Difference of Trace Element Exposed Routes and their Health Risks between Agriculture and Pastoral areas in Bay Xinjiang China

Muyesaier.Tudi^{1,3, 4, 5}, Dung Tri Phung³, Huada Daniel RUAN², Lin-sheng Yang^{1, 5, *}, Hai-Jun Guo³, Des Connell⁴, Ross Sadler³, Cordia Chu³

1 Key Laboratory of Land Surface Pattern and Simulation, Institute of Geographical Sciences and Natural Resources Research, Chinese Academy of Sciences, 11 A Datun Road, Beijing 100101, China;

2 Beijing Normal University-Hong Kong Baptist University United International College, 2000 Jintong Road, Tangjiawan, Zhuhai, Guangdong Province, China

3 Centre for Environment and Population Health, Griffith University, 170 Kessel Road, Nathan QLD 4111, Australia.

4 School of Environment and Science, Griffith University, 170 Kessel Road, Nathan QLD 4111, Australia.

5 University of Chinese Academy of Sciences, Beijing 100049, China

Corresponding author: Lin-sheng Yang

Note: Agri., TECL and HHR are agricultural, trace element contamination level and human health risk respectively

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Jun Guo³, Des Connell⁴, Ross Sadler³, Cordia Chu³

1 Key Laboratory of Land Surface Pattern and Simulation, Institute of Geographical Sciences
and Natural Resources Research, Chinese Academy of Sciences, 11 A Datun Road, Beijing
100101, China;

2 Beijing Normal University–Hong Kong Baptist University United International College,
2000 Jintong Road, Tangjiawan, Zhuhai, Guangdong Province, China;

3 Centre for Environment and Population Health, Griffith University, 170 Kessel Road,
Nathan QLD 4111, Australia;

4 School of Environment and Science, Griffith University, 170 Kessel Road, Nathan QLD
4111, Australia;

5 University of Chinese Academy of Sciences, Beijing 100049, China.

Corresponding author: Lin-sheng Yang

Email: yangls@igsnr.ac.cn

Abstract: The concentration level related to toxicities of trace elements in drinking water, rice, wheat flour, and their associated negative impacts on human health have become an emergent issue in China. Because Xinjiang is the largest province in China with the majority of arable pasture land available for cultivation, it is important to analyse the concentrations of trace elements in relation to their toxicities in water, rice, and wheat flour and to investigate the health risk differences between agricultural and pastoral areas in Bay County, Xinjiang. The study results showed that (1) metal concentrations from drinking water, rice, and wheat flour were within the permissible limits; (2) concentration levels of trace elements and their total risk from drinking water and rice were higher in the agricultural areas than those in the pastoral areas, whereas concentration levels of trace elements and their total risk from wheat flour were higher in the pastoral areas than those in the agricultural areas; (3) the concentration level of the trace elements in rice were higher than in the wheat flour, but the risk from the wheat flour was higher than the risk from rice; (4) total non-cancer risk from the flour (H_{if}) in both areas exceeded the respective safe reference doses; (5) total cancer risk from the wheat flour, rice, and water exceeded the safety limit (1×10^{-4}); (6) for the exposed population, arsenic was suggested as the most evident pollutant leading to carcinogenic concerns regarding the water, rice, and wheat flour; (7) the risk index from the wheat flour made up the highest percentage both in the total cancer risk and the non-cancer risk, followed by rice and then water; and (8) the human health risk was attributed to influence from the local environment in the agriculture areas, while it was attributed to the external environment in the pastoral areas.

Key words: health risk assessment, trace elements, water, rice, wheat flour, , Xinjiang, China

1. Introduction

Although essential trace elements are essential for animal and human life, they are only needed at minimum levels (Chen et al., 2018). Concentrations of trace elements are increased by waste disposal, vehicular exhausts, atmospheric deposition, fertilization, use of pesticides, and application of sewage sludge in arable land (Cheng et al., 2018). When more trace elements are taken up by humans than are discharged, the extra trace elements will accumulate in the body (Zheng et al., 2013). Generally, it takes several decades for trace elements to be excreted from human body (Liu, 2014). Trace elements have a significant toxicity, persistence, and bio-accumulation (Cheng et al., 2018).

Trace elements may enter the human body through oral intake, inhalation, or dermal contact, and this may cause negative impacts on human health (Zheng et al., 2013). Food and water constitute important sources of human oral exposure to trace elements (Doabi et al., 2018, Chen et al., 2018). Exposure to trace elements from drinking water has affected human health with different diseases (Singh & Kumar, 2017). Aside from drinking water, crops—especially rice and wheat—may be the most important pathway for human uptake of trace elements. The main cause of excessive trace elements in drinking water and crops in rural areas is land reclamation, including the use of chemicals and wastewater irrigation in agricultural activities (Yang et al., 2018). Pesticides and some chemicals from wastewater irrigation circulate through the atmosphere, water, soil, and biosphere, but some of these are vestigial in surface water, ground water, and crops (Sánchez-González et al., 2013). The chemical residues may not be completely removed via modern drinking water treatment. As a result of this, drinking water and crops in rural areas may not always be safe for human consumption, but safety of the drinking water and crops always neglected (Guo et al., 2014).

Owing to rapid economic development in the last two decades, the China's environment has been badly polluted by trace elements (Chen et al., 2018; Cheng et al., 2018; , Li et al., 2015). The concentration levels related to toxicities of trace elements in drinking water, rice, and wheat and their associated negative impacts on human health have become an emergent issue in China. Thus, it is important to monitor and improve the quality of drinking water and crops to ensure the health and safety of people in China. Although many studies have focused on the trace element concentration levels in drinking water and crops and their human health risk assessment, most of the studies investigated the industrial areas (Wu et al., 2018, Tepanosyan et al., 2018, Oguri et al., 2018). Trace element pollution has already had adverse impacts on the environment and threatened human life in the agricultural areas (Yang et al., 2018).

Xinjiang has the largest areas of reclaimed land in China, and it is being reclaimed at the fastest rate (Liu 2014). Consequently, this land reclamation increases trace element concentrations and causes many health problems. Furthermore, land cultivation in Xinjiang has transformed pastoral areas into agricultural lands through the applications of different technologies and chemicals, which could result in two negative impacts. First, it may cause the pollution of water and soil resources in reclaimed land. Second, the consumption of crops in cultivated areas is increasing. Both these aspects increase the exposure to trace elements in terms of their health risk. Moreover, Xinjiang is a temperate zone with a typical continental climate (Hu, 2014); therefore, trace elements tend to easily accumulate in the surface of the soil, which leads to the problem of contamination of trace elements in water and crops in Xinjiang (Liu, 2014). However, information on the concentration of multiple trace elements from multiple pathways such as drinking water, rice, and wheat flour, and their potential health risks on Xinjiang, is insufficient so far. As a cultivated land development zone that has

been reserved, Xinjiang provides the largest strategic support for food production and arable land security in China.

As such, it is important to analyze the concentration levels of the environmental multiple trace elements in drinking water and crops as well as their health risk differences between agricultural and pastoral areas in Xinjiang to investigate how land reclamation from pastoral areas to agricultural areas increases the environmental trace element concentration level, and its human health risk. In addition, it is necessary to analyses the health risk of the multiple trace element from three main pathways in the rural areas of Xinjiang in order to provide a strategy for food safety and agriculture development in the Xinjiang regions and in other regions of China.

In this study, drinking water, rice, and wheat flour are the main routes for trace element exposure, so samples of drinking water, rice, and wheat flour were collected from 180 households in typical agricultural and animal husbandry regions. The distribution and human health risk differences among the elements Fe, Zn, As, Mn, Ni, Se, Cu, Co, Pb, and Cd from multiple exposure routes including drinking water, rice, and wheat flour in these regions were investigated. This study may provide basic information for determining the impacts of land cultivation on the concentration variations of trace elements and their exposure levels in drinking water, rice, and wheat flour that pose a threat to human health. It may also provide experimental data and local evidence for food and drinking water risk management in Xinjiang.

2. Materials and methods

2.1 Study area

This study was conducted in Bay County, Xinjiang, China, which is located in the southwest of the Xinjiang Uyghur Autonomous Region. Specifically, the region is between $80^{\circ}37'39''$ – $83^{\circ}02'25''$ E and $41^{\circ}24'08''$ – $42^{\circ}38'52''$ N. The Keyir (KYR), Kariwah (KRW), Dawanqi (DW), and Yaturi (YTR) are typical agricultural areas, and Yangchang (Y) and Bulong (B) are pastoral areas in this region (Figure. 1). In DW, KYR, KRW, and YTR, farmers cultivate wheat for production of wheat flour but rice must be imported from other regions and sold in supermarkets. In the pastoral areas of Y and B, both rice and wheat flour is imported.

2.2. Sample collection and preparation

In July 2015, methods of the multi-stage random sampling (Guo et al., 2015) were used to collect samples of tap drinking water, flour, and rice from 180 families. The drinking water was taken from the water containers (the population collects drinking water from the taps and stores the water in a container), and the rice and wheat flour samples were collected from the food containers of every family in the four typical agricultural (Keyir, Kariwah, Dawanqi, Yaturi) and two natural pastoral (Bulong, Yangchang) areas of Bay County (Figure 1). A total of 30 samples for each of the three media from every region were collected.

About one liter of water was collected in a plastic container and then samples were filtered through the filter papers. A total of 10 ml of nitric acid was added to preserve the drinking water samples. The acidified drinking water samples were stored at 0 – 4 °C and analyzed within seven days of collection. The water samples were collected and preserved in accordance with the GB/T5750.2-2006 Standard test for Drinking Water (Magesh et al., 2017).

The wheat flour and rice samples were collected separately in plastic bags. Five hundred grams of polished rice and wheat flour were packed in polyethylene bags and sent to a laboratory. The rice samples were rinsed with tap water followed by deionized water to remove dust. After drying in an oven at 50 °C for 24 hours, each rice or wheat flour sample was homogenize by grinding with an agate mortar and passing through a 0.15 mm sieve (Ma et al., 2016). All samples of rice and wheat flour powders were stored in sealed polyethylene bags and preserved in a desiccator before analysis.

2.3 Instrumental analysis and quality control

2.3.1 Instrumental Analysis

Drinking water samples

Each 10 mL water sample was acidified with 100 µL of nitric acid, and the concentrations of trace elements (Fe , Zn , As , Mn , Ni , Se , Cu , Co , Pb , Cd) were determined using Agilent 7700x inductively coupled plasma mