contaminated areas should be considered.

1 | INTRODUCTION

Wheat (*Triticum aestivum* L.) is a worldwide staple food crop, with ∼20% of human dietary caloric and protein consumption coming from wheat (Liu, Liu, et al., 2020; Senapati & Semenov, 2020; Wu et al., 2020). Wheat consumption by humans is also important for the intake of essential micronutrients (Liu, Liu, et al., 2020; White & Broadley, 2009). In developing countries, it is believed that 20% of human Zn intake is from wheat (Liu, Liu, et al., 2020). Furthermore, micronutrients in wheat grain are unevenly distributed, with greater Cu, Fe, and Zn concentrations often found in the bran (i.e., pericarp) of the wheat kernel (Cardoso et al., 2018; Li et al., 2011; Liu et al., 2007; Yan et al., 2020). Calculated from the results of Li et al. (2017), concentrations of Zn, Fe, and Cu in wheat bran were 2.60–3.22, 3.60–5.73, and 1.21–2.70 times of those of the endosperm, respectively. Cardoso et al. (2018) separated wheat grain into embryo and endosperm, finding that the mean Fe and Zn concentrations in embryo were 6.92 and 8.29 times of those in endosperm, the latter of which is the main component of wheat flour.

Unfortunately, the uneven micronutrient distribution in wheat grain also occurs with nonessential, potentially toxic

Abbreviations: ADD, average daily dose; EF, exposure frequency; HI, hazard index; HQ, hazard quotient; MPC, maximum permissible concentration; PI, pollution index.

trace elements such as As, Cd, and Pb. Giordano and Blandino (2018) found that As, Cd, and Pb concentrations in wheat bran were greater than the endosperm, with the greatest concentration differences found for Pb. Cubadda et al. (2005) showed that pearling (i.e., abrasively removing the bran) of wheat grain by only 30% resulted in a 32–68% decrease in (non)essential microelements. Thus, the consumption of whole wheat flour may result in greater human intake of both essential and potentially harmful trace elements.

Human ingestion of potentially harmful elements may be exacerbated when food crops such as wheat are grown in soils contaminated by heavy metals. In support of this contention, numerous researchers have proven that wheat grain can accumulate heavy metals to unsafe concentrations for human consumption (Li et al., 2020b; Liu et al., 2017; Liu, Cui, et al., 2020; Wu et al., 2020; Xing et al., 2016). Long-term consumption of whole wheat flour from areas where heavy metals are of concern may result in greater-than-tolerable heavy metal accumulation in human tissues (Bermudez et al., 2011; Wang et al., 2020).

In recent years, the hazard quotient (HQ), which is a comparison of the maximum tolerable intake value of heavy metals by humans to the observed intake value, has been widely used to assess the health risk of heavy metals in soil, dust, and food. Chen et al. (2020) investigated the effect of P rates on the heavy metal accumulation of wheat grains and the resulting health risk in North China. Wang et al. (2020) investigated the absorption of heavy metals from wheat in different parts of human digestion systems. Zhang et al. (2018) studied the effect of different N fertilizer treatments on wheat grain heavy metal accumulation and its effect on human health. Unfortunately, given the uneven distribution of heavy metals in wheat grain, the consumption of whole wheat flour versus wheat flour will quite possibly result in different risks of heavy metal accumulation in the human body and subsequently health risks. This contention was not addressed by the above authors and has not been adequately addressed in the literature.

The objective this of study was to investigate the differences between heavy metal concentrations within whole wheat flour and flour (i.e., with the bran removed) when the crop was raised in a heavy metal contaminated area affected by a Pb smelter. Based on the collected data, human health risks were estimated for consumption of either wheat flour or whole wheat flour.

Core Ideas

- Greater heavy metal concentrations were found in bran than in wheat flour.
- Pearling reduced the Pb risk the most compared with Cd, Cu, and Zn.
- Hazard quotients of wheat flour were lower than whole wheat flour.
- Cd hazard quotients of whole wheat flour and wheat flour exceeded safe levels.

2 | MATERIALS AND METHODS

2.1 | Study area and sample collection

Soil and wheat were collected from an area affected by Pb smelting in northern China. Detailed study area conditions and soil and wheat grain sample collection information can be found in Li et al. (2020b). The study area was affected by a Pb smelter that produces about 400,000 t of Pb annually, with farmland in this area mainly under a winter wheat–summer corn (*Zea mays* L.) rotation. Thirteen pairs of soil and wheat grain samples were collected from fields within three villages, namely QD, GF, and BS (i.e., $n = 13$ at each village). The distances between QD, GF, and BS and the nearby Pb smelting facility are about 1, 3, and 6 km, respectively.

