

# Heavy metal levels, potential health risk, and uncertainty analysis in a plant–soil–irrigation system of the Yellow River irrigation area of northern Ningxia (China)

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## Research article

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## Abstract

Background Industrial development results in elevated levels of heavy metals in the local environment, including the air, soil, and water. These heavy metals can contaminate crops in the surrounding area, which may pose severe health risks to local inhabitants. The aims of this study were to determine the levels of heavy metals in plant–soil–irrigation system and the associated human health risk with deterministic and probabilistic approaches. Methods In this study, samples of soil, irrigation water, and maize crops were collected from the Yellow River irrigation area in northern Ningxia, China. Inductively coupled plasma-atomic emission spectrometry(ICP-AES)was applied to determine the heavy metal contents in maize grains, soil samples, and irrigation water. Potential health risks were assessed by deterministic and probabilistic estimation. Results The average concentrations of chromium and lead in maize exceeded the maximum allowable concentrations in food. The average concentrations of metals in the associated soil and irrigation water were both below the safe limits allowed in China. Deterministic estimation indicated a hazard index of 0.0986 for all inhabitants, implying no significant non-carcinogenic risk. The lifetime cancer risk value was  $3.261 \times 10^{-5}$ , lower than the maximum acceptable level of  $1 \times 10^{-4}$  suggested by USEPA, while above the negligible level of  $1 \times 10^{-6}$  (USEPA) and  $1 \times 10^{-5}$  (WHO), with females facing a greater health risk than males. Probabilistic estimation indicated that approximately 0.62% of inhabitants are exposed to non-carcinogenic risk due to maize ingestion, while carcinogenic risk exceeds the maximum acceptable level ( $1 \times 10^{-4}$ ) for 8.23% and negligible level ( $1 \times 10^{-5}$ ) for 64.26% of inhabitants. Sensitivity analysis indicated that the concentration of arsenic in maize, the daily intake of maize, and exposure frequency of maize are the primary contributing factors in both non-carcinogenic and carcinogenic risks. Therefore, the content of arsenic in maize is of concern in the study region. Conclusions Based on deterministic and probabilistic risk estimation, there are no obvious non-carcinogenic health risks to inhabitants, while carcinogenic risk from As exposure is higher than the acceptable risk level. Females are at greater risk than males, and inhabitants under 20 years of age have the highest risk among age groups.

## Background

As a result of rapidly increasing population growth, industrial and commercial development, and accelerated urbanization, environmental contamination has become a serious problem in developing countries [1]. Heavy metals are an important type of contaminant that can accumulate in the environment from sources such as mining, pesticides, and chemical fertilizers [2]. In China, the environment is more heavily contaminated by heavy metals in regions with higher degrees of industrialization. Industrial wastes such as waste water, waste residue, and flue gas can affect the surrounding agricultural land, water, and air. In a previous study, the heavy metal concentration in 29.4% of soil samples collected from 2,523 industrial parks exceeded the standard for soil environmental quality [3]. Heavy metals may accumulate until they reach toxic levels that harm the quality of human life [4–6]. For example, cadmium (Cd) exposure can cause adverse health effects, including damage to the lung, liver, testicles, brain, bone, and blood system along with cancer; Cd can accumulate for 10 to 20 years in the human body and is considered one of the most toxic heavy metals [7–10]. The main toxic effects of lead (Pb) are neurological effects, especially intelligence quotient (I. Q.) deficits [11]. In children, Pb may cause behavioral disturbances along

with learning and concentration difficulties [12]. While arsenic (As) is a metalloid, it is considered as a metal in many studies because it behaves like a heavy metal in many aspects. When As enters the body, it is primarily deposited in the hair, bones, and other organs, and it can lead to disorders of the respiratory, nervous, and circulation systems along with cancer [13–14]. Although some heavy metals including copper (Cu), zinc (Zn), and chromium (Cr) are considered essential micronutrients at low concentrations [15–16], they can pose non-carcinogenic hazardous effects on human health when present at concentrations exceeding the tolerable doses [17].

Ningxia Province is a typical developing region in China. Northern Ningxia Province is an important industrial area that is home to several industrial parks, and untreated emissions have resulted in environmental pollution. In recent years, researches on the contamination status of this area have been conducted. The groundwater and soil in this area have been polluted to some extent by multiple heavy metals. For example, the soil is seriously contaminated with Cd, with Cd concentrations exceeding the standard level, and the concentrations of Zn, Pb, Cu, and Cr in soil are higher than the background values for Ningxia [18]. However, the soil samples analyzed in Ref. 18 were collected from industrial parks; the surrounding agricultural soil was not evaluated. Since crops could be strongly affected by metals in the soil, it is necessary to determine the concentrations of heavy metals in agricultural soil. The Yellow River is the second largest river in China and provides the drinking water, domestic water, and agricultural irrigation water for the regions along the river. However, rapid population growth and industrial development have resulted in the direct discharge of pollutants into the river, including metals, causing the water quality to deteriorate [19–20]. For example, filtered water from the Yellow River in the Ningxia area was reported to be severely polluted by Cr [21].

The consumption of food grown in local fields contaminated with heavy metals presents a health risk for local inhabitants [22]. Risk assessments have been performed using various indices, including the hazard quotient (HQ) [23–24], hazard index (HI) [25–26], and morbidity status (MS) [27]. While numerous studies have been carried out in developed areas of China [5, 28–30], much less attention has been paid to developing regions such as Ningxia province. Therefore, it is necessary to evaluate the potential health risks to local inhabitants resulting from heavy metal contamination in developing regions.

The present study was carried out in northern Ningxia, China, where the Yellow River has been used to irrigate crops for several decades. Maize (*Zea mays* L.) is the most widely grown crop in the study area. The objectives of this study were to (1) determine the levels of six common heavy metals (Cd, Pb, Cr, Zn, Cu, and As) in maize along with the associated soil and irrigation water; (2) determine the daily consumption of maize by local inhabitants; (3) evaluate the hazardous health effects resulting from heavy metals exposure by maize consumption; and (4) analyze the main factors affecting health risk.

## Methods

### *Sample collection*

The study area was located in Northern Ningxia, China, which contains many industrial parks. Water from the Yellow River in this area is used to irrigate the surrounding agricultural fields. 42 fresh, raw maize samples were collected from the cultivated area in August 2017, and the map of sampling sites was shown in Fig. 1. In addition, soil from each maize cultivated area was also sampled. The maize and soil samples were packed, labeled and immediately transported to the laboratory. Five samples of irrigation water were collected from the Yellow River near the study area. Each water sample was collected in a polypropylene bottle, and 1 mL of concentrated nitric acid ( $\text{HNO}_3$ ) was added to each water sample to eliminate microbial activity. The samples of maize and soil were collected by the permission of the peasant household, while irrigation water can be collected without any permission.

*Fig. 1*

#### *Chemical analysis*

The maize samples were dried in an oven (DHG-9030A, China) at 60°C and powdered using a grinding mill (0.2-mm sieve). The sifted samples (0.5 g) were weighed into digestion tubes, and 10 mL of digestion solution ( $v:v$ ,  $\text{HNO}_3:\text{HClO}_4 = 4:1$ ) was added. After cold-digestion overnight, the mixture was further digested with a block digester at 120°C until the solution was clear. The digested samples were then diluted to a volume of 10 mL with Milli-Q water. The soil samples were air-dried until reaching a constant weight and powdered using a grinding mill (0.2-mm sieve). Subsequently, 12 mL of nitric acid:hydrofluoric acid ( $v:v$ ,  $\text{HNO}_3:\text{HF} = 5:1$ ) was added to 0.35 g of each soil sample in a Teflon digestion tube. The mixture was heated at 120°C until the solution volume reached approximately 3 mL. Next, 5 mL of perchloric acid was added to continue the digestion until the solution was clear. The acid in the sample was evaporated at the same temperature until 1–2 mL of solution remained. The solution was then transferred to 50 mL colorimetric tube and diluted to a final volume of 50 mL with water. The samples of irrigation water were filtered with 0.45- $\mu\text{m}$  filters and then 20 mL of each filtered solution was digested with 5 mL nitric acid. In the last step, digested samples were diluted to 10 mL with Milli-Q water.

The contents of heavy metals (Cd, Pb, Cr, Zn, Cu, and As) were performed by ICP-AES (Varian710-ES, USA). That each analyses was performed in triplicate, blank reagents and standard reference reagents were used in each batch, and recoveries of the metals were tested to make sure the accuracy and precision of the experiment procedures.

#### *Questionnaires on maize consumption*

To determine the maize consumption habits of residents, questionnaires were distributed in villages close to the sampling sites. A total of 103 local inhabitants were randomly selected to complete the questionnaire considering age distribution and gender balance. The questionnaire include information about their age, gender, body weight, frequency and quantity of maize consumption, and maize source. The maize consumption information for residents was used to realize health risk assessment.

#### *Health risk assessment*

## Deterministic estimation of health risk

Among the six metals considered, Cd, Pb, Cr, Zn, and Cu pose non-carcinogenic health risks through oral exposure, while As poses both non-carcinogenic and carcinogenic health risks upon oral exposure. The non-carcinogenic effect of an individual metal could be evaluated by HQ value calculated using Eq. (1) [30]:

$$HQ = EXPO/RfD, \quad (1)$$

where EXPO is daily exposure to metals (mg/(kg·day)), and RfD is the reference dose (mg/(kg·day)) suggested by the United States Environmental Protection Agency (USEPA) or World Health Organization (WHO). To evaluate exposure to two or more metals, HI [Eq. (2)] value was used to evaluate the total non-carcinogenic health risk [31]:

$$HI = \sum^1_n HQ_n. \quad (2)$$

If HQ or HI is less than 1, no obvious non-carcinogenic risk exists. EXPO was determined using Eq. (3) [32]:

$$EXPO = (C \times DI \times EF \times ED) / (BW \times LT), \quad (3)$$

where  $C$  (mg/kg) represents concentration of heavy metals in maize; DI (g/day) is the daily intake of maize; EF (day/year) is the exposure frequency determined from the questionnaire; ED (year) is the exposure duration; BW (kg) is the body weight of residents determined from the questionnaire; and LT (year) is the lifetime of residents in days (presumed to be 70 years).

The carcinogenic risk ( $R$ ) caused by As was determined using Eq. (4) [32]:

$$R = SF \times EXPO. \quad (4)$$

The value of SF suggested by the USEPA is  $1.5 \text{ (mg/kg/day)}^{-1}$  (USEPA, 2010). The negligible carcinogenic risk level suggested by the USEPA is  $10^{-6}$ , while the level set by the WHO is  $10^{-5}$ , and the maximum acceptable level suggested by USEPA is  $10^{-4}$  [33].

## Probabilistic estimation of health risk

To assess uncertainty and variability in risk assessment, a probabilistic estimation was performed using Monte Carlo technique because the deterministic estimation only provides the mean value of population exposure, which cannot accurately estimate the exposure of the population. Monte Carlo technique was performed to calculate the distribution of exposure and the health risk of the population.

Probabilistic distributions of the health risk were generated by inputting the variability in exposure factors. From the chemical analyses and questionnaires, the distributions of parameters (like concentration of metals, daily intake of maize, exposure frequency, and body weight, and so on) were determined. The best-fitting distribution for each variable was determined by fitting a number of parametric distributions (e.g.,

lognormal, gamma, and Weibull). Anderson–Darling (AD) test combined with other tests was used to determine the goodness-of-fit for each distribution. The process was realized using Oracle<sup>©</sup> Crystal Ball software.

The probabilistic estimation of health risks, which was used Monte Carlo technique in Crystal Ball software was based on the 10<sup>th</sup>, 50<sup>th</sup>, and 90<sup>th</sup> values. In the present study, Monte Carlo simulation was allowed to run for 10,000 iterations by drawing parameter values randomly from the distribution functions obtained before. Finally, the proportion of the population exceeding the acceptable health risk level was calculated.

### *Sensitivity analysis*

Sensitivity analysis was performed in order to confirm which variables pose the greatest effect on health risk in Crystal Ball software. First, the rank correlation coefficients between the exposure factors and health risk were determined using probabilistic estimation. Subsequently, the contribution of each variable was calculated by squaring the variance. Finally, the results were normalized to 100%, and the sequence of contributing variables was generated.

### *Statistical methods*

Mean and standard deviation (SD) were calculated in Microsoft Excel 2010. The determination of the best-fitting distribution, Monte Carlo simulation, and sensitivity analysis were all carried out in Crystal Ball software.

## **Results**

### *Heavy metal levels in maize, associated soil, and irrigation water*

The heavy metal concentrations in maize grains, associated soil, and irrigation water are presented in Table 1. The average concentrations of Cd, Pb, Cr, Zn, Cu, and As in maize grains were 0.037, 0.41, 2.36, 17.02, 1.04, and 0.17 mg/kg, respectively. The maximum allowable concentrations of these six metals in food are 0.1 (Cd), 0.2 (Pb), 1.0 (Cr), 50 (Zn), 10 (Cu) and 0.5 (As) mg/kg (GB 2762–2017, NY 861–2004) [34,35]. Thus, the average concentrations of Pb and Cr in this study exceeded the maximum allowable concentrations in food. However, the concentrations of four metals exceeded the standards in some samples. The measured metal level exceeded the limit in the highest percentage of samples for Cr (69%) followed by Pb (47%), As (18%), and Cd (2%). The average heavy metal concentrations in the soil samples were 0.14, 18.16, 37.25, 138.20, 19.61, and 14.18 mg/kg for Cd, Pb, Cr, Zn, Cu, and As, respectively, with large variation among samples. The average Cd and Zn concentrations exceeded the standards (GB15618–1995) for soils in China [36]. The background concentrations in Ningxia were 0.11 (Cd), 20.6 (Pb), 60.0 (Cr), 58.8 (Zn), 22.1(Cu) and 11.90 (As) mg/kg [37]. Thus, the Zn level in soil measured in this study was significantly higher than the background value, while the Cr level was lower, and the levels of other metals were similar to the background values. Some sampling sites in this study were close to industrial parks, which might explain the high metal concentrations found at these sites. However, source identification should be conducted to better understand the results. The transfer factor (TF) is an index for

evaluating the transfer potential of a metal from soil to plant [38]. TF reflects the concentration of metal in maize relative to that in the corresponding soil. A TF value higher than 1 indicates a high level of metal accumulation in the plant [39]. In this study, the TF values were far less than 1 for all six heavy metals, indicating low levels of metal accumulation in maize grains. In irrigation water samples, the mean concentrations of the six heavy metals were all far below the limits acceptable in China (GB 5084–2005) [40], and none of the measured concentrations exceeded the standards.

*Table 1*

*Body weight, daily consumption of maize, and exposure frequency of maize*

The results for resident BW, DI, and EF are summarized in Table 2. Of the 103 local inhabitants that participated in this study, 49% were male, and 51% were female. The participants were classified into four age groups: under 20, 20–40, 40–60 (including 40), and over 60 years old (containing 25%, 34%, 26%, and 15% of participants, respectively). The average BWs of all participants, male participants, and female participants were 54.60, 58.45, and 50.67 kg, respectively. Based on reported statistics, the average BW for adults in China is 62.7 kg for males and 54.4 kg for females [41]. The average BWs in this study was slightly lower than the reported values, possibly because nearly half the participants in the under 20 age group were children (less than 14 years old).

In this study, the maize DI was slightly less for males (160.20 g/day) than for females (162.08 g/day). Among the different age groups, DI was the highest (186.37 g/day) in the 20–40 age group followed by the 40–60 (181.48 g/day) and >60 (136.36 g/day) groups. The <20 group had the lowest DI of 118.46 g/day. Maize EF was also lower for males (14.34 day/year) than for females (16.67 day/year). However, unlike DI, EF was highest in the <20 age group (17.81 day/year) followed by the 40–60 (17.11 day/year) and 20–40 (14.06 day/year) age groups. The >60 group had the lowest EF of 10.36 day/year. BW, DI, and EF all affect health risk, and their degrees of influence were evaluated by sensitivity analysis.

*Table 2*

*Human health risk assessment*

Deterministic estimation

The oral RfD values were established as 1, 1500, 300, 40, and 0.3 µg/kg/day for Cd, Cr, Zn, Cu, and As, respectively [17]. The RfD value for Pb was 3.57 µg/kg/day according to the provisional tolerable weekly intake level (25 µg/kg/week) [42] set by the WHO.

The results of the deterministic estimation of health risk are shown in Table 3. The HQ values indicate that no individual metal posed a significant non-carcinogenic risk (HQ < 1). For all inhabitants, the non-carcinogenic health risk posed by the different metals decreased in the following order: As > Pb > Zn > Cd > Cu > Cr. The combined non-carcinogenic health risk HI values were also less than 1, indicating that maize consumption is not associated with an obvious non-carcinogenic health risk due to heavy metals. The HI

values for females were greater than those for males, implying that females experience more potential non-carcinogenic health risks from heavy metals than males. This can be attributed to the higher DI and EF but lower BW of females compared to males. Among the different age groups, HI decreased in the following order: under 20 > 40–60 > 20–40 > over 60. Similarly, there was one study indicated that children are at a greater health risk than adults from the consumption of an individual metal in maize, for example, HI caused by Cd for adults is  $8.5 \times 10^{-2}$  while for children is  $1.3 \times 10^{-1}$  [43].

In terms of carcinogenic risk caused by As, the average *R* value for all participants was  $3.261 \times 10^{-5}$ , lower than the maximum acceptable carcinogenic level set by the USEPA ( $10^{-4}$ ) but higher than the negligible risk levels set by the USEPA ( $10^{-6}$ ) and WHO ( $10^{-5}$ ). Among age groups, the *R* values decreased in the same order as the HI values: under 20 > 40–60 > 20–40 > over 60 years old.

Table 3

#### Probabilistic estimation

The best-fitting distributions of exposure factors were determined using Crystal Ball software. The concentrations of metals in maize were fitted to lognormal distributions with the exception of Cu concentration, which was fitted to a beta distribution. Similarly, a previous study found that the concentrations of most contaminants in the environment follow lognormal distributions [44]. All DI, EF, and BW values were fitted to lognormal distributions with the exceptions of the BWs of males (Poisson) and females (negative binomial).

The results of the probabilistic estimation of health risk are summarized in Table 4. For non-carcinogenic risk, all HI values were fitted to lognormal distributions. The 10<sup>th</sup>, 50<sup>th</sup>, and 90<sup>th</sup> percentile HI values were 0.02, 0.06, and 0.25 for all inhabitants; 0.01, 0.05, and 0.18 for males; and 0.02, 0.07, and 0.29 for females, respectively. Approximately 0.62% (all inhabitants; Fig. 2a), 0.22% (male inhabitants; Fig. 2b), and 1.17% (female inhabitants; Fig. 2c) had HI values greater than 1, indicating slight non-carcinogenic risk. Among age groups, the 90<sup>th</sup> percentile HI value was highest (0.40) for inhabitants under 20 years old and lowest for those over 60 years (0.11). Approximately 2.07% (under 20 years old), 0.28% (20–40), 0.53% (40–60), and 0.04% (over 60 years old) of inhabitants had HI values greater than 1 (Fig. 3), indicating very low non-carcinogenic risk in all age groups. Based on the HI values determined via probabilistic estimation, we can conclude that the non-carcinogenic health risks resulting from the studied metals are not significant.

Table 4

#### Fig. 2

#### Fig. 3

To evaluate carcinogenic risk, all *R* values were also fitted to lognormal distributions. The 10<sup>th</sup>, 50<sup>th</sup>, and 90<sup>th</sup> values of *R* for all inhabitants were  $0.29 \times 10^{-5}$ ,  $1.61 \times 10^{-5}$ , and  $8.62 \times 10^{-5}$ , respectively. The value of *R* exceeded maximum acceptable level of  $1 \times 10^{-4}$  (USEPA) in 8.23% of inhabitants (Fig. 2d), while *R* was

greater than the negligible level of  $1 \times 10^{-5}$  (WHO) for approximately 64.26% of inhabitants (Fig. 4a). The 10<sup>th</sup>, 50<sup>th</sup>, and 90<sup>th</sup> values of  $R$  were  $0.26 \times 10^{-5}$ ,  $1.28 \times 10^{-5}$ , and  $6.35 \times 10^{-5}$  for males and  $0.34 \times 10^{-5}$ ,  $1.85 \times 10^{-5}$ , and  $10.03 \times 10^{-5}$  for females, respectively. Approximately 5.12% of male inhabitants (Fig. 2e) and 10.46% of female inhabitants (Fig. 2f) had  $R$  values greater than  $1 \times 10^{-4}$ , while 58.28% of male inhabitants and 68.30% of female inhabitants had  $R$  values greater than  $1 \times 10^{-5}$  (Fig. 4b and Fig. 4c); thus, the carcinogenic risk was greater for females than for males. The respective 10<sup>th</sup>, 50<sup>th</sup>, and 90<sup>th</sup> values of  $R$  were  $0.40 \times 10^{-5}$ ,  $2.31 \times 10^{-5}$ , and  $13.40 \times 10^{-5}$  for inhabitants under 20 years old;  $0.32 \times 10^{-5}$ ,  $1.51 \times 10^{-5}$ , and  $7.11 \times 10^{-5}$  for inhabitants aged 20–40;  $0.30 \times 10^{-5}$ ,  $1.54 \times 10^{-5}$ , and  $7.70 \times 10^{-5}$  for inhabitants aged 40–60; and  $0.19 \times 10^{-5}$ ,  $0.88 \times 10^{-5}$ , and  $4.07 \times 10^{-5}$  for inhabitants over 60 years old. Approximately 14.81% (under 20 years old), 7.18% (20–40), 5.83% (40–60) and 2.08% (over 60 years old) of inhabitants had  $R$  values greater than  $1 \times 10^{-4}$  (Fig. 5), while 73.24% (under 20 years old), 63.26% (20–40), 63.47% (40–60), and 45.74% (over 60 years old) of inhabitants had  $R$  values greater than  $1 \times 10^{-5}$  (Fig. 6). Thus, the carcinogenic risk differed among age groups.

*Fig. 4*

*Fig. 5*

*Fig. 6*

#### *Sensitivity results*

The sensitivity analyses showed that the contributions of As concentration, maize DI, maize EF, and PB concentration to non-carcinogenic risk were 35.8%, 29.4%, 20.7%, and 1.8%, respectively. The contributions of As concentration, maize DI, and maize EF to carcinogenic risk were 61.0%, 18.5%, and 13.1%, respectively. These results imply that controlling the concentration of As would effectively reduce the health risk for local inhabitants.

## **Discussion**

### *Heavy metal levels in plant–soil–irrigation system*

Samples of maize cultivated near mine sites and industrial areas or irrigated by waste water were most likely to have heavy metal concentrations exceeding the limits. The average concentrations of Cr, Zn, Pb, and Cr in maize irrigated with waste water were significantly higher than the WHO standards [45]. Heavy metal concentrations of 0.306 (Cd), 0.24 (Pb), 4.17 (Cr), 22.46 (Zn), and 0.82 (Cu) mg/kg in maize grains around mining area of Anhui Province [46], other elements levels are similar to the concentrations detected in this study, while Cd is nearly 10 times higher than ours. One of the possible reason is that Cd level in their study soil is 0.22 ~ 0.29 mg/kg, nearly double higher than that in this study (0.14 mg/kg). Heavy metal concentrations in maize of 0.129, 0.021, 3.4, and 2.6 mg/kg for Pb, Cd, Zn, and Cu, respectively [47], below the standard values and our study values. Although their study area was an industrial area, the maize samples were collected from a market and may not have been grown near an industrial zone.

Former studies found that compared to other parts of maize, the grains have lower TF values [39,48], indicating low heavy metal uptake and accumulation in maize grains. The TF values of the six metals in this study followed the following sequence: Cd > Zn ≈ As > Cr > Cu > Pb. Researchers also found that Cd and Pb had the highest and lowest TF values in maize grains which were collected in Heilongjiang Province [48]. In this study, the soil samples were most polluted with Cd and Zn, while the maize samples were most contaminated with Cr and Pb. This indicates that the percentage of samples exceeding the concentration limit for a heavy metal was not related to TF.

The metal levels determined in this study were in agreement with other values for water samples reported in the literature. Compared to this study, one study showed similar metal concentrations in filtered water samples: Cd (ND–0.11 µg/L), Cr (74.80–94.70 µg/L), Cu (0.68–2.79 µg/L), Pb (ND–0.82 µg/L), and Zn (0.19–1.82 µg/L) [21]. However, the above samples along with the water samples in this study were filtered and did not include suspended particles or sediment. According to another study, the heavy metal concentrations in suspended particulate matter of the Yellow River were 0.428 mg/kg for Cd, 74.9 mg/kg for Cr, 40.1 mg/kg for Cu, 32.6 mg/kg for Pb, and 13.6 mg/kg for As [49], below the standards set for soils but higher than the background values in local soil. One of above studies also reported heavy metal concentrations in suspended particles and sediment from the Yellow River [21]: Cd (0.23–1.09 mg/kg), Cr (64.50–84.90 mg/kg), Cu (25.40–42.20 mg/kg), Pb (20.80–31.70 mg/kg), and Zn (72.50–107.00 mg/kg) for suspended particles and Cd (0.23–1.09 mg/kg), Cr (64.50–84.90 mg/kg), Cu (25.40–42.20 mg/kg), Pb (20.80–31.70 mg/kg), and Zn (72.50–107.00 mg/kg) for sediment. These results imply that heavy metals in the Yellow River primarily accumulate in suspended particles and sediment, which should receive more attention.

#### *Health risk assessment*

Previous studies also have reported that inhabitants are exposed to metals through maize consumption [43,50]. The dietary intakes of metals from maize in the Swat District of northern Pakistan were reported as 0.043, 0.013, 0.075, and 0.083 µg/kg/day for Cd, Cr, Cu, and Zn, respectively [43], and the HQ values for individual metals along with the combined HI value were far less than 1, implying that maize consumption does not result in obvious non-carcinogenic risk. The dietary intakes of metals from maize in Tonglushan mine in Hubei, China were reported as 0.027, 0.021, 1.10, and 0.032 µg/kg/day for Cd, Pb, Cu, and As, respectively [50], with the HI value less than 1 (0.42). The carcinogenic risk caused by As via maize consumption has rarely been reported. Many reports have focused on carcinogenic risk resulting from rice consumption [51–53] because among different foods, rice has the greatest contribution to total As daily intake [54].

In the present study, the estimates of health risk from heavy metal exposure are only for maize consumption and do not account for any other sources. In reality, health risk is affected by various sources of heavy metals such as different foodstuffs (e.g., rice, wheat, and vegetables) and drinking water. Other pathways of heavy metal exposure including dust inhalation and dermal contact also contribute to health risks. Therefore, exposure from other sources should be considered to estimate the total risk in future works.

### *The uncertainties of risk assessment*

The uncertainties in risk assessment can be represented in various forms, including probability density functions, fuzzy numbers, and arithmetic intervals [54]. Probability density functions were used in this study. What's more, Monte Carlo simulation is the most widely used method in risk assessment as it can combine multiple probability density functions of risk to quantify uncertainty or variability in a probabilistic framework using computer simulation [55]. Many researchers have used Monte Carlo simulation to evaluate probabilistic risks [56–59]. In recent years, Monte Carlo simulation has been combined with other approaches to evaluate risk. Kentel and Aral [60] combined Monte Carlo simulation with fuzzy calculation to evaluate certain percentiles of risk. Kentel and Aral [61] also developed a probabilistic-fuzzy method called two-dimensional fuzzy Monte Carlo simulation. In this approach, Monte Carlo simulation is used to capture variability, while fuzzy calculation is used to capture uncertainty. To assess the health risk from maize consumption more precisely, Monte Carlo simulation could be combined with fuzzy calculation in a future study.

## Conclusion

Although the average heavy metal concentrations in soil and irrigation water did not exceed the corresponding national standards, the average contents of two metals (Pb and Cr) in maize exceeded the standards for food. Thus, exposure to heavy metals via maize consumption poses potential health risks to local inhabitants. Based on deterministic and probabilistic risk estimation, there are no obvious non-carcinogenic health risks to inhabitants, while carcinogenic risk from As exposure is lower than the maximum acceptable level of  $1\times10^{-4}$  and higher than the negligible level of  $1\times10^{-5}$ . Females are at greater risk than males, and inhabitants under 20 years of age have the highest risk among age groups. To reduce risk, mitigation strategies should be adopted, such as: controlling metal-containing emissions from industry; controlling the application of metal-containing fertilizers and pesticides; and implementing efficient irrigation methods to reduce metal contamination.

## Abbreviations

As: arsenic; BW: body weight; C: concentration of heavy metals; Cd: cadmium; Cr: chromium; Cu: copper; DI: daily intake; ED: exposure duration; EF: exposure frequency; EXPO: exposure to metals; HQ: hazard quotient; HI: hazard index; MS: morbidity status; ICP-AES: inductively coupled plasma-atomic emission spectrometry; Pb: lead; I. Q.: Intelligence Quotient; R: carcinogenic risk of As; RfD: reference dose; SF: slope factor; SD: standard deviation; TF: transfer factor; USEPA: United States Environmental Protection Agency; WHO: World Health Organization; Zn: zinc

## Declarations

### *Ethics approval and consent to participate*

The study was approved by the institutional research ethics committee of Ningxia Medical University, and written informed consent was obtained from each participant.

#### *Consent for publication*

The study was approved for publication by all authors.

#### *Availability of data and material*

All data analyzed during this study are included in this published article.

#### *Competing interests*

The authors declare that they have no conflicts of interest.

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#### *Author Contributions*

PL, MZ and JT conceived and designed the experiments. YZ and NF performed the experiments. PL and YZ analyzed the data. JT and MZ provided the analysis tools. PL wrote the manuscript. All authors read and approved the final manuscript.

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## Tables

Table 1 The concentrations of heavy metals in the maize grains, the associated soil and the irrigated water.

Metals	Maize(mg/kg)			Associated soil (mg/kg)			Transfer factor (TF=C <sub>corn</sub> /C <sub>soil</sub> )		Irrigated water (µg/L)		
	Mean	Safe limits <sup>a</sup>	Over-limit ratio	Mean (mix-max)	Safe limits <sup>b</sup>	Over-limit ratio		Mean (mix-max)	Safe limits <sup>c</sup>	Over-limit ratio	
Cd	0.037 (0.00025- 0.14)	0.1	2%	0.14 (ND- 0.45)	0.3	24.4%	0.26	0.11 (ND- 0.4)	10	0%	
Pb	0.41 (ND- 2.66)	0.2	47%	18.16 (0.64- 35.08)	300	0%	0.023	1.12 (0.045- 5.9)	200	0%	
Cr	2.36 (ND- 23.08)	1.0	69%	37.25 (19.71- 65.74)	200	0%	0.063	41.03 (ND- 71)	100	0%	
Zn	17.02 (0.84- 39.4)	50	0%	138.20 (39.21- 542.27)	250	20.2%	0.12	0.91 (0.095- 1.9)	2000	0%	
Cu	1.04 (0.0080- 2.57)	10	0%	19.61 (2.37- 77.82)	100	0%	0.053	1.6 (0.74- 2.7)	500	0%	
As	0.17 (ND- 0.73)	0.5	18%	14.18 (9.08- 21.24)	25	0%	0.12	3.6 (2.4- 6.1)	50	0%	

<sup>a</sup> means allowable levels of heavy metals for corns in China by GB 2762-2017 and NY 861-2004 standards;

<sup>b</sup> means allowable levels of heavy metals for soil in China by GB15618-1995 standard;

<sup>c</sup> means allowable levels of heavy metals for irrigated water in China by GB 5084-2005 standard.

ND means not detected.

Table 2 Exposure factors of the inhabitants by gender and age groups (mean±SD)

	n(person)	Body weight(kg)	Daily intake of corn (g/day)	Exposure frequency (day/year)
All	103	54.60±18.56	161.13±97.16	15.50±8.45
Male	50	58.45±18.28	160.20±96.27	14.34±7.47
Female	53	50.67±18.19	162.08±99.06	16.67±9.28
<20	26	30.92±15.37	118.46±83.03	17.81±8.75
20~40	35	62.55±10.73	186.37±95.42	14.06±6.13
40~60	27	65.78±7.68	181.48±105.75	17.11±10.11

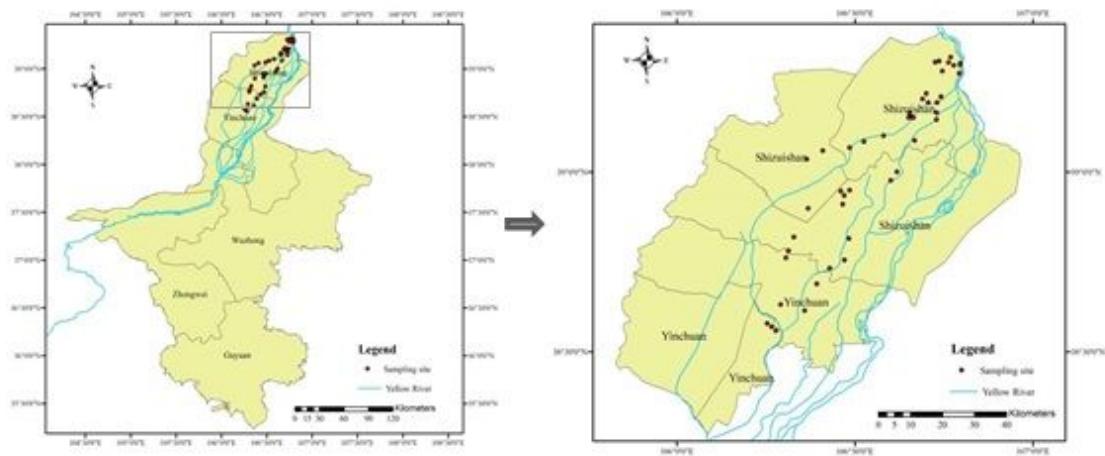
Table 3 Non- carcinogenic risk HQ, HI value of heavy metals and carcinogenic risk R value of As due to consumption of corn by deterministic estimation method

	Non- carcinogenic risk (HQ)							Carcinogenic risk (R)
	Cd	Pb	Cr	Zn	Cu	As	HI	
All	0.0048	0.0109	0.0002	0.0071	0.0033	0.0725	0.0986	3.261E-05
Male	0.0041	0.0093	0.0002	0.0061	0.0028	0.0623	0.0848	2.804E-05
Female	0.0055	0.0127	0.0002	0.0083	0.0038	0.0845	0.1150	3.802E-05
<20	0.0071	0.0162	0.0003	0.0106	0.0049	0.1081	0.1472	4.864E-05
20~40	0.0044	0.0100	0.0002	0.0065	0.0030	0.0664	0.0904	2.987E-05
40~60	0.0049	0.0112	0.0002	0.0073	0.0034	0.0748	0.1018	3.366E-05
>60	0.0025	0.0057	0.0001	0.0037	0.0017	0.0378	0.0514	1.700E-05

Table 4 The statistics of probabilistic estimation of HI and R values

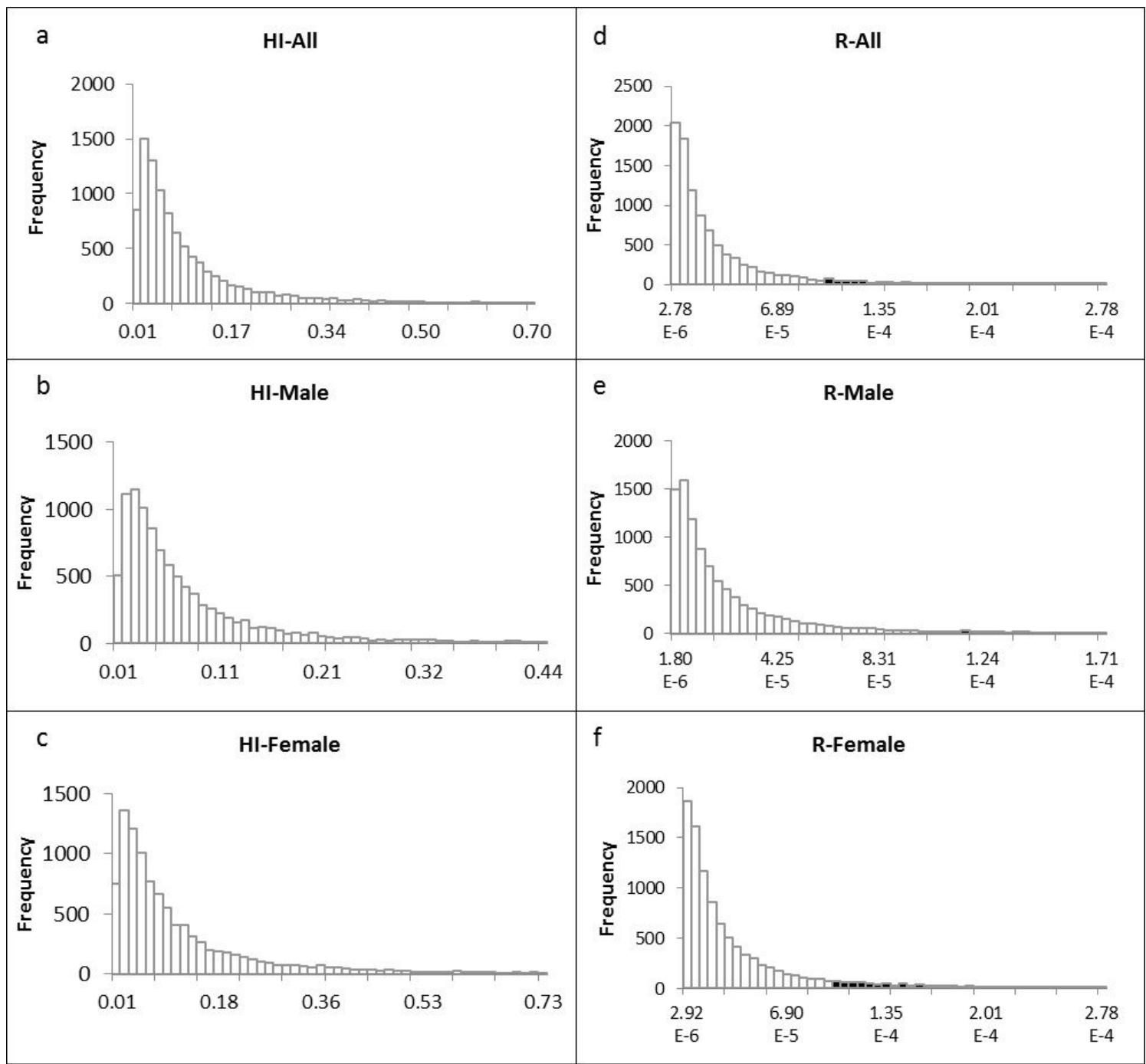
Non- carcinogenic risk $\text{OR}^{\text{HII}}$							Carcinogenic risk $\text{OR}^{\text{R}} \times 10^{-5}$						
	Distribution	Parameters	10%	50%	90%		Distribution	Parameters	10%	50%	90%		
All	Lognormal	Location:0.00, Mean:0.11, SD:0.18	0.02	0.06	0.25		Lognormal	Location:0.00, Mean: 3.8, SD: 8.3	0.29	1.61	8.62		
Male	Lognormal	Location:0.01, Mean:0.08, SD:0.11	0.01	0.05	0.18		Lognormal	Location:0.00, Mean:4.4, SD: 9.5	0.26	1.28	6.35		
Female	Lognormal	Location:0.00, Mean:0.13, SD:0.20	0.02	0.07	0.29		Lognormal	Location:0.00, Mean: 3.3, SD: 2.0	0.34	1.85	10.03		
<20	Lognormal	Location:0.00, Mean:0.18, SD:0.29	0.02	0.09	0.40		Lognormal	Location:0.00, Mean: 2.5, SD: 3.7	0.40	2.31	13.40		
20~40	Lognormal	Location:0.00, Mean:0.09, SD:0.12	0.02	0.06	0.20		Lognormal	Location:0.00, Mean: 3.1, SD: 5.7	0.32	1.51	7.11		
40~60	Lognormal	Location:0.00, Mean:0.10, SD:0.14	0.02	0.06	0.22		Lognormal	Location:0.00, Mean: 3.4, SD: 6.9	0.30	1.54	7.70		
>60	Lognormal	Location:0.00, Mean:0.05, SD:0.06	0.01	0.03	0.11		Lognormal	Location:0.00, Mean: 1.8 SD: 3.2	0.19	0.88	4.07		

## Figures



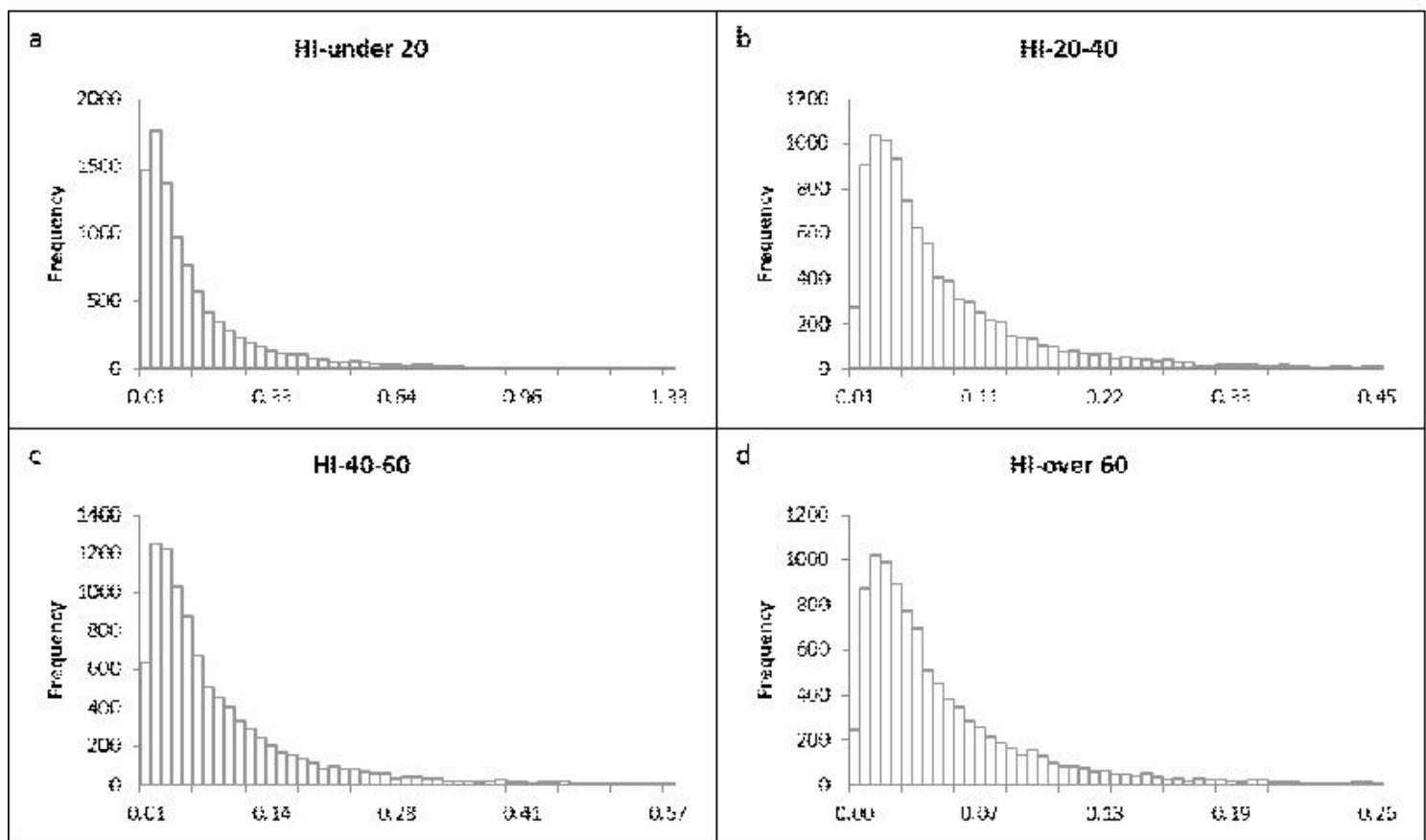
**Figure 1**

Fig. 1 Location of the study area and distribution of the sampling sites (●represents a sampling site)



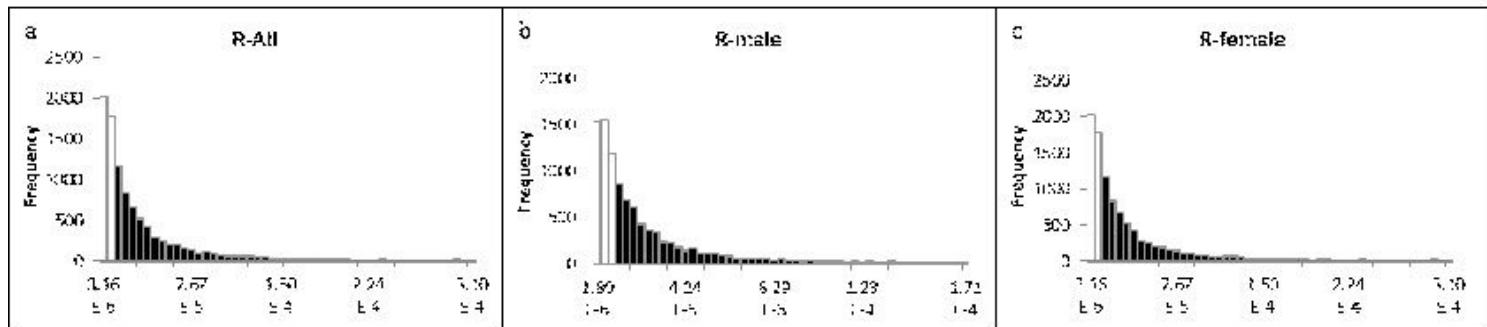
**Figure 2**

Fig. 2 Probability exceeding 1 of HI (a: all inhabitants, b: male, c: female) and 10-4 of R (d: all inhabitants, e: male, f: female). The black area represents the exceeding probabilities, which were 0.62%, 0.22%, 1.17%, 8.23%, 5.12% and 10.46% for a, b, c, d, e and f, respectively.



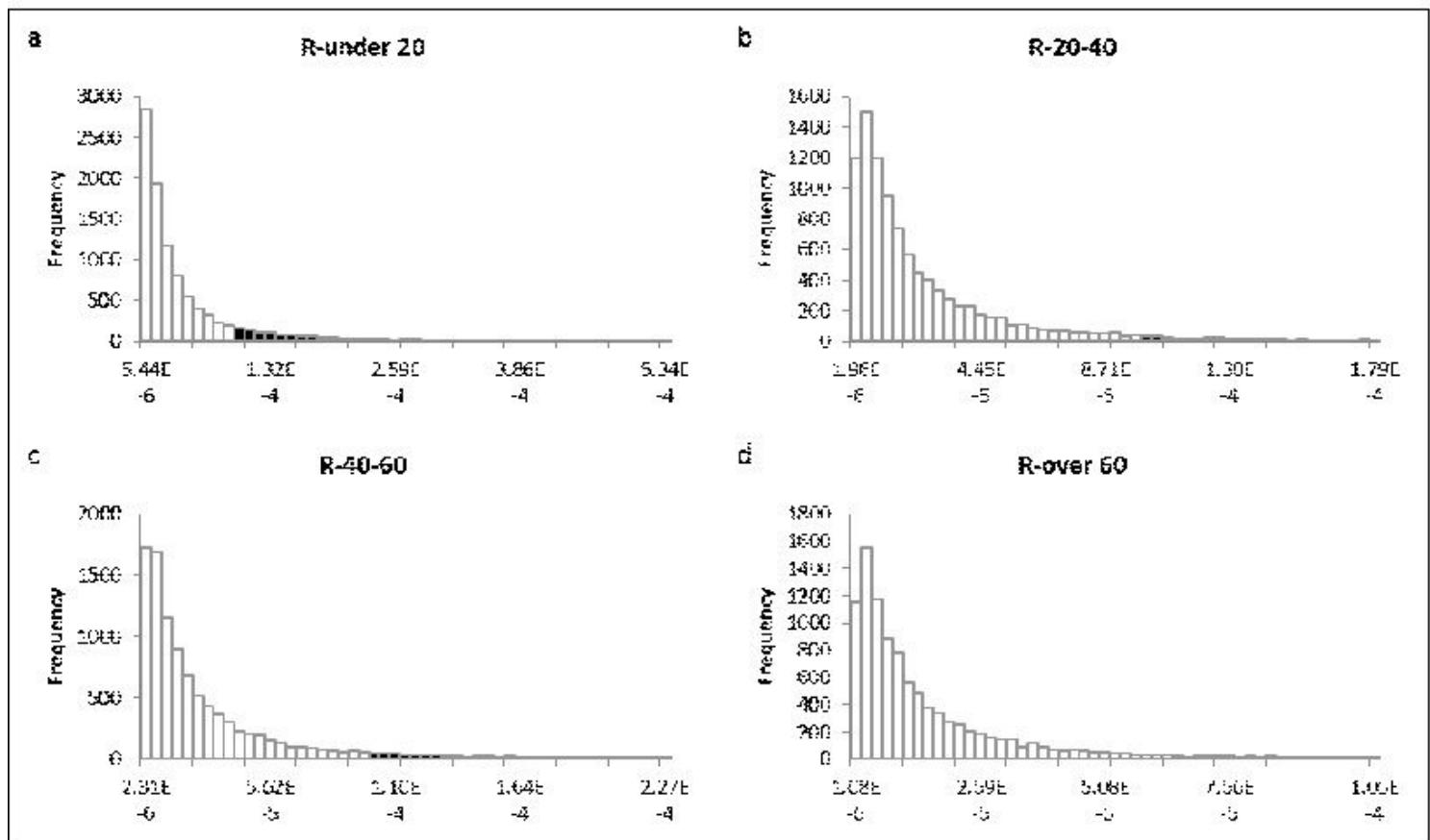
**Figure 3**

Fig.3 Probability exceeding 1 of HI (a: under 20 years old, b: 20~40, c: 40~60, d: over 60 years old). About 2.07%, 0.28% , 0.53% and 0.04% of inhabitants had HI values greater than 1 for a, b, c, and d, respectively.



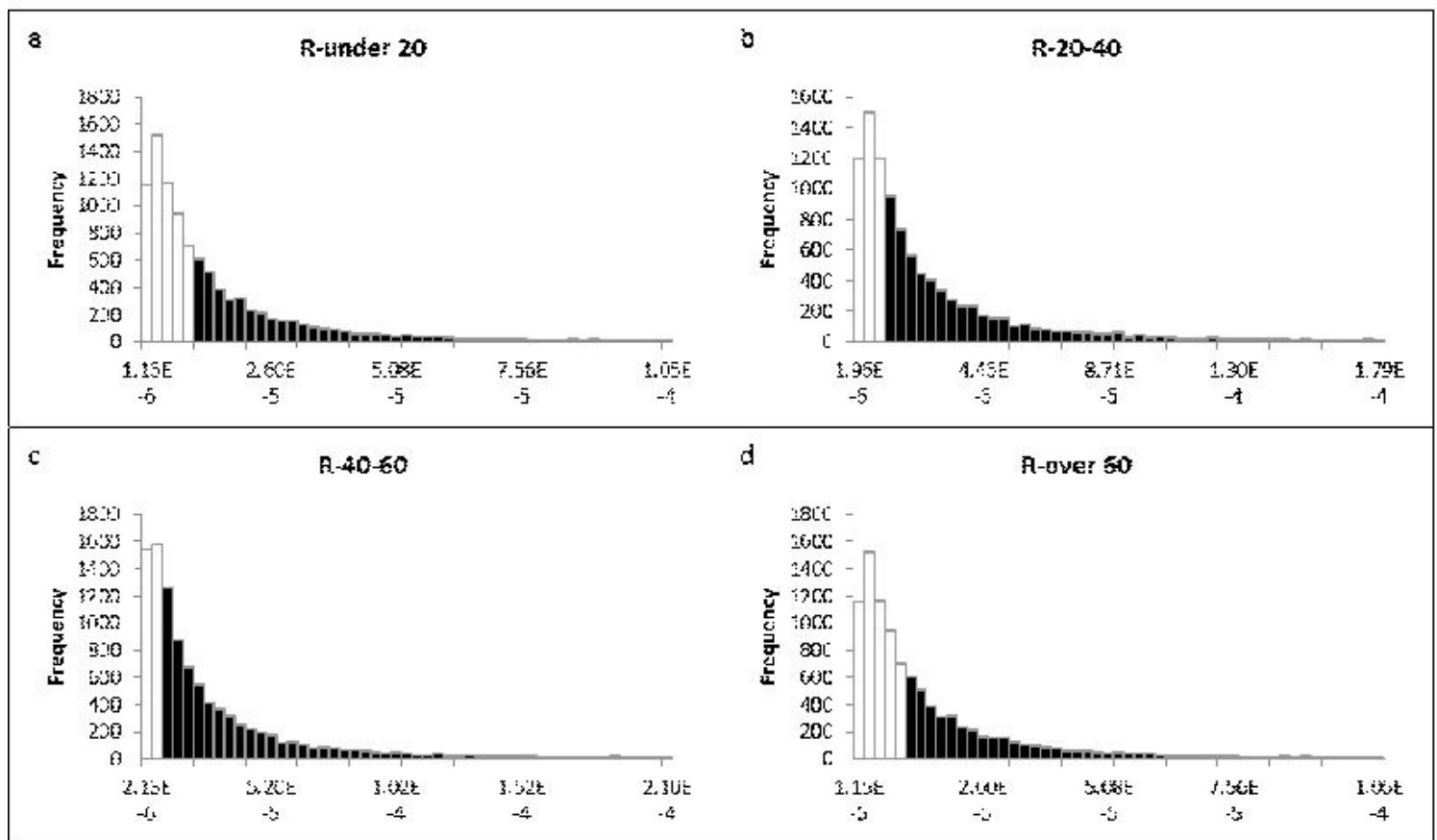
**Figure 4**

Fig.4 Probability exceeding 10-5 of R (a: all inhabitant, b: male c: female), the black area represents the exceeding probabilities, and about 64.26%, 58.28% and 68.30% for a, b, c, and d, respectively.



**Figure 5**

Fig.5 Probability exceeding 10-4 of R (a: under 20 years old, b: 20~40, c: 40~60, d: over 60 years old), the black area represents the exceeding probabilities, and about 14.81%, 7.18%, 5.83% and 2.08% for a, b, c, and d, respectively.



**Figure 6**

Fig.6 Probability exceeding 10-5 of R (a: under 20 years old, b: 20~40, c: 40~60, d: over 60 years old), the black area represents the exceeding probabilities, and about 73.24%, 63.26%, 63.47% and 45.74% for a, b, c, and d, respectively.