



Occurrence and dietary intake risk assessment of heavy metals in Fuji apple

Mengying Shuai^{a,1}, Yihui Liu^{a,1}, Yishan Wei^b, Peng Jiang^b, Mingyu Liu^a, Kunyu Wang^a, Zhichao Li^a, Xiaoming Li^a, Xiabing Kong^a, Min Xu^a, Jiyun Nie^{a,*}

^a College of Horticulture, Qingdao Agricultural University/Laboratory of Quality & Safety Risk Assessment for Fruit (Qingdao), MARA/Technical Agency for the Whole Process Control of Agricultural Product Quality & Safety, MARA/Qingdao Key Lab of Modern Agriculture Quality and Safety Engineering, Qingdao 266109, China

^b Qingdao Agricultural Products Quality and Safety Center, Qingdao 266109, China

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ABSTRACT

Heavy metal is one of the main safety risk factors for fruits and has garnered growing scientific interest. In this study, the occurrence and the health risks to consumers of heavy metals in Fuji apples were assessed. The contents of the heavy metals in whole fruit (WF) and pulp (PP) of Fuji apple were detected by ICP-MS. None of the samples exceeded the MLs of the studied heavy metals, and both SPI and IPI of the heavy metals in all samples were at safe levels. The contents of As, Cd, Mn, Cu, and Ni in WF samples demonstrated strong positive correlations with those in PP samples, with r of 0.92, 0.90, 0.87, 0.76, and 0.75, respectively. The DIR of the heavy metals in Fuji apple was far below the tolerance value. The values of THQ, TTHQ, and CR of heavy metals in Fuji apples decreased with the age of consumers and for the same age group, the values to males were lower than those to females. For all the assessed heavy metals (Cd, Pb, and As), the CR values were all below 1×10^{-6} , which reflected a low and negligible risk to the consumers of Fuji apple.

1. Introduction

Apple (*Malus domestica*) belongs to the apple genus in the Rosaceae family (Zhang, et al., 2019). Its fruit is rich in sugars, acids, vitamins, flavonoids, dietary fiber, pectin, amino acids, and minerals, which have beneficial effects to human health (Zhang, et al., 2023; Arnold and Gramza-Michalowska, 2024; Wang, et al., 2024), such as reducing cardiovascular risk, lowering blood lipids, preventing obesity, and moistening the intestines (Skinner et al., 2018). Apple is widely cultivated throughout the world. It is the first important deciduous fruit tree in China and has an annual production of nearly 48 million tons, accounting for about 50 % of the world's total apple production (Liang et al., 2021). China has four main apple producing areas, including Bohai Bay (Liaoning, Shandong, and Hebei provinces), Loess Plateau (Shanxi, Shaanxi, Gansu, and Henan provinces), Southwest Cool Highlands (Sichuan, Yunnan, and Guizhou provinces), and the Yellow River communist-held (Eastern Henan, Southwestern Shandong, Northern Jiangsu, and Northern Anhui provinces), and Fuji is its first dominant

cultivar, accounting for about 70 % of its apple production (Cong, 2010; Nie et al., 2016; Ministry of Agriculture and Rural Affairs, 2022).

Food contamination includes heavy metals, mycotoxins, pathogenic microorganisms, and persistent pollutants, etc. (Alabi et al., 2024). At a global scale, about 600 million people (almost one in ten) and 0.42 million people fall ill or die each year due to the consumption of contaminated food (Širić et al., 2022). The heavy metals in fruits mainly include Mn, Ni, Cu, Zn, As, Cd, and Pb (Nie, 2020). The most common exposure route for human to heavy metals is dietary intake (Al-Huqail et al., 2023). Compared with other routes of exposure such as inhalation and dermal contact, food ingestion accounts for more than 90 % of the intake of heavy metals (Širić et al., 2022). Consuming heavy metal-contaminated meals may result in heavy metal buildup in various tissues of human body, producing both chronic and acute health effects, such as cancer and acute poisoning (Alabi et al., 2024).

The per capita apple possession of Chinese people has been increasing year by year, and reached 32.8 kg in 2022 (Ministry of Agriculture and Rural Affairs, 2022). When people eat apples, they may

Abbreviations: ICP-MS, Inductively coupled plasma mass spectrometry; MLs, maximum limits; SPI, single pollution index; IPI, integrated pollution index; DIR, daily intake rate; THQ, target hazard quotient; TTHQ, total target hazard quotient; CR, carcinogenic risk; LOD, the limit of detection; LOQ, the limit of quantification; CSF, cancer slope factor; ANOVA, one-way analysis of variance.

* Corresponding author.

E-mail address: jiyunnie@163.com (J. Nie).

¹ These authors contributed equally to this paper.

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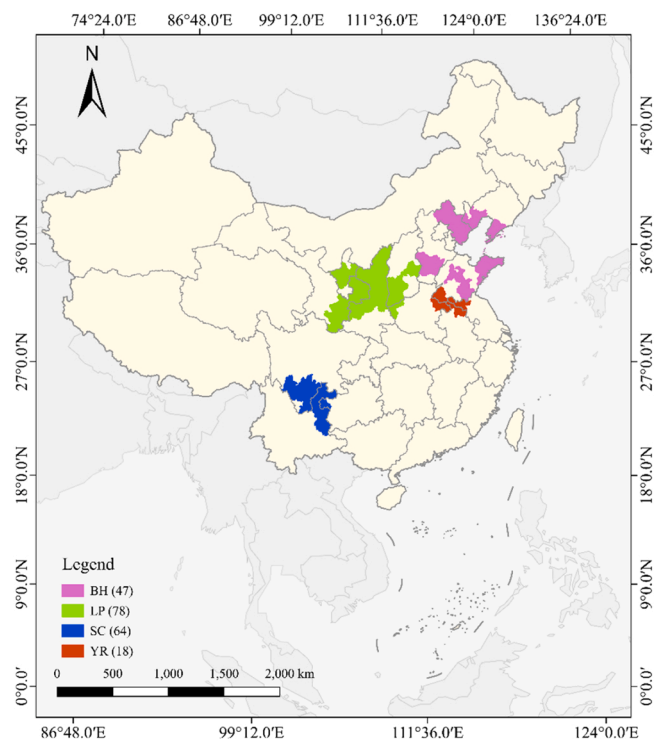


Fig. 1. Sampling locations of Fuji apple samples.

peel them or not, depending on personal habits. As the most important fresh fruit in China, if containing high level of heavy metals, apple will inevitably pose a risk to consumer health. Due to the differences in the tolerance and intake of heavy metals, the toxicity of heavy metals will vary among different population groups (Özcan et al., 2012). Therefore, it is necessary to determine the content of the heavy metals in apple of the whole fruit (WF) and the pulp (PP) and assess their health risks to different groups of consumers.

In this study, we aim to clarify the level and the diet intake risk of heavy metals in the WF and PP of Fuji apple from the main producing areas in China. Seven heavy metals in 414 Fuji apple samples (207 WF samples and 207 PP samples, respectively) were determined, including Mn, Ni, Cu, Zn, As, Cd, and Pb. The heavy metal contamination risk of apple consumption to different consumer groups was systematically assessed, including SPI, IPI, DIR, THQ, TTHQ, and CR. The findings may provide some insight into heavy metal pollution of apple in China, and serve as a basis for comparison with other countries and other fruits.

2. Materials and methods

2.1. Fuji apple sample collection

At the harvest season in the autumn of 2021, 207 Fuji apple samples were collected from Bohai Bay (BH; $n = 47$), Southwest Cool Highlands (SC; $n = 64$), Loess Plateau (LP; $n = 78$), and Yellow River communist-held (YR; $n = 18$) in China (Fig. 1). Apples were sampled using a randomized sampling protocol. Within each selected orchard, five apple trees were randomly chosen. From each tree, a mature fruit were collected from each of the four cardinal directions (east, west, south, and north) of the canopy, resulting in a representative sample of 20 apples per orchard. The samples were cleaned with deionized water. And each sample was divided into two subsamples to serve as the WF and PP samples. The apples were peeled using a professional peeling knife with circumferential peeling along the equatorial plane of the fruits. The peeling thickness was controlled at 1.0 mm. Then, the WF and PP samples were all sliced with a ceramic knife and baked at 65°C in a

blower oven (GFL-70, Tianjin Labotery Instrument Equipment Co., Ltd.) until a constant weight. Finally, the WF and PP samples were ground into powder in the mono mill (Pulverisette 6, FRITSCH GmbH) and passed through 100-mesh nylon sieves for preservation.

2.2. Determination of heavy metals in Fuji apple samples

2.2.1. Extraction of heavy metals from Fuji apple

Extraction and determination were conducted based on previous studies (Nie et al., 2016; Nie, 2018; Xu et al., 2023). Approximately 0.300 g of sieved Fuji apple powder sample was accurately and rapidly weighed and 7 mL of HNO_3 (65 %, Guaranteed reagent) was added to a TeflonTM-coated digestion vessel (BHW-09C Heating Block, Beijing Hengao Instrument Co., Ltd.). A microwave-assisted digestion system (MARS 6, CEM, USA) was used to completely digest the apple samples. The samples were heated in the MARS according to the following programmer: the temperature was increased from 0 °C to 120 °C over 5 min, then heated to 150 °C over 1 min. The temperature reached a maximum of 180 °C and maintained 20 min, after which it was cooled down over 30 min (Nie et al., 2016; Xu et al., 2023). After digestion, the TeflonTM-coated digestion vessels were transferred to an acid digestion evaporator for acid evaporation and concentration until the solution volume was reduced to below 2 mL, followed by termination of heating and natural cooling to ambient temperature. The solution was diluted to 20 mL with ultrapure water, thoroughly mixed, filtered through a 0.22 μm membrane, and stored at 4 °C for heavy metal analysis.

2.2.2. Optimization of detection methods

ICP-MS (7900, Agilent) was used to determine the heavy metals (Mn, Ni, Cu, Zn, As, Cd, and Pb). The key operating parameters for the analysis of the diluted samples were as follows: radio frequency power, 1550 W; plasma gas flow rate, 15 L/min; carrier gas flow rate, 0.99 L/min; and auxiliary gas flow rate, 0.9 L/min. Interference equation was used to correct the mass spectral interference. The stability of the instrument was calibrated with the internal standard solution. Internal standard was selected according to the mass number of the measured element. ^{45}Sc , ^{115}In , and ^{209}Bi were used as the internal standards for ^{55}Mn , ^{111}Cd , and ^{208}Pb , respectively. ^{72}Ge was used as the internal standard for ^{60}Ni , ^{63}Cu , ^{66}Zn , and ^{75}As .

2.2.3. Methodological assessment

Three-fold standard deviation was used as the LOD of the element. Ten-fold standard deviation was used as the LOQ of the element. The LOD and LOQ ranged from 0.004 ng/kg to 0.2 ng/kg and 0.01 ng/kg to 1.1 ng/kg, respectively. The analytical curve for each heavy metal was of an r^2 value between 0.9995 and 1.0000. The ICP-MS technique in this study showed good recoveries of 94.1 %–118.7 %. This method meets the requirements of National Standards of the People's Republic of China for elemental content analysis (Nie, 2018).

2.3. Calculation of pollution levels

SPI was calculated using Eq. (1), and the assessment standard of heavy metals is listed in Table S1 (Nie et al., 2016; Wei et al., 2018; Yang et al., 2022).

$$P_i = \frac{C_i}{S_i} \quad (1)$$

In the equation, P_i is the heavy metal SPI value of heavy metal i ; C_i is the concentration of heavy metal i , (mg/kg); S_i is the evaluation standard of heavy metal i , (mg/kg).

IPI was calculated using Eq. (2), and the results were evaluated using the comprehensive pollution classification standard of heavy metals in Table S2 (Cheng et al., 2014; Nie et al., 2016; Wei et al., 2018; Uraguchi et al., 2017; Yang et al., 2022).

Table 1

Comparison of heavy metal content between whole fruit and pulp of Fuji apple.

Fuji apple	Mn (mg/kg)	Zn (mg/kg)	Cu (mg/kg)	Ni (μg/kg)	Pb (μg/kg)	As (μg/kg)	Cd (μg/kg)
WF	0.513 ± 0.217 aA	0.510 ± 0.218 aA	0.350 ± 0.108 aA	7.49 ± 5.07 aA	8.23 ± 5.58 aA	2.50 ± 2.82 aA	0.627 ± 0.659 aA
PP	0.319 ± 0.148 bB	0.419 ± 0.178 bB	0.314 ± 0.102 bB	6.14 ± 4.74 bB	3.87 ± 2.46 bB	1.96 ± 2.52 bA	0.391 ± 0.485 bB

Note: WF and PP stand for whole fruit and pulp, respectively. Data with the same lowercase letter in the same column are not significantly different at the level of $p < 0.05$, and data with the same uppercase letter are not significantly different at the level of $p < 0.01$. The same as below.

$$IPI = \sqrt{\frac{P_{\max}^2 + P_{\text{ave}}^2}{2}} \quad (2)$$

In the equation, $P_{\max}$ refers to the maximum value of SPI of the seven studied heavy metals in each sample; P_{ave} indicates the average value of SPI of the seven studied heavy metals in each sample.

2.4. Calculation of daily intake of heavy metals from Fuji apple

Point assessment and probability assessment models were used to assess the safety risk of human dietary intake of heavy metals from Fuji apple. The DIR of heavy metals caused by Fuji apple consumption was estimated with Eq. (3). In Eq. (3), DIR is the estimated daily intake rate of some heavy metal, [$\mu\text{g} / (\text{d} \cdot \text{kg } W_b)$]; I is the ingestion rate of Fuji apple, (kg/d); C is the content of some heavy metal in Fuji apple, (mg/kg); and W_b is the body weight, (kg).

$$DIR = \frac{C \times I}{W_b} \times 1000 \quad (3)$$

In this study, consumers were divided into different age groups for estimation and analysis. Table S3 lists the average weight of different age groups and consumption data of Fuji apple. The ingestion rate of Fuji apple (I) was estimated according to the total yield, export rate, processing proportion, and storage loss rate (Nie et al., 2016). The consumption of Fuji apple in different age groups was estimated by referring to Wu et al. (Wu et al., 2018).

2.5. Calculation of target hazard quotient of heavy metals from Fuji apple

The THQ was calculated using Eq. (4), where: E_f is the exposure frequency (d/a), taken as 120 d (Mao et al., 2019); E_d is the exposure duration (a), set as 74.7 years for adult males, 80.5 years for adult females (WHO, 2021), and 18 years for children in China (Chen et al., 2015); C is the concentration of the heavy metal in Fuji apple samples ($\mu\text{g}/\text{kg}$); T_e is the non-carcinogenic exposure time (d), calculated as 365 $\text{d}/\text{year} \times E_d$; RfD is the oral reference dose [$\mu\text{g}/(\text{d} \cdot \text{kg } W_b)$], with values for Mn (0.14), Ni (0.02), Cu (0.04), Zn (0.3), As (0.0003), Pb (0.0035), and Cd (0.001) (Shaheen et al., 2016; Chen et al., 2015). The meanings of “ I ” and “ W_b ” were the same as those in Eq. (3).

$$THQ = \frac{E_f \times E_d \times I \times C}{W_b \times T_e \times RfD} \quad (4)$$

$THQ < 1$ indicates that the intake of a heavy metal in Fuji apple by a population group is safe. Conversely, when $THQ \geq 1$, the heavy metal was considered to have a non-acceptable health risk for humans (Shaheen et al., 2016; Mao et al., 2019). In addition, humans could be simultaneously exposed to several metals, which might produce a superposition effect. TTHQ assessment was conducted according to Eq. (5). TTHQ is the total THQ of the studied seven heavy metals. In this study, considering the behavioral and physiological differences, the consumers were divided into different age groups for probability assessment.

$$TTHQ = \sum THQ \quad (5)$$

Table 2

Content comparison between heavy metals in whole fruit and pulp of Fuji apple.

Heavy metal	WF	PP
Mn (mg/kg)	0.513 ± 0.217 aA	0.319 ± 0.148 bB
Zn (mg/kg)	0.510 ± 0.218 aA	0.419 ± 0.178 aA
Cu (mg/kg)	0.350 ± 0.108 bB	0.314 ± 0.102 bB
Ni (μg/kg)	7.49 ± 5.07 cC	6.14 ± 4.74 cC
Pb (μg/kg)	8.23 ± 5.58 cC	3.87 ± 2.46 cC
As (μg/kg)	2.50 ± 2.82 cC	1.96 ± 2.52 cC
Cd (μg/kg)	0.627 ± 0.659 cC	0.391 ± 0.485 cC

Note: WF and PP stand for whole fruit and pulp, respectively. Data with the same lowercase letter in the same column are not significantly different at the level of $p < 0.05$, and data with the same uppercase letter are not significantly different at the level of $p < 0.01$. The same as below.

2.6. Calculation of carcinogenic risk of heavy metals from Fuji apple

CR was used to estimate the probability of cancer occurrence in an individual's lifetime due to exposure to potential carcinogens (Shi et al., 2023). Here, CR was calculated according to Eq. (6). For the CSF [$\text{mg}/(\text{d} \cdot \text{kg } W_b)$], Cd, Pb, and As were 15, 0.0085, and 1.5, respectively (Mao et al., 2019; Shaheen et al., 2016). CR was divided into five grades, including very low ($CR \leq 1 \times 10^{-6}$), low ($1 \times 10^{-6} < CR \leq 1 \times 10^{-4}$), moderate ($1 \times 10^{-4} < CR \leq 1 \times 10^{-3}$), high ($1 \times 10^{-3} < CR \leq 1 \times 10^{-1}$), and very high ($CR > 1 \times 10^{-1}$) (Wu et al., 2019).

$$CR = \frac{E_f \times E_d \times I_f \times C \times CSF}{W_b \times T_e} \quad (6)$$

where T_e refers to non-carcinogenic exposure time, (d); and CSF refers to carcinogenic slope factor, [$\text{mg}/(\text{d} \cdot \text{kg } W_b)$].

2.7. Statistical analysis

The coefficient of variation and data distribution of different heavy metals in Fuji apple samples were analyzed using @Risk 5.5 software (Palisade, USA). The content level of each heavy metal in Fuji apple was divided into three levels (namely low, medium, and high) by the 30th and 70th percentiles of its best-fit distribution, which has the smallest Chi-Sq Statistic (Gupta et al., 2018). The test of significance of the difference was performed through ANOVA and Duncan's test for multiple comparisons for equal variances using the SPSS (Version 26.0, IBM, USA). Correlation analysis was performed using Chiplot (<https://www.chiplot.online/>). Origin (Origin 2021, USA) was used to plot the heat maps.

3. Results and discussion

3.1. Heavy metal content in Fuji apple

3.1.1. Comparison between different parts of Fuji apple

Significant differences in heavy metal contents were found between WF and PP samples of Fuji apples (Table 1). As showed significant difference ($p < 0.05$), while Zn, Mn, Pb, Ni, and Cd exhibited highly significant differences ($p < 0.01$). Among WF samples, Pb, Ni, As and Cd differed significantly ($p < 0.01$), whereas Zn and Mn showed no significant variation. For PP samples, all measured metals except Cu and Mn

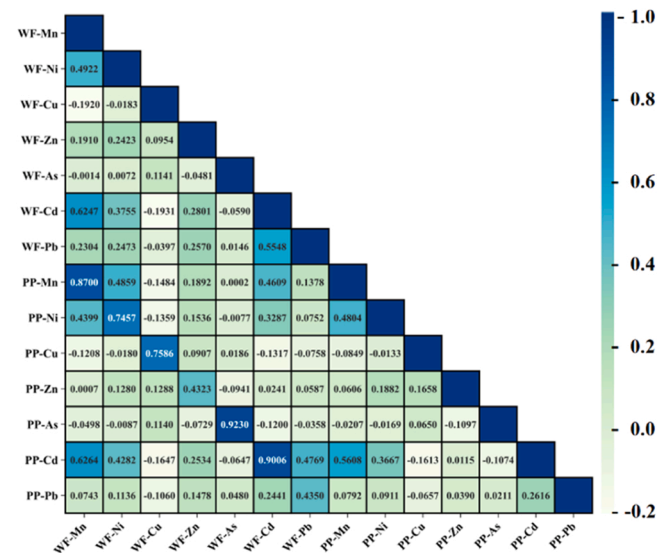


Fig. 2. Correlation analysis of heavy metals in whole fruit and pulp of Fuji apples. (Note: The deepening of the green color represents a gradual increase in the positive correlation. The critical value of correlation coefficient is $r = 0.1364$ at $\alpha = 0.05$ and $r = 0.1787$ at $\alpha = 0.01$).

demonstrated significant differences ($p < 0.01$) (Table 2). The heavy metal contents followed an order of Mn (0.513 mg/kg) and Zn (0.510 mg/kg) > Cu (0.350 mg/kg) > Pb (8.23 µg/kg) > Ni (7.49 µg/kg) > As (2.50 µg/kg) > Cd (0.627 µg/kg) in WF samples, and Zn (0.419 mg/kg) > Mn (0.319 mg/kg) and Cu (0.314 mg/kg) > Ni (6.14 µg/kg) > Pb (3.87 µg/kg) > As (1.96 µg/kg) > Cd (0.391 µg/kg) in PP samples (Table 1).

The MLs for heavy metals in fruits refers to the highest permissible content of heavy metals allowed in fruits (Xu et al., 2023). The results showed that the average contents of each heavy metal in all samples were lower than its ML. The apple peel, which is directly exposed to the external environment and rich in pectin, is more prone to accumulating pollutants. Its pectin content provides a strong binding capacity for certain heavy metals, such as Cd, further enhancing its tendency to accumulate pollutants compared to the pulp (Özcan et al., 2012). The contents of Cu, Mn, and Zn were significantly higher than those of Ni, As, Cd and Pb in both WF and PP samples, indicating that these three heavy metals are more likely to accumulate in Fuji apple (Özcan, et al., 2012). Previous studies have demonstrated that heavy metals in apple follow an order of Zn > Cu > Pb > Ni > Cd, which is consistent with the results obtained for WF samples of Fuji apple in this study (Imširović et al., 2018).

Mn, Ni, Cu, As, and Cd contents in WF samples had a strong positive correlation with PP, the Pearson's correlation coefficients (r) were 0.8700, 0.7457, 0.7586, 0.9230, and 0.9006, respectively, while Zn ($r = 0.4320$) and Pb ($r = 0.4350$) showed relatively weaker correlations (Fig. 2). The statistical analysis revealed highly significant correlations

($p < 0.01$) between As and Cd in both PP and WF samples, exhibiting strong correlation coefficients of 0.9230 and 0.9010, respectively. The strong correlation of heavy metal concentrations between WF and PP in Fuji apples stems primarily from shared anthropogenic pollution sources (Luo et al., 2021), mediated through three interconnected processes including heavy metal accumulation in agricultural soils, contaminant translocation via polluted irrigation water (Cai et al., 2019), and atmospheric particulate deposition (Liu et al., 2023). These pollutants are absorbed through both epidermal and mesocarp tissues (Uraguchi et al., 2017), with their co-accumulation patterns being intensified by the concurrent use of agricultural chemicals, such as phosphate fertilizers and sewage sludge-based soil amendments (Luo et al., 2021; Yang et al., 2021). The preferential binding of cadmium to pectin in apple peel (Özcan et al., 2012) modulates peel-specific Cd accumulation dynamics, consequently affecting the metal's distribution profile throughout the fruit (Liu et al., 2022), which provides a plausible explanation for the consistent Cd concentration relationships observed between peel and whole fruit samples.

3.1.2. Comparison between different producing areas

The content of Mn, Ni, Cd, As, and Pb in Fuji apples samples from SC region showed significant differences compared to the other three producing regions ($p < 0.05$), with Mn and Cd in SC region exhibiting extremely significant differences compared to BH, LP, and YR regions ($p < 0.01$). WF samples from SC contained significantly higher levels of Mn (0.685 mg/kg) and Pb (12.2 µg/kg) than those from YR (Mn: 0.401 mg/kg; Pb: 7.08 µg/kg), BH (Mn: 0.509 mg/kg; Pb: 6.33 µg/kg), and LP (Mn: 0.44 mg/kg; Pb: 4.48 µg/kg) ($p < 0.01$). Similarly, PP samples from SC region showed significantly higher contents of Mn (0.405 mg/kg), Ni (8.72 µg/kg), Cd (0.955 µg/kg), and Pb (4.63 µg/kg) compared to those from YR (Mn: 0.223 mg/kg; Ni: 2.35 µg/kg; Cd: 0.133 µg/kg; Pb: 2.63 µg/kg), BH (Mn: 0.322 mg/kg; Ni: 6.32 µg/kg; Cd: 0.214 µg/kg; Pb: 2.63 µg/kg), and LP (Mn: 0.270 mg/kg; Ni: 4.78 µg/kg; Cd: 0.096 µg/kg; Pb: 3.92 µg/kg) ($p < 0.01$) (Table 3).

The geological backgrounds of the four major apple production regions (BH, SC, LP, and YR) exhibit distinct characteristics that fundamentally determine their soil properties and, consequently, the heavy metal accumulation in Fuji apples. Regionally, the SC area is characterized by carbonate and purple sedimentary rocks, which are naturally enriched with Mn, Cd, and As, establishing an inherently high geochemical background (Chen et al., 1991). In contrast, the BH and YR regions lie within the vast alluvial plains of the Yellow and Haihe Rivers, where fertile soils derived from alluvial-lacustrine deposits are rich in clay minerals and organic matter such as humus (Sier et al., 2016). The LP region, meanwhile, is defined by deep, alkaline, and porous loess deposits, which form the parent material for its characteristic loess and loe soils (Chen et al., 1991).

In this study, the contents of Mn, Ni, Cd, As, and Pb (excluding Cu and Zn) in WF and PP samples from the SC region were significantly higher than those in the BH, LP, and YR regions (Table 3). This pattern can be primarily attributed to the generally acidic soils in the SC region (Chen et al., 1991). From a soil chemistry perspective, acidic conditions

Table 3
Comparison of heavy metal content between Fuji apples from four main production areas of China.

Part	Area	Sample	Mn (mg/kg)	Zn (mg/kg)	Cu (mg/kg)	Ni (µg/kg)	Pb (µg/kg)	As (µg/kg)	Cd (µg/kg)
WF	BH	47	0.509 ± 0.171 bB	0.418 ± 0.606 bB	0.336 ± 0.108 bB	7.75 ± 3.26 bAB	6.33 ± 3.02 bcB	3.24 ± 4.26 aA	0.334 ± 0.241 bB
	SC	64	0.685 ± 0.267 aA	0.604 ± 0.305 aA	0.319 ± 0.094 bB	10.49 ± 6.97 aA	12.2 ± 4.92 aA	1.95 ± 0.68 aA	1.48 ± 0.492 aA
	LP	18	0.400 ± 0.171 cB	0.614 ± 0.501 aA	0.432 ± 0.151 aA	4.38 ± 2.05 cC	4.48 ± 2.64 cB	2.12 ± 1.00 aA	0.268 ± 0.193 bB
	YR	77	0.401 ± 0.083 cB	0.452 ± 0.181 bB	0.363 ± 0.097 bB	5.57 ± 2.89 cBC	7.08 ± 6.10 bB	2.62 ± 3.05 aA	0.183 ± 0.163 bB
PP	BH	47	0.322 ± 0.122 bB	0.406 ± 0.171 aA	0.298 ± 0.097 bB	6.35 ± 2.52 bAB	3.26 ± 2.52 bcAB	2.74 ± 4.13 aA	0.214 ± 1.193 bB
	SC	64	0.405 ± 0.207 aA	0.409 ± 0.186 aA	0.295 ± 0.087 bB	8.72 ± 6.55 aA	4.63 ± 2.27 aA	1.34 ± 0.76 bA	0.955 ± 0.494 aA
	LP	18	0.270 ± 0.071 bcBC	0.426 ± 0.182 aA	0.321 ± 0.100 bB	4.78 ± 3.15 bBC	3.92 ± 2.62 abAB	2.07 ± 2.37 abA	0.096 ± 0.138 bB
	YR	77	0.223 ± 0.046 cC	0.442 ± 0.149 aA	0.392 ± 0.138 aA	2.35 ± 1.43 cC	2.63 ± 0.98 cB	1.72 ± 0.77 abA	0.133 ± 0.083 bB

Note: WF and PP stand for whole fruit and pulp, respectively. Data with the same lowercase letter in the same column are not significantly different at the level of $p < 0.05$, and data with the same uppercase letter are not significantly different at the level of $p < 0.01$.

Table 4
Grading of heavy metal content in the whole fruit and pulp of Fuji apples.

Part	Heavy metal	Best-fit distribution	p	Low	Medium	High
WF	Cd (μg/kg)	InvGauss	0.0000	< 0.182	0.182–0.633	> 0.633
	As(μg/kg)	Pearson 5	0.0129	< 1.39	1.39–2.52	> 2.52
	Ni (μg/kg)	LogLogistic	0.5015	< 4.82	4.82–8.22	> 8.22
	Pb (μg/kg)	Pearson 5	0.7095	< 5.02	5.02–9.23	> 9.23
	Zn (mg/kg)	InvGauss	0.7954	< 0.359	0.359–0.570	> 0.570
	Mn (mg/kg)	InvGauss	0.8341	< 0.383	0.383–0.563	> 0.563
	Cu (mg/kg)	LogLogistic	0.8522	< 0.291	0.291–0.392	> 0.392
PP	Cd (μg/kg)	InvGauss	0.0023	< 0.080	0.080–0.337	> 0.337
	As(μg/kg)	Pearson 5	0.2908	< 0.941	0.941–1.86	> 1.86
	Ni (μg/kg)	LogLogistic	0.3352	< 0.366	0.366–0.687	> 0.687
	Pb (μg/kg)	Pearson 5	0.6753	< 2.41	2.41–4.37	> 4.37
	Zn (mg/kg)	LogLogistic	0.5821	< 0.319	0.319–0.471	> 0.471
	Mn (mg/kg)	LogLogistic	0.5473	< 0.238	0.238–0.338	> 0.338
	Cu (mg/kg)	LogLogistic	0.9908	< 0.260	0.260–0.353	> 0.353

enhance the solubility of elements including Mn, Zn, Cu, Ni, and As, while simultaneously reducing the adsorption capacity of soil colloids. This dual effect increases the concentrations of these elements in the soil solution, thereby promoting their phytoavailability. Furthermore, plants may physiologically increase the uptake of soluble elements under acidic stress by regulating root exudate composition or modulating the expression of specific transport proteins (Li et al., 2014). The SC region's abundant precipitation further amplifies this process; rainfall-driven mineral leaching facilitates the infiltration of heavy metals into groundwater and irrigation sources, subsequently promoting their translocation and bioaccumulation in vegetation (Luo et al., 2009; Sungur et al., 2015; Xu et al., 2023).

In the LP region, the alkaline soil pH inherent to its loess-derived soils creates chemical conditions that are generally unfavorable for the plant uptake of most heavy metals (Chen et al., 1991), explaining the lower overall accumulation observed there. Soils in the BH and YR regions, being rich in humus which has a strong enrichment effect on metallic elements (Sier and Monteith, 2016), exhibit a greater capacity for metal retention. This characteristic likely explains why heavy metal concentrations in these two regions were significantly higher ($p < 0.01$) than those in the LP region (Liu et al., 2023). Apart from these geogenic and pedogenic factors, specific agricultural practices also contribute to heavy metal profiles. Research has demonstrated that prolonged use of Cu-based fungicides (e.g., Bordeaux mixture) leads to marked accumulation of copper in apple fruits (Shan et al., 2022), a phenomenon widely reported across apple-producing regions and attributed to the persistent nature of copper in the soil–plant system (Nie et al., 2020). Similarly, elevated Zn levels in plants are often linked to wastewater irrigation practices (Lü et al., 2021), suggesting this pathway may be a significant contributor to Zn accumulation in Fuji apples.

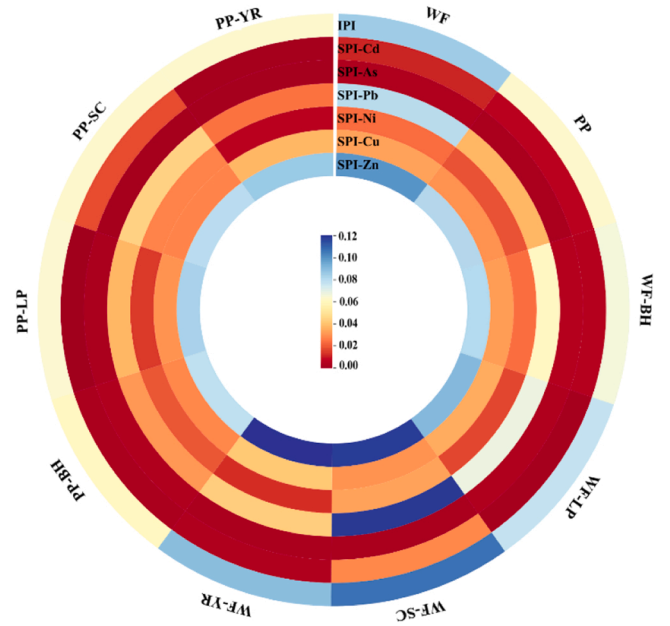


Fig. 3. Heavy metal contamination of SPI and IPI in different parts and production areas of Fuji apples. (Note: The SPI and IPI of heavy metals in Fuji apple gradually decreased from blue to red).

3.1.3. Grading of heavy metal content in Fuji apple

For a more intuitive understanding of the heavy metal content in Fuji apple samples, the best fit distribution functions of the seven heavy metals were obtained through content distribution fitting (Table 4). The content of heavy metals to Fuji apples was divided into three grades: low (<30 %), medium (30 %–70 %), and high (>70 %). Through distribution fitting analysis of seven heavy metal content in Fuji apple samples, we gained in-depth understanding of the pollution characteristics and potential sources of different metals.

The fitting distribution for Pb (WF: $p = 0.7095$; PP: $p = 0.6753$) and As (WF: $p = 0.0129$; PP: $p = 0.2908$) was the Pearson 5, whose right-skewed (skewness >2) and heavy-tailed (kurtosis >6) properties suggest intermittent high-intensity pollution events, possibly from non-point sources such as atmospheric deposition or irregular fertilizer applications (Delignette-Muller and Dutang, 2015; EPA, 2021). Cu (WF: $p = 0.8522$; PP: $p = 0.9908$) and Ni (WF: $p = 0.5015$; PP: $p = 0.3352$) showed good fitting with LogLogistic distribution ($p > 0.5$) (Table 4). The accumulation patterns of Cu and Ni demonstrate multifactorial regulation characteristics ($p < 0.05$). Distribution profiles reveal significant synergistic effects between natural soil background levels and anthropogenic activities (particularly agricultural fertilization) ($p < 0.01$), which aligns with the LogLogistic distribution model's capability to characterize multi-mechanism interactions (Özcan et al., 2012; Wang et al., 2021). The interception effect of the cuticular wax layer leads to an InvGauss distribution of Mn and Zn in WF, demonstrating a transport characteristic of "high initial resistance followed by gradual stabilization" (Li et al., 2018). In the PP samples, due to regulation by multiple physiological barriers including vascular unloading and vacuolar compartmentalization, Mn and Zn exhibited a LogLogistic distribution, reflecting their multi-stage rate-limited accumulation properties (Uraguchi et al., 2017). Cd in Fuji apples consistently followed an InvGauss distribution pattern in both WF ($p = 0.0000$) and PP ($p = 0.0023$) samples. The fitting results for Cd in both WF and PP were unsatisfactory ($p < 0.01$), indicating that its pollution process might not be regulated by biological barriers but rather occurs through systemic pathways (such as irrigation water absorption) into the fruit interior (Sungur et al., 2015; Xu et al., 2023).

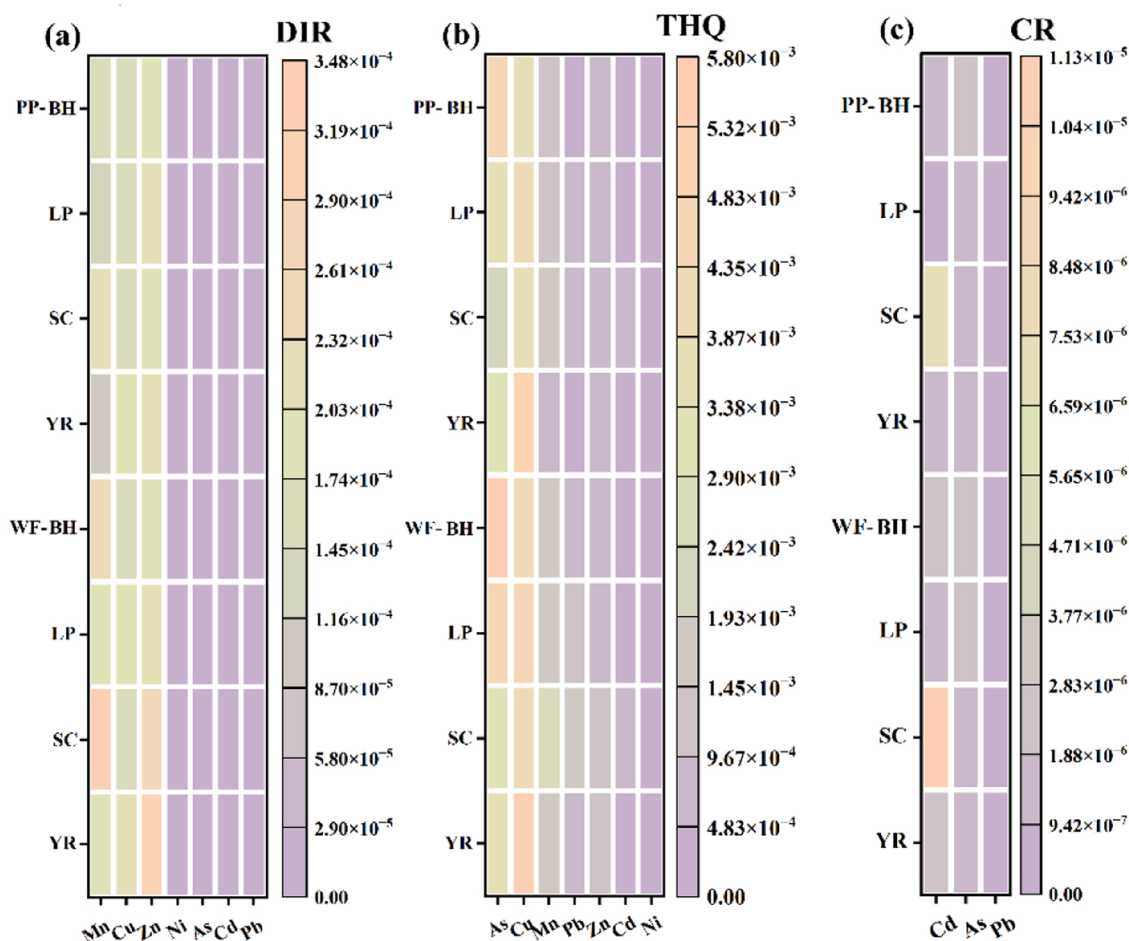


Fig. 4. The risk of Fuji apples from four regions (a: DIR value; b: THQ value; c: CR value).

3.2. Heavy metal contamination in Fuji apple

3.2.1. Comparison between different parts

The heavy metals in WF and PP samples of Fuji apple were analyzed by SPI and IPI. Since there is no ML for Mn in apple, this study only evaluated the pollution of the remaining six heavy metals. In both WF and PP samples, the SPI followed the order of Zn (0.102; 0.084) > Pb (0.082; 0.039) > Cu (0.035; 0.031) > Ni (0.025; 0.020) > Cd (0.013; 0.008) > As (0.005; 0.004) (Fig. 3). Sample with an SPI < 1 was identified as being uncontaminated with heavy metals (Xu et al., 2023). It could be seen that the SPI of WF and PP samples of Fuji apple was far below 1. In the WF samples, the SPIs of Zn and Pb were significantly higher than those of Ni, Cu, As, and Cd ($p < 0.01$), while the SPIs of Ni and Cu were significantly higher than those of Cd and As ($p < 0.01$). The SPI of Zn in WF samples was significantly higher than that of Pb ($p < 0.05$). In the PP samples, there was no significant difference in SPI between Cu and Pb or between As and Cd ($p > 0.05$). However, the SPIs of Zn and Pb in PP were significantly higher than that of Ni, Cu, As, and Cd ($p < 0.01$), while the SPIs of Ni and Cu remained significantly higher than that of Cd and As ($p < 0.01$). Notably, in PP samples, the SPI of Zn was significantly higher than that of Pb ($p < 0.01$). The average IPI of six heavy metals was 0.087 and 0.065 for WF and PP samples, respectively (Fig. 3). The IPI of WF samples was slightly higher than that of PP samples. All the samples had IPI ≤ 0.7 , indicating that these samples were at safe levels (Xu et al., 2023). Our results indicated that the levels of heavy metals in Fuji apple of China are generally low, which is similar with some other studies (Mawari et al., 2022; Gupta et al., 2018). In conclusion, both the SPI and IPI values of heavy metals in Fuji apples were all far below 1, demonstrating that neither WF nor PP samples

showed any heavy metal contamination.

3.2.2. Comparison between different producing areas

Heavy metal contamination content in Fuji apple samples from different producing areas was investigated using SPI and IPI. The SPI of WF and PP samples from four producing areas was all lower than 1, while the IPI was all lower than 0.7, both of which were at safe levels. Zn and Pb exhibited the two highest SPIs among the seven heavy metals analyzed in Fuji apples from the four main producing regions. In WF samples, the highest and lowest SPI were found for Zn (0.1) in Fuji apple from YR and As (0.003) in Fuji apple from SC, respectively. In PP samples, the highest and lowest SPIs were found for Zn (0.09) in Fuji apples from YR and Cd (0.02) in Fuji apples from LP, respectively (Fig. 3). The IPI of heavy metals in WF and PP samples ranged from 0.07 to 0.11 and 0.06 to 0.07, respectively. The IPI of heavy metals in Fuji apple from the four producing areas followed an order of SC > YR > LP > BH in WF samples, while LP > YR > SC > BH in PP samples (Fig. 3).

SPI analysis revealed distinct patterns of heavy metal accumulation in Fuji apple samples from SC, YR, LP, and BH regions (Fig. 3). SPI values in PP samples of Zn and As showed accumulation across regions with no significant differences ($p > 0.05$). The SPI values of Ni in WF and PP samples from LP and YR regions showed significant differences from BH and SC regions ($p < 0.01$). The SPI values of Cu in WF and PP samples revealed a single significant difference between SC and YR regions ($p < 0.05$). The SPI analysis revealed significantly higher Pb accumulation levels in both WF and PP samples from SC region compared to LP, BH, and YR regions ($p < 0.01$), while no significant differences were observed among the latter three regions. For Cd contamination, even more obvious regional disparities were found, with the SC region

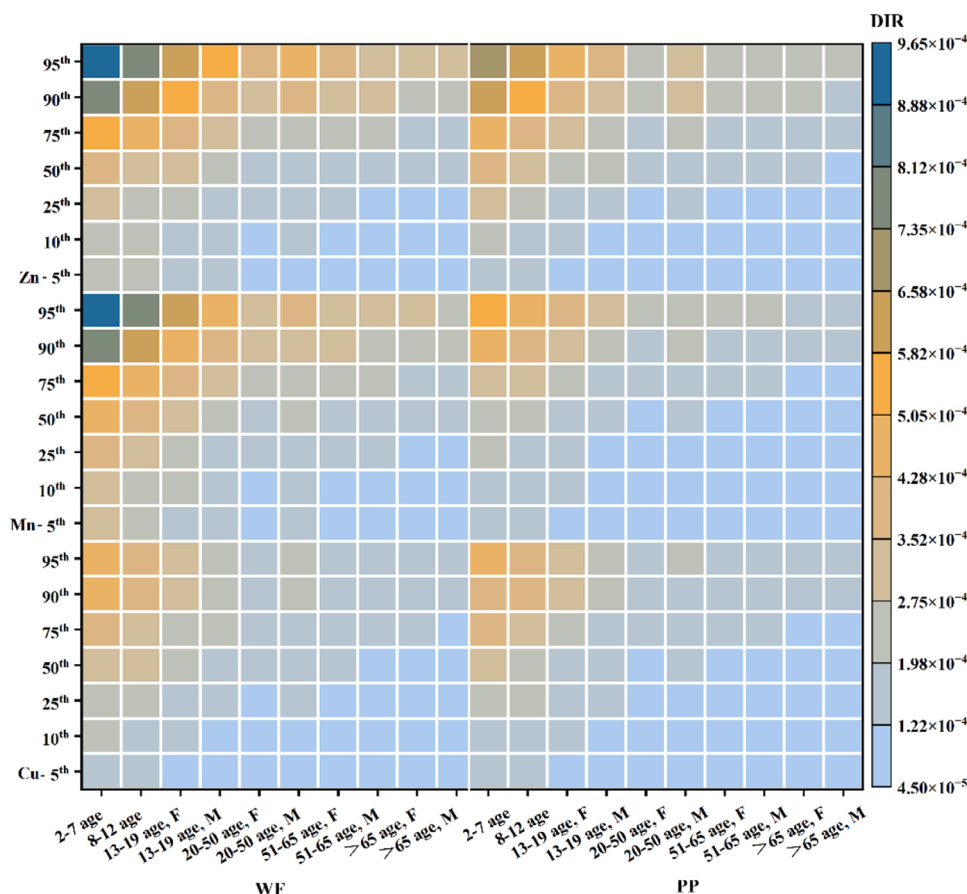


Fig. 5. DIR values of heavy metals in Fuji apples for different populations.

showing highly significant differences from all other regions ($p < 0.01$) (Fig. 3). These results clearly demonstrated that the SC region exhibited distinct accumulation characteristics for Cd and Pb compared to other regions. The observed variations in heavy metal accumulation might be attributed to soil composition, agricultural practices, or environmental factors across production regions.

3.3. Risk assessment of heavy metals in Fuji apple

3.3.1. Chronic risk assessment

Point estimation and probability estimation were investigated to estimate the level of risk posed by heavy metals in Fuji apple to different population groups. The probabilistic health risk assessment of each heavy metal was carried out by the 5th, 10th, 25th, 50th, 75th, 90th, and 95th percentile of its content in Fuji apple (Xu et al., 2023). To test the health risks of heavy metals in Fuji apple to consumers, the DIR, THQ, TTHQ, and CR of heavy metals were assessed through point assessment and probabilistic assessment. The DIR for WF samples followed the order of Mn (2.53×10^{-3}) > Zn (2.49×10^{-3}) > Cu (1.84×10^{-4}) > Ni (3.57×10^{-6}) > As (1.26×10^{-6}) > Pb (2.87×10^{-7}) $\approx$ Cd (2.87×10^{-7}), and that for PP samples followed the order of Zn (2.00×10^{-4}) > Cu (1.66×10^{-4}) > Mn (1.55×10^{-4}) > Ni (2.81×10^{-6}) > As (9.95×10^{-7}) > Pb (1.77×10^{-7}) $\approx$ Cd (1.77×10^{-7}) (Fig. 4 a). The DIR of WF samples was about 1.3-fold that of PP samples. Heavy metals tend to accumulate in fruit peels, and direct consumption leads to higher exposure levels. Peeling can effectively remove a portion of these heavy metals, particularly those concentrated in the peel (Li et al., 2023). The DIR values of the seven heavy metals were overall higher in SC, followed by YR, BH, and LP. The DIR of Mn and Zn in WF samples from SC and YR was 3.47×10^{-4} and 2.91×10^{-4} , respectively, which were higher than two producing areas (Fig. 4 a). The mean values of estimated DIR

corresponding to their tolerance values were 0.1636 % (WF) and 0.1342 % (PP) for LP, 0.1806 % (WF) and 0.1501 % (PP) for BH, 0.1680 % (WF) and 0.1360 % (PP) for YR, and 0.1715 % (WF) and 0.1279 % (PP) for SC. These data indicated that apple from these four producing areas does not pose a threat to the health of consumers.

The DIR value of seven heavy metals in apple decreased with the age of consumers (Fig. 5). The DIR value of heavy metals for the group aged 2–7 years of apple consumption was about 3.17-fold that for the group aged 65 years. Previous a study showed that children and adolescents demonstrate 30 %–50 % higher fruit consumption than adults due to developmental requirements, resulting in significantly elevated heavy metal exposure risks ($p < 0.01$) (Smith et al., 2021). Another study showed that children exhibit 2.1–3.3 times higher weight-normalized DIR of heavy metals compared to adults due to their lower body weight (Barr et al., 2022). The combination of growth requirements, developmental needs, and lower body weight significantly increased heavy metal exposure risk from apple consumption in children aged 2–7 years ($p < 0.05$). Conversely, age-related changes in masticatory function, metabolic rate, and taste preferences significantly reduced exposure risk in elderly populations (≥ 65 years) ($p < 0.05$) (Barr et al., 2022; Johnson et al., 2021; Smith, et al., 2021). Apple intake in adult males was 25 % higher than in females ($p < 0.05$), corresponding to an 18 %–22 % increase in Cd and Pb intakes (Zhang et al., 2022). DIR values for all seven heavy metals in apples were higher in males than females in this study (Fig. 5), which was attributed to higher basal metabolic rate, fruit consumption, and body weight in males (Table S4) (Zhang et al., 2022).

The THQ values of the seven heavy metals in PP samples for different population groups followed the order of Cu > As > Mn > Zn > Pb > Cd > Ni, which was generally consistent with the order observed in WF samples (Fig. 6). For children aged 2–7 years, the 95th percentile of the

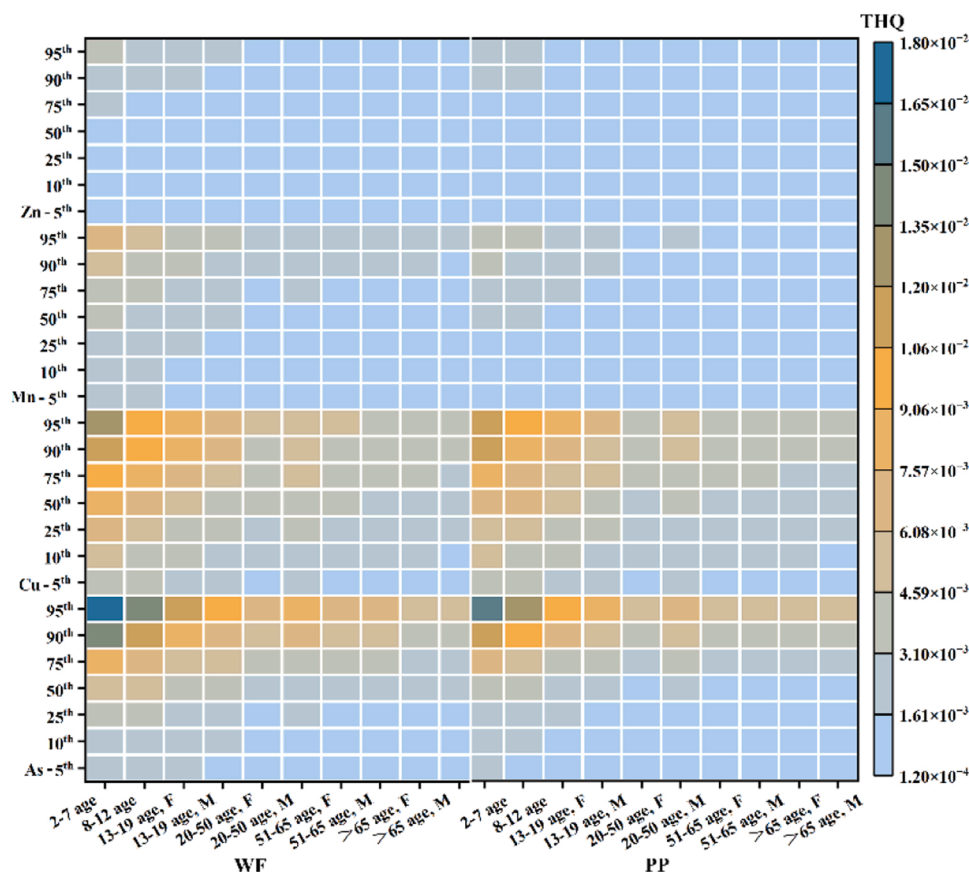


Fig. 6. THQ values of heavy metals in Fuji apples for different populations.

THQ values for seven heavy metals in Fuji apples ranked as As (WF: 1.79×10^{-2} ; PP: 1.58×10^{-2}) > Cu (WF: 1.26×10^{-2} ; PP: 1.20×10^{-2}) > Mn (WF: 6.45×10^{-3} ; PP: 4.04×10^{-3}) > Zn (WF: 3.21×10^{-3} ; PP: 2.38×10^{-3}) > Pb (WF: 1.80×10^{-5} ; PP: 8.22×10^{-6}) > Ni (WF: 1.29×10^{-5} ; PP: 1.37×10^{-5}) > Cd (WF: 1.76×10^{-6} ; PP: 1.50×10^{-6}). The THQ values of the four heavy metals in Fuji apples showed a declining trend as consumer age increased, with females consistently exhibiting higher values than males. This trend is different with that observed for the DIR values. Notably, the THQ values for Cd, Ni, and Pb in Fuji apples from the four producing areas were below 1.2×10^{-4} and therefore were not displayed in Fig. 6. The THQ values for As, Mn, and Cu in WF samples from the BH, SC, and YR regions were 1.64×10^{-6} , 3.47×10^{-4} , and 2.19×10^{-4} , respectively; which were higher than those in the other three regions and like the values observed in PP samples (Fig. 4 b). Similarly, significant variations in TTHQ values were observed between PP and WF samples collected from different geographical regions. For WF samples, the TTHQ values followed the order of SC (1.36×10^{-2}) > BH (1.35×10^{-2}) > LP (1.25×10^{-2}) > YR (1.24×10^{-2}), while for PP samples, the order was BH (1.1×10^{-2}) > LP (1.0×10^{-2}) > YR (9.9×10^{-3}) > SC (9.5×10^{-3}) (Fig. 7). Overall, the THQ and TTHQ values of WF samples were greater than those of PP samples, indicating a higher potential health risk associated with the consumption of whole apples compared to peeled apples.

3.3.2. Carcinogenic risk assessment

Cr^{3+} and Cr^{6+} are the most important forms of Cr in the environment and biological systems. Cr^{6+} has attracted significant attention due to its high toxicity and carcinogenicity, while Cr^{3+} is relatively safe and an essential trace element for humans. Cr in apples primarily exists as Cr^{3+} , which poses a relatively low carcinogenic risk (Zhitkovich, 2011). However, in cases of Cr^{6+} contamination, the CR should be assessed using the CSF (EPA IRIS Database, 2023). ICP-MS primarily measures

total elemental concentrations and cannot directly differentiate valence states (e.g., Cr^{3+} and Cr^{6+}) (Michalski et al., 2011). Furthermore, sample pretreatment procedures may alter the original valence states. Therefore, this study did not conduct a risk assessment for Cr species in Fuji apples. The CR was classified as very low ($\text{CR} \leq 1 \times 10^{-6}$), low ($1 \times 10^{-6} \leq \text{CR} \leq 1 \times 10^{-4}$), intermediate ($1 \times 10^{-4} \leq \text{CR} \leq 1 \times 10^{-3}$), high ($1 \times 10^{-3} \leq \text{CR} \leq 1 \times 10^{-1}$) and very high ($\text{CR} \geq 1 \times 10^{-1}$). CR is higher than 1×10^{-6} , indicating that the risk to humans is unacceptable (Xu et al., 2023). Fuji apple from BH had the highest CR value of As (WF: 2.46×10^{-6} ; PP: 2.08×10^{-6}), and the CR values of Pb in Fuji apples from four producing areas was all lower than 1.0×10^{-6} . The CR of As in the WF samples from RY (1.61×10^{-6}) and SC (1.48×10^{-6}) was all low risk level. The CR value for Cd in Fuji apples from SC was significantly higher than that from other producing areas (Fig. 4 c).

This study systematically assessed the CR of As, Pb, and Cd in WF and PP for different age groups. Probabilistic assessment indicated that the order of risk contribution in all age groups at the 5th–90th percentile was Cd (WF: 5.48×10^{-6} ; PP: 3.48×10^{-6}) > As (WF: 1.89×10^{-6} ; PP: 1.50×10^{-6}) > Pb (WF: 3.77×10^{-8} ; PP: 1.75×10^{-8}) (Fig. 8). For children aged 2–7 years old, the CR value of Pb in WF (1.53×10^{-7}) was 2.2 times higher than that in PP (6.99×10^{-8}). Similarly, the CR value of Cd in WF (3.18×10^{-5}) was 41 % higher than that in PP (2.25×10^{-5}) (Fig. 8). Notably, CR demonstrated a clear declining trend with increasing age. Relative to pediatric groups, 66 %–70 % decrease in CR values for Pb and Cd exposure through Fuji apple consumption among elderly individuals (≥ 65 years).

Gender disparity analysis revealed higher CR value of As in females (5.37×10^{-6}) compared to males (4.32×10^{-6}) aged 13–19 groups. While males aged 20–50 group showed elevated CR values (3.73×10^{-6}) relative to females (3.05×10^{-6}) (Fig. 8). Notably, gender-based differences in CR values diminished significantly in elderly populations. For instance, in the 2–7-year-old age group, the CR value of As in WF

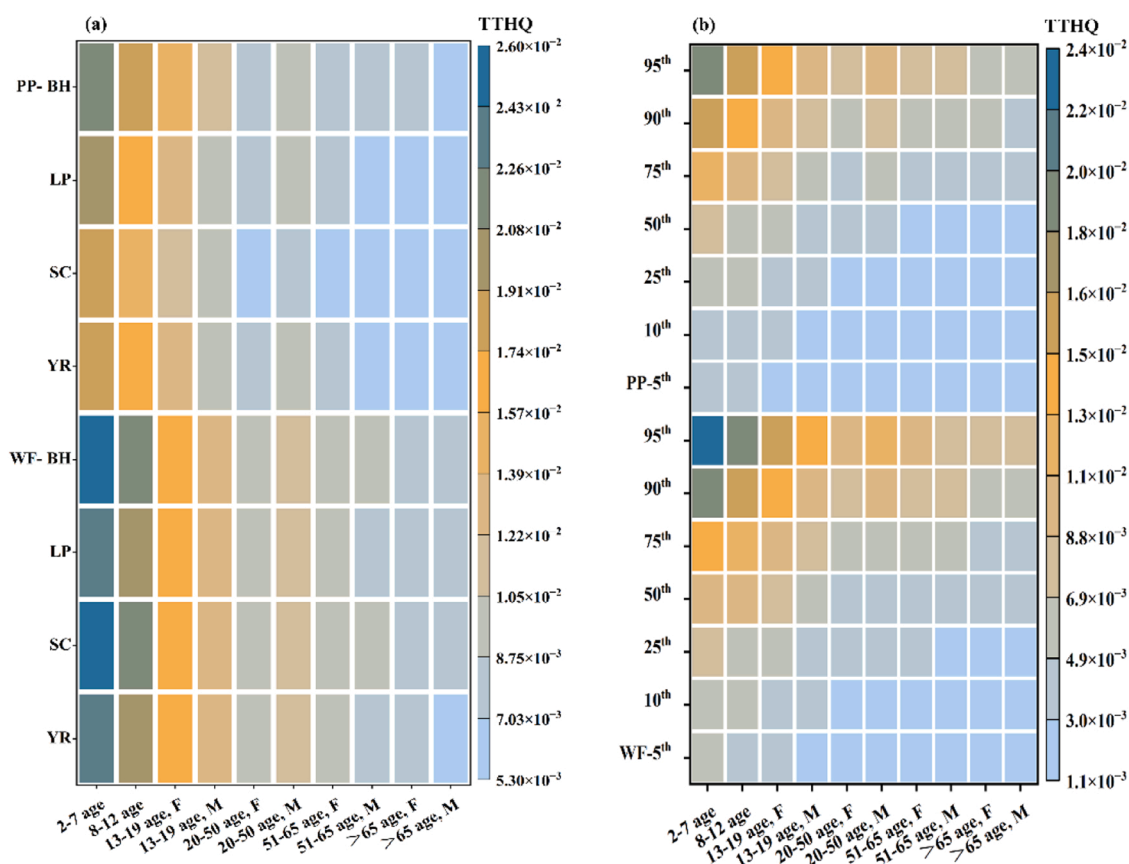


Fig. 7. TTHQ values of heavy metals in Fuji apples of different regions (a) and different parts of Fuji apples (b). (Note: F stands for female; M stands for male).

samples reached 8.06×10^{-6} , indicating higher exposure risk in children. In contrast, elderly individuals (≥ 65 years) exhibited substantially lower CR values of -2.71×10^{-6} for females and 2.38×10^{-6} for males, which indicated a pronounced age-dependent reduction in arsenic-associated health risk (Fig. 8). This result showed that the CR values of Cd, As, and Pb for adults was all lower than 1×10^{-6} , suggesting that normal consumption of Fuji apple from these producing areas would not cause adverse effect on adult health. Children exhibited the highest susceptibility to heavy metals from Fuji apple. Since peeled apple could only reduce the CR of heavy metal by 10 %–45 %, we recommend promoting peeled apple, especially for vulnerable groups like children.

4. Discussion and conclusion

The primary sources of heavy metal contamination in Fuji apples include the natural soil background, atmospheric deposition, irrigation with wastewater, and the discharge of industrial three-wastes, mine wastewater pollution, mine waste dumping, the use of agricultural inputs (such as fertilizers, metallic pesticides [e.g., Cu or Zn-based formulations], and agricultural plastic film), etc. (Nie, 2020). Agricultural plastic films incorporate heavy metals through the use of heat stabilizers. These multiple-source pollutants are ultimately absorbed by apple trees through the three primary environmental media (soil, air, and water) by the routes of soil-to-tree transfer, foliar uptake, and direct atmospheric deposition (Nie, 2020). Root systems activate heavy metals in soil by secreting organic acids and other compounds, enabling their entry via the plant root uptake and transport pathway (Li et al., 2025). Heavy metal ions are actively loaded into the xylem vessels and transported upward to stems, leaves, and fruits via transpiration-driven flow (Uraguchi & Fujiwara, 2012). Atmospheric heavy metal particulates or solutions can directly enter leaf tissues through stomata or slowly permeate the cuticle via hydrophilic micropores after dissolution

(Schreck et al., 2012). Young or damaged leaves exhibit heightened susceptibility to such absorption. After being absorbed by the leaves, heavy metals may accumulate in mesophyll cells, while mobile elements can further transfer to the phloem and translocate to edible organs along with products of photosynthetic products (Uraguchi et al., 2017). Based on the above mechanisms, a multi-layered mitigation strategy can be formulated. Orchards can deploy multiple strategies to control heavy metal pollution. Key measures include choosing locations with certified environmental quality, selecting fertilizers that comply with relevant standards for heavy metal content, adopting fruit bagging and washing, topsoil replacement, and applying chemical, biological, and organic amendments (Nie, 2020).

This study determined the levels of seven heavy metals (Cd, Pb, Ni, Cu, Zn, As, and Mn) in Fuji apple samples from four main producing areas in China. The content of each heavy metal in WF was significant differences than that in PP. The heavy metal contents in either WF or PP of Fuji apple from the four producing areas followed the order of SC > YR > BH > LP. None of the samples exceeded the MLs for the studied seven heavy metals. Both SPI and IPI were at safe levels, and the DIR was well below the tolerance level. Additionally, the THQ and TTHQ for all population groups were all lower than 1, indicating that the consumption of Fuji apple from these producing areas poses no threat to human health. The health risks associated with heavy metal exposure exhibited significant variations depending on demographic characteristics and tissue distribution. The risk level decreased with age, and the values of risk to male groups in the same age group were lower than those to females. The risk levels of each element in WF samples were higher than those in PP samples. In a whole, the analyses of DIR, THQ, TTHQ, and CR for the seven heavy metals demonstrated that normal consumption of Fuji apples from these production regions is unlikely to cause any harmful effects on human health.

The results of this study indicate that the current heavy metal

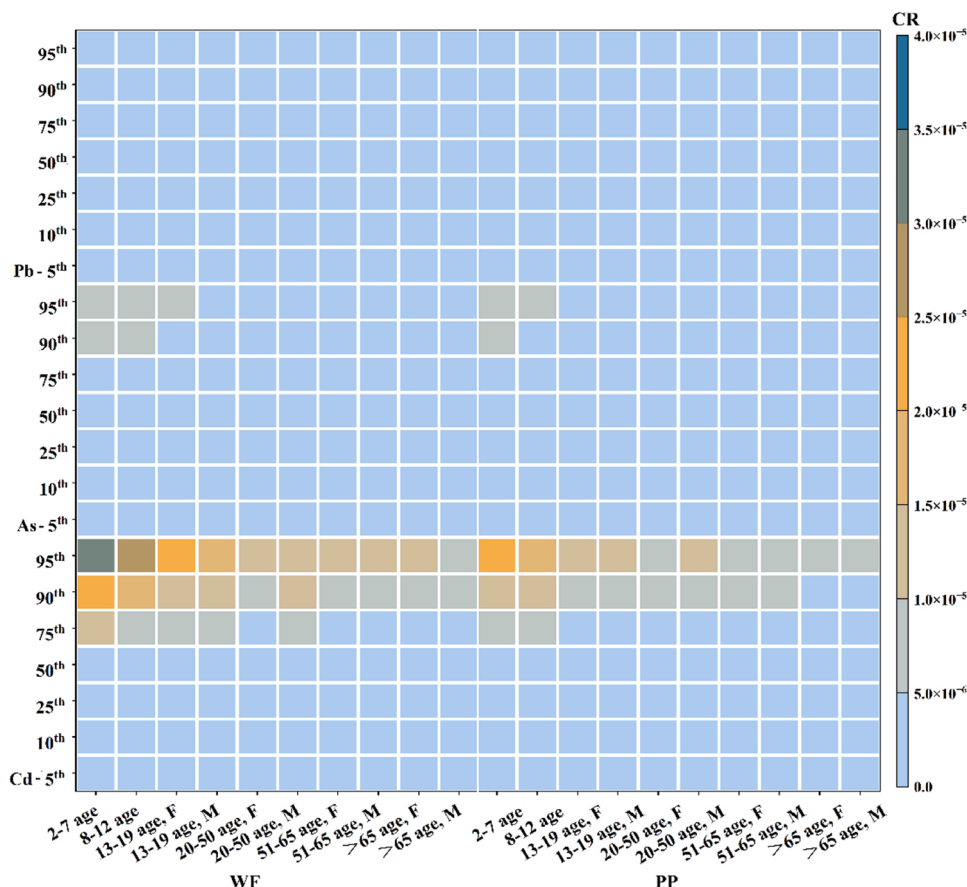


Fig. 8. CR values of heavy metals in Fuji apple for different populations. (Note: F stands for female; M stands for male).

contamination status of Fuji apples from the four major production regions is generally safe, with negligible health risks to consumers. Nevertheless, as a preventive strategy to safeguard against future contamination, it is prudent to implement the following measures during production. It is recommended that orchard should be situated away from pollution sources (such as industrial "three wastes"), with heavy metal content in its soil, irrigation water, and ambient air complying with national standards. These strategies provide a scientific basis and practical guidance for ensuring the sustainable development of the apple industry and consumer safety. Furthermore, consumers can adopt scientific washing methods, such as soaking and washing apples with solutions of baking soda, salt, and vinegar (Nie, 2020). This practice has been proven to effectively reduce heavy metals on the fruit surface. It serves as an effective final barrier to safeguard dietary safety.

CRediT authorship contribution statement

Peng Jiang: Writing–review & editing, Writing–original draft, Resources, Formal analysis, Data curation, Conceptualization. **Mingyu Liu:** Writing –review & editing, Writing–original draft, Visualization, Validation, Formal analysis, Data curation, Conceptualization. **Yihui Liu:** Writing– review & editing, Writing–original draft, Visualization, Validation, Supervision, Formal analysis, Data curation, Conceptualization. **Yishan Wei:** Writing–review & editing, Writing–original draft, Visualization, Validation, Formal analysis, Data curation, Conceptualization. **Jiyun Nie:** Writing–review & editing, Writing–original draft, Visualization, Validation, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation. **Mengying Shuai:** Writing–review & editing, Writing–original draft, Visualization, Validation, Formal analysis, Data curation, Conceptualization. **Xiabing Kong:** Writing–review & editing,

Writing–original draft, Visualization, Formal analysis, Data curation, Conceptualization. **Min Xu:** Writing–review & editing, Writing–original draft, Visualization, Validation, Formal analysis, Data curation, Conceptualization. **Zhichao Li:** Writing–review & editing, Writing–original draft, Visualization, Validation, Data curation, Conceptualization. **Xiaoming Li:** Writing–review & editing, Writing–original draft, Visualization, Validation, Formal analysis, Data curation, Conceptualization. **Kunyu Wang:** Writing–review & editing, Writing–original draft, Visualization, Validation, Formal analysis, Data curation, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jfca.2025.108482](https://doi.org/10.1016/j.jfca.2025.108482).

Data availability

Data will be made available on request.

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