Soil samples were collected from the 0-to-20-cm depth with a core drill, air-dried, and ground to pass a 0.149-mm sieve. Wheat grain samples were washed three times with tap water followed by three times with deionized water then oven-dried at 75 ˚C for 48 h. A portion of each wheat grain sample was then pearled into wheat bran and nonbran parts with a laboratory mill (JNMJ3, Taizhou Grain Instrument Co.). The average bran percentage was 5.65 ± 0.98%. Earlier work found that the first 3% of the pearled wheat was mainly wheat pericarp and that at 6% the endosperm was pearled (De Brier et al., 2015). Wheat grain, bran, and nonbran parts were ground to a <0.1-mm powder for chemical analysis. The samples were designated as either whole wheat flour, bran, or wheat flour.

2.2 | Chemical analysis

Soil samples were digested with H_2O_2 (30%)–HF (40%)–HNO_3 (68%)–$HClO_4$ (70%) on a hotplate for total heavy metal concentration analysis; wheat samples were digested with HNO_3–$HClO_4$ (5:1, v/v) on a hotplate, both as described in Lu (2000). All samples were digested in triplicate. Concentrations of Cd, Cu, Pb, and Zn in soil digestates and Cu and Zn in wheat digestates were determined using atomic absorption spectrometry (TAS-990, PGeneral). Concentrations of Cd and Pb in wheat digestates were determined by inductively coupled plasma–optical emission spectroscopy (iCAP 7200, Thermo Fisher).

2.3 | Quality assurance and quality control

All glassware was soaked in a 5% nitric acid solution for more than 4 h and then triple-rinsed with deionized water before use. Blank samples, duplicate samples, and standard reference materials (GBW08303 for soil and GBW10035 for wheat) were used during the analytical process for quality control. Reference material recovery of Cd, Cu, Pb, and Zn in soil ranged from 98.9 to 102.4%, and those of wheat ranged from 97.4 to 106.5%.

2.4 | Data analysis

2.4.1 | Exposure assessment

According to the *Exposure Factors Handbook* (USEPA, 2018), the average daily dose (ADD) (mg kg^{-1} d^{-1}) of an element via food ingestion was estimated using Equation 1 for each whole wheat flour and wheat flour sample:

$$\mathrm{ADD} = \frac{C \times \mathrm{IngR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \tag{1}$$

where C is the concentration of the element in wheat product (mg kg^{-1}), IngR is the ingestion rate (kg d^{-1}), EF is the exposure frequency (d yr^{-1}), ED is the exposure duration (yr; which was set at 70 yr), BW is body weight (kg), and AT is the average time (d; which was set as EF×10 and EF×30 for noncancer risk analysis for children and adults, respectively, and 25,500 d for cancer risk analysis). Values of all parameters are shown in Supplemental Table S1.

2.4.2 | Risk calculation

A HQ indicating the noncarcinogenic risk during a lifetime can be calculated by dividing the ADD by a specific reference dose. The HQ for each element was calculated using Equation (2) (The University of Tennessee Knoxville & Oak Ridge National Laboratory, 2020) for each whole wheat flour and wheat flour sample,

$$\mathrm{HQ} = \frac{\mathrm{ADD}}{\mathrm{RfD}} \tag{2}$$

where RfD is the estimated maximum permissible risk to humans through daily ingestion exposure (mg kg^{-1} d^{-1}). Experiencing adverse health effects is unlikely when the HQ is ≤1, whereas there may be concern for potential noncarcinogenic effects when the HQ is >1 (The University of Tennessee Knoxville & Oak Ridge National Laboratory, 2020). The hazard index (HI) is the sum of HQs of different elements.

The pollution index (PI) was calculated as the ratio between the heavy metal concentration in a sample and the maximum permissible concentration (MPC) of this element in the food quality standard (National Health Commission of China & China Food & Drug Administration, 2017) for wheat fractions. The PI was also determined for soil heavy metal concentrations as compared to background soil heavy metal concentrations presented in Cheng et al. (2014).

2.4.3 | Statistical analysis

Data analysis was conducted using SPSS for Windows 25.0 (SPSS Inc.). Analysis of variance was conducted with the post hoc Tukey's test method for samples between different villages or different metals, and a paired sample t-test was conducted for individual metal carcinogenic or noncarcinogenic risks between whole wheat flour and wheat flour consumption, or between children and adults for the consumption of the same wheat fractions. Statistical differences were determined using a $p < .05$. Data plotting and graphical presentation were performed using Origin 9.0 software (Origin Lab Corp.).

3 | RESULTS

3.1 | Soil heavy metal concentrations

Soil heavy metal concentrations from the three villages are presented in Supplemental Table S2. The concentration ranges of Cd, Cu, Pb, and Zn in soil were 0.45–9.90, 26.1–70.5, 107–642, and 73.8–215 mg kg^{-1}, respectively. Results indicate significant Cd and Pb accumulation and, to a lesser extent, Cu and Zn accumulation in the study area. As the distance from the smelter increased, the concentrations of these metals decreased.

3.2 | Heavy metal concentrations in the whole wheat flour, wheat bran, and flour

Wheat sample heavy metal concentrations are shown in Table 1. Differences existed among heavy metal concentrations of different grain fractions. For all four metals from these three sites, the mean concentrations ranked bran > whole wheat flour > wheat flour, with bran heavy metal concentrations significantly greater than those of whole wheat flour and wheat flour ($p < .05$; Table 1).

In most cases, the mean concentrations of the same heavy metal in the same kind of wheat samples between these three villages ranked QD > GF > BS, except for bran Pb concentrations, which followed the order BS > GF > QD. Cadmium and Pb concentrations of all three wheat grain fractions from QD

TABLE 1 Mean heavy metal concentrations in wheat grain, bran and flour samples collected from three villages of QD, GF, and GS near a large Pb smelter in northern China

Village	Value[a]	Cd			Cu			Pb			Zn		
		WWF[b]	Bran	Flour	WWF	Bran	Flour	WWF	Bran	Flour	WWF	Bran	Flour
QD (n = 13)	Mean	0.674Ab	1.30Aa	0.636Ab	5.18Ab	40.4Aa	3.14Ab	0.645Ab	4.50Aa	0.411Ab	36.9Ab	103Aa	33.0Ab
	SD	0.369	0.615	0.36	1.27	12.9	1.24	0.089	1.40	0.136	11.4	31.0	10.3
	PI[a]	6.74		6.36				3.22		2.06			
GF (n = 13)	Mean	0.277Bb	0.901Ba	0.239Bb	3.45Bb	24.7Ba	2.25Ab	0.385Bb	4.82ABa	0.137Bb	28.0Bb	57.6Ba	26.2ABb
	SD	0.065	0.210	0.070	1.42	6.09	1.30	0.130	1.17	0.104	5.57	13.0	5.56
	PI	2.77		2.39				1.92		0.687			
BS (n = 13)	Mean	0.188Bb	0.698Ba	0.159Bb	2.87Bb	17.4Ba	2.04Ab	0.419Bb	5.79Ba	0.112Bc	22.8Bb	60.0Ba	20.7Bb
	SD	0.045	0.214	0.024	0.818	2.04	0.921	0.104	0.422	0.110	7.12	9.80	7.37
	PI	1.88		1.59				2.09		0.558			
	MPC[†]	0.1		0.1				0.2		0.2			

Note. Different uppercase letters signify significant differences ($p < .05$) between concentrations of the same metal in the same wheat fraction across villages. Different lowercase letters signify significant difference ($p < .05$) between concentrations of the same metal in different wheat fractions from the same village (based on ANOVA). The distances between these villages and the nearby Pb smelter were 1, 3, and 6 km, respectively.

[a]MPC, maximum permissible concentration (from National Health Commission of China and China Food and Drug Administration, 2017); PI, pollution index.

[b]Whole wheat flour.

were greater than those from GF and BS ($p < .05$), whereas no significant difference was found between GF and BS samples ($p > .05$). For Cu and Zn concentrations, significant differences were only found for bran and whole wheat flour samples between samples from QD as compared to GF and BS ($p < .05$).

The Cd concentrations of all samples from all locations were all greater than the MPC of 0.1 mg kg^{-1}, whereas flour Pb concentrations from the GF and BS sites were lower than the MPC of 0.2 mg kg^{-1} (National Health Commission of China & China Food & Drug Administration, 2017). The Pb concentrations of all wheat samples from nearest the Pb smelter (QD), and most wheat samples from the GF and BS sites, were greater than the MPC value. Compared with the Cd and Pb MPCs, 100 and 95% of all the whole wheat flour samples had Cd and Pb concentrations greater than their MPCs, whereas 100 and 36% of the wheat flour samples had Cd and Pb concentrations greater than their MPCs.

Concentration ratios of Cd, Cu, Pb, and Zn between different wheat grain fractions are presented in Supplemental Table S3. Results indicate that Cu and Pb had greater whole wheat flour/wheat flour ratios at all three sites as compared to the Cd and Zn ratios. The Cd and Zn whole wheat flour/wheat flour ratios were close to 1. The bran/wheat flour and bran/whole wheat flour ratios were generally greater than the whole wheat flour/wheat flour ratios for all metals. The bran/wheat flour or bran/whole wheat flour Pb ratios were greater than 7 and in some instances were as great as 50. Overall, wheat grain had unsafe Pb and Cd concentrations; pearling reduced heavy metal concentrations, especially that of Pb; and bran had greater heavy metal concentrations than wheat flour.

3.3 | Correlations between soil and wheat heavy metal concentrations

Significant linear correlations were found between all three wheat products for all four metals and soil heavy metal concentrations ($p < .05$), except between bran Pb and soil Pb concentrations (Figure 1). All metals accumulated to the greatest degree in the bran. The greatest r values were found for whole wheat flour Cd, Cu, and Pb and bran Zn.

3.4 | Risk assessment

The ADDs of heavy metals by adults and children at these three villages are presented in Table 2. It is obvious that if consumption of wheat harvested from QD (1 km from the Pb smelter) occurs, this would lead to the greatest heavy metal ADD among all three sites. In most cases, consumption of wheat harvested at GF (3 km from the Pb smelter) also has greater heavy metal intake than wheat from BS (6 km from the Pb smelter).

Consumption of whole wheat flour would result in more heavy metal intake than consumption of wheat flour. The ratios between ADD of whole wheat flour to wheat flour ranged between 1.06 (Cd; QD Village) to 3.76 (Pb; BS Village); the average ratios over all three villages for Cd, Cu, Pb, and Zn also indicate that pearling decreased the intake of Pb and Cu more than Cd and Zn. Finally, ADD values in children were greater than in adults.

The HQ and HI values are presented in Figure 2. The average HQ of each heavy metal of all samples at these three villages were calculated. For adults, the average whole wheat flour consumption HQs of Cd, Cu, Pb, and Zn were 3.16, 0.80,

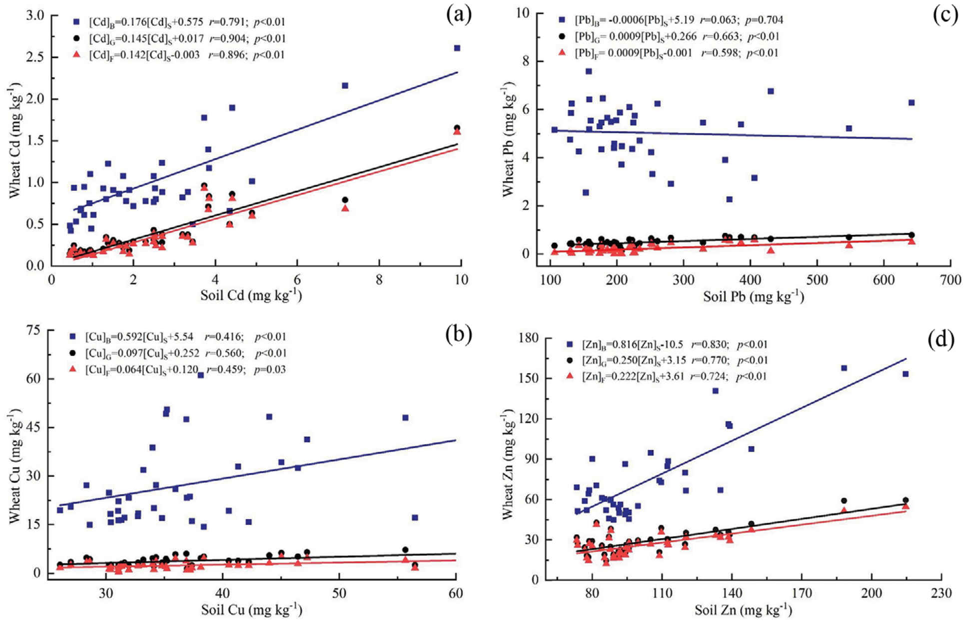

FIGURE 1 Correlations between whole wheat flour, wheat bran, wheat flour, and total soil (a) Cd, (b) Cu, (c) Pb, and (d) Zn concentrations near a large Pb smelter in northern China. $[\text{Metal}]_B$, $[\text{Metal}]_G$, $[\text{Metal}]_F$, and $[\text{Metal}]_S$ denote the heavy metal concentrations of wheat bran, whole wheat flour, wheat flour, and soil, respectively ($n = 39$)

TABLE 2 Average daily dose for noncarcinogenic risk of Cd, Cu, Pb, and Zn by consumption of whole wheat flour and wheat flour in the villages of QD, GF, and BS

Element	Sample sites	Whole wheat flour		Wheat flour		Whole wheat flour/wheat flour	
		Children	Adults	Children	Adults	Ratio	Mean
		mg kg^{-1} d^{-1}					
Cd	QD	7.45×10^{-3}	5.61×10^{-3}	7.04×10^{-3}	5.30×10^{-3}	1.06	1.13
	GF	3.06×10^{-3}	2.31×10^{-3}	2.64×10^{-3}	1.99×10^{-3}	1.16	
	BS	2.08×10^{-3}	1.57×10^{-3}	1.76×10^{-3}	1.32×10^{-3}	1.18	
Cu	QD	5.74×10^{-2}	4.32×10^{-2}	3.47×10^{-2}	2.61×10^{-2}	1.65	1.53
	GF	3.81×10^{-2}	2.87×10^{-2}	2.41×10^{-2}	1.81×10^{-2}	1.53	
	BS	3.17×10^{-2}	2.39×10^{-2}	2.25×10^{-2}	1.70×10^{-2}	1.41	
Pb	QD	7.13×10^{-3}	5.37×10^{-3}	4.55×10^{-3}	3.43×10^{-3}	1.57	2.71
	GF	4.26×10^{-3}	3.21×10^{-3}	1.52×10^{-3}	1.14×10^{-3}	2.80	
	BS	4.63×10^{-3}	3.49×10^{-3}	1.23×10^{-3}	9.30×10^{-4}	3.76	
Zn	QD	4.09×10^{-1}	3.08×10^{-1}	3.65×10^{-1}	2.75×10^{-1}	1.12	1.10
	GF	3.10×10^{-1}	2.33×10^{-1}	2.90×10^{-1}	2.19×10^{-1}	1.07	
	BS	2.52×10^{-1}	1.90×10^{-1}	2.29×10^{-1}	1.72×10^{-1}	1.10	

Note. The distances between these villages and the nearby Pb smelter were 1, 3, and 6 km, respectively.

1.15, and 0.81, respectively; the average wheat flour consumption HQs were 2.87, 0.52, 0.52, and 0.74, respectively. For children, the whole wheat flour consumption HQs of Cd, Cu, Pb, and Zn were 4.19, 1.06, 1.53, and 1.07, respectively, and the average wheat flour consumption HQs were 3.81, 0.68, 0.70, and 0.98, respectively. The HQ results clearly indicate that the greatest noncarcinogenic risk of consuming wheat in the study area is from the intake of Cd, with the greatest Cd HQ present via the consumption of whole wheat flour by children in the QD village (see statistical results in Supplemental Table S4).

For all three villages, the heavy metal HQs for consumption of either whole wheat flour or wheat flour were in the order of QD > GF > BS except Pb, indicating a more likely effect on human health of heavy metal intake by local residents as distance to the smelter decreases (Figure 2). For whole wheat

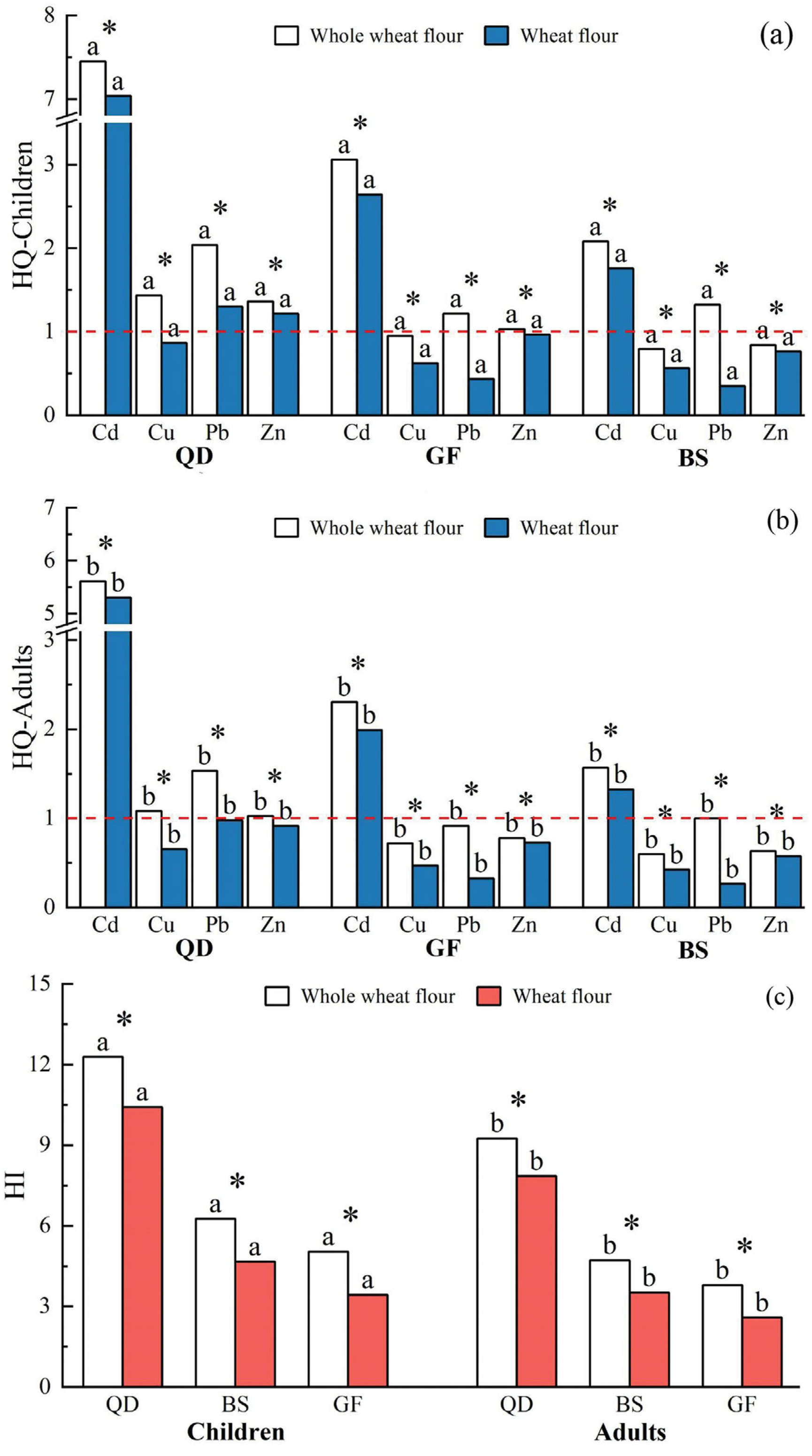

FIGURE 2 Hazard quotient (HQ) of (a) children and (b) adults and (c) hazard index (HI) (sum of HQ) of children and adults for consumption of wheat harvested in the villages of QD, BS and GF near a large Pb smelter in China. Different lowercase letters in (a) and (b) signify significant HQ differences ($p < .05$) between children and adult consumption of the same metal within the same wheat fraction in the same village. Different lowercase letters in (c) signify significant HI differences ($p < .05$) between children and adults for consumption of same wheat fraction in the same villages. The distances between QD, GF, and BS villages and the nearby Pb smelter were 1, 3, and 6 km, respectively. *Significant differences ($p < .05$) between whole wheat flour and wheat flour consumption in the same village for either children or adults. See Supplemental Table S3 for significant HI differences between different villages

flour consumption by children, health risks existed for Cd and Pb at all three villages (Figure 2a). At the QD village, children's health issues may also exist for Cu and Zn intake. Compared with children, adults had a less noncarcinogenic health risk (Figure 2b).

Pearling reduced the noncarcinogenic risk of wheat consumption, with Pb being reduced to the greatest extent among all four metals (Figure 2). Mean Cd, Cu, Pb, and Zn reduction rates were 15.6, 34.4, 53.7, and 8.76%, respectively. Regardless of pearling, the noncarcinogenic risk of Cd existed for both children and adults at all three villages; the risk of Pb and Zn still existed for children after pearling at the QD village. The HQ for wheat flour consumption was significantly lower than whole wheat flour consumption for both children and adults for all metals at all three villages except for Zn in adults (Figure 2). These findings indicate the beneficial effect of pearling of wheat grains on the health of local residents.

The HI results of the three villages (Figure 2c) followed the order QD > GF > BS for both children and adults; as distance to the smelting facility increased, the HI decreased. Pearling resulted in a reduction in HI of 15.2, 23.4, and 31.2% for QD, GF, and BS, respectively. The differences between QD and the other two villages were significantly different ($p < .05$; Supplemental Table S5).

4 | DISCUSSION

4.1 | Wheat grain heavy metal concentrations

The Cd and Pb concentrations of most whole wheat flour and wheat flour samples were greater than the MPCs of these two elements in the food quality standard of China (Table 1; National Health Commission of China & China Food & Drug Administration, 2017), indicating potential health risk following consumption of these products in this area. The grain Cd and Pb concentrations are in agreement with results of Xing et al. (2016) and with those of Xing et al. (2018) and Liu, Cui, et al. (2020), who also showed elevated wheat Cd and Pb concentrations from areas affected by the same Pb smelter as in the current work (soil Cd = 1.90, Pb = 145 mg kg^{-1} in Xing et al. [2018]; soil Cd = 2.65, Pb = 261 mg kg^{-1} in Liu, Cui, et al. [2020]). The grain heavy metal concentrations in the present work were also comparable to results in other areas affected by heavy metal mining and smelting (Douay et al., 2008; Liu et al., 2017).

Concentrations of Cu, Pb, and Zn concentrations in wheat samples (Table 1; Supplemental Table S3) were similar to those found in earlier works (e.g., Cardoso et al., 2018; Cubadda et al., 2005; Giordano & Blandino, 2018; Yan et al., 2020). Pearling, or the removal of wheat bran, also affected heavy metal distribution in different wheat grain fractions (Table 1; Supplemental Table S3). As wheat grain is gradually pearled, a declining trend of trace microelement concentrations is typically observed (Li et al., 2011; Liu et al., 2007; Vignola et al., 2016). Liu et al. (2007) and Li et al. (2011) gradually pearled wheat grain, finding that the Zn concentration of the outer 4% of wheat grain was about two to three times the inner most wheat grain portion. According to Vignola et al. (2016), the Zn concentration of whole wheat flour is about 1.5–2 times that of the wheat flour. Giordano and Blandino (2018) found that the Pb concentration of the outer 0–5% of wheat grain was about four times greater than that of the neighboring wheat grain interior (5–10%). In the present work, Cd and Zn bran concentrations were 2.04–4.39 times that of the flour concentrations, and bran Cu and Pb concentrations were 8.55–50.0 times of the flour concentrations (Table 1; Supplemental Table S3). These sharp differences in element concentrations between bran and flour are in agreement with the results of Li et al. (2017) and Cardoso et al. (2018). The results of the present work also indicate that the outer 5% of wheat grain is heavy metal/micronutrient enriched. Giordano and Blandino (2018) found relatively similar Cd concentrations in the outer 0–5% and the neighboring 5–10% in wheat grain across six cultivars (0.188 vs. 0.178 mg kg^{-1}, respectively), whereas Pb concentrations in these wheat grain fractions across the six cultivars were much greater (0.170 vs. 0.044 mg kg^{-1}, respectively); these differences are in agreement with the Cd and Pb concentrations presented in Table 1 and Supplemental Table S3.

In the present work, Cd had the greatest PI (Table 1) and the greatest health risk (Figure 2) among all four metals studied. The whole wheat flour/wheat flour ratio results (Supplemental Table S3) indicate that pearling of about 5% of the outer wheat grain layer does not greatly reduce metal concentrations. In conjunction with current study findings and those of Giordano and Blandino (2018), we anticipate that pearling a greater percentage of the outer grain layer (e.g., 30–50% for commercial wheat flour) will result in a more pronounced reduction of edible heavy metals in the final wheat product. Greater pearling may also potentially lead to human health risk reductions.

The correlations between total soil and wheat grain sample heavy metal concentrations indicate that total soil heavy metal concentrations are an important predictor of wheat grain Cd, Cu, and Zn accumulation (Figure 1) in the study area. For Pb, no significant correlation was found between bran and total soil Pb concentration ($p > .05$). The responses of whole wheat flour and wheat flour Pb to total soil Pb concentrations did not increase dramatically as compared to other elements. This implies that total soil Pb should not be used as a predictor for wheat Pb accumulation in these soils. Earlier research suggested that, in areas where atmospheric Pb deposition occurs, wheat grain Pb is mainly attributed to leaf absorption of Pb particulates (Douay et al., 2008; Ma, Liu, Hu, et al., 2019; Ma, Liu, Jin, et al., 2019; Yang et al., 2015). The Pb atmospheric deposition in the study area was previously determined to be ∼100 mg $(m^2\ 30\ d)^{-1}$, which is much greater than "cleaner" areas (Qiu et al., 2016). Thus, the poor correlation between bran and total soil Pb concentrations and the near flatline Pb accumulation in (whole wheat) flour with increasing total soil Pb content imply that other Pb sources (e.g., atmospherically deposited) may be contributing to Pb accumulation in outer wheat grain layers.

The two main pathways by which heavy metal accumulation occurs in wheat grain are (a) the atmospheric deposition–leaf–grain pathway and (b) the soil–shoot–grain pathway (Li et al., 2020a; Ma, Liu, Jin, et al., 2019). The proportions of these two pathways differ between metals, although common heavy metals found in wheat grain can essentially be divided into two groups according to their dominant accumulation pathway. The first wheat grain group contains Pb and, as outlined above, is affected by atmospheric deposition. Lead is more difficult to be absorbed from soil and translocated into wheat grain as compared to other metals. Wheat grain Pb often has a bioconcentration factor (BCF) of <0.1 or even <0.01 in the context of wheat plants absorbing Pb from the soil (Boussen et al., 2013; Chen et al., 2020; Si et al., 2015; Wang et al., 2020). The second wheat grain group includes Cd, Zn, and Cu, for which wheat grain has BCF values >0.1.

The difference between heavy metal accumulation in plants is affected by other factors, including soil concentration and availability, interactions between different metals, and wheat genetics (Khaneghah et al., 2020; Rezapour et al., 2019). More specifically, the accumulation of heavy metals in wheat grain via plant root absorption plays an important role. Metals must enter plant roots via transmembrane transporter mechanisms, metal ions must be transported via the xylem (or even in the phloem), and translocation of heavy metals into wheat grain from the wheat peduncle must occur (Harris & Taylor, 2013; Yamaji & Ma, 2014). There are likely a multitude of heavy metal interactions that occur within plants, at various stages during absorption to translocation to the wheat grain, that are still not fully understood (Harris & Taylor, 2013; Li et al., 2020a; Luo et al., 2019). However, there is abundant evidence to confirm that atmospheric absorption plays a more important role in Pb accumulation in wheat grain than other metals (Douay et al., 2008; Ma, Liu, Hu, et al., 2019; Ma, Liu, Jin, et al., 2019). Compared with previous results of foliar Pb absorption (e.g., Douay et al., 2008; Ma, Liu, Hu, et al., 2019; Ma, Liu, Jin, et al., 2019), results from the current work indicated that the foliar accumulated Pb mainly existed in the wheat grain bran. As suggested above, removing the bran would reduce Pb in the flour and likely reduce human health risk.

4.2 | Health risk of consumption of wheat grain from the polluted areas

Unsafe heavy metal concentrations in wheat grain, with respect to human consumption, have been reported (Douay et al., 2008; Huang et al., 2008; Li et al., 2020b; Liu et al., 2017; Liu, Cui, et al., 2020; Ma et al., 2015; Rezapour et al., 2019). Because wheat is an important worldwide food source (Senapati & Semenov, 2020; Wu et al., 2020), elevated heavy metal concentrations in wheat grain imply that human health may be adversely affected.

However, most of the past research focused on heavy metal accumulation in wheat grain has compared wheat grain heavy metal concentrations with recommended safe levels (e.g., Douay et al., 2008; Li et al., 2020b; Liu, Cui, et al., 2020; Ma et al., 2015; Xing et al., 2016). Recently, the HQ method has been used for health risk assessments from food consumption (e.g., Chen et al., 2018; Huang et al., 2008; Kang et al., 2018; Liu, Liu, et al., 2020; Sharma et al., 2018). In our case, this method considered the daily consumption amount of wheat products and the tolerable daily intake of heavy metals of individuals in the risk assessment, based on the assumption that wheat grain heavy metals can be totally absorbed into human blood following consumption. This, undoubtedly, is different from actual conditions. For example, research has found that less than 20% of Cd in wheat or rice grain can be extracted with simulated intestinal fluid (Liu et al., 2017; Wang et al., 2020; Yang et al., 2012). Thus, the calculations presented, based on total heavy metal concentrations in wheat products, likely overestimate risk. However, the approach used in the current study could be considered a relatively conservative estimate of health risk.

On the other hand, other bioaccessibility assessments have used simulated body fluids to assess heavy metal bioaccessibility in materials such as foods, with the results of such scenarios affected by operational parameters; no methods with commonly accepted parameters have been universally adopted (Kastury et al., 2018; Xing et al., 2019). Consequently, heavy metals bioaccessibility in wheat estimated with such methods varies greatly from study to study and even for the same element in the same study (Liu et al., 2017; Praveena & Omar, 2017). Furthermore, the bioaccessibility methodology of heavy metals in particulate matters and soil have been widely investigated, whereas those for food have been less studied (Kastury et al., 2018; Ren et al., 2020). The bioaccessibility assessment method and factors affecting the assessment results of food still need further investigation and validation to human absorption. According to existing results, food-borne Ni bioavailability in the human gastrointestinal tract ranges between 1.7 and 10% (Babaahmadifooladi et al., 2020), and larger variations in bioavailability (low to 100%) have been found in baby food for Fe, Zn, Cu, and Mn (da Silva et al., 2013). Rice Cd bioaccessibility within the human gastrointestinal tract has also been shown to vary substantially (7.9–24%) (Tefera et al., 2020). Until standard guidelines are universally accepted, it is reasonable and reliable to use a conservative human health risk estimation with respect to wheat grain total heavy metal concentrations. With this stated, results of the present work regarding potential wheat consumption health risks in the area studied (Figure 2; Supplemental Table S3) suggest that (a) children have greater noncarcinogenic risk than adults; (b) adults have greater carcinogenic risks than children; (c) greater risks of Cd existed than for Cu, Pb, and Zn; and (d) pearling reduced the risk for Pb to a greater extent than for Cd, Cu, and Zn.

Heavy metal concentrations in crops or vegetables harvested in nonferrous smelting areas have been previously investigated (Douay et al., 2008; Li et al., 2020b; Liu et al., 2017; Liu, Cui, et al., 2020; Xing et al., 2016); yet, estimation of potential health issues associated with human heavy metal intake have rarely been studied. The HQ was used in the current study to estimate the noncancer risk of consuming such materials over the course of a human's lifetime. In the present study, the HQ values of individual heavy metals, and especially those of Cd and Pb, were more than 50 times those from relatively "clean" areas (Huang et al., 2008; Kang et al., 2018; Liu, Liu, et al., 2020). Zhou et al. (2018) reported a Cu ADD of approximately 0.034–0.065 mg kg^{-1} d^{-1} from food consumption for residents living close to a Cu mining area. In the current work, the Cu ADDs via wheat flour consumption were 0.017–0.0347 mg kg^{-1} d^{-1} (Table 2), which are comparable to Zhou et al. (2018). Overall, the current study results are important for understanding and estimating heavy metal intake by residents in nonferrous metal smelting areas, especially with respect to consumption of food products produced in the vicinity of heavy metal contamination sources.

5 | CONCLUSIONS

Lead smelting activities in this area of China have resulted in soil and wheat grain Cd, Cu, Pb, and Zn accumulation. Compared with the recommended safe levels presented in food quality standards, Cd was the most accumulated wheat grain metal. Pearling reduced Cu and Pb wheat concentrations to the greatest extent; unfortunately, pearling did not greatly reduce Cd human health risk, and thus additional research is required to reduce Cd accumulation in wheat in this and similar areas. Pearling did, however, reduce the noncarcinogenic Pb risk by the greatest extent of all metals studied. Among all four metals investigated, the greatest carcinogenic and noncarcinogenic risks were from Cd. Children appeared to be at greater noncarcinogenic health risk and at less carcinogenic health risk as compared to adults. Soil heavy metal sequestration methodologies, transforming local farmlands to nonfood plant production systems, or relocating residents may be the only practical means by which reductions in human health risks are realized.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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SUPPORTING INFORMATION

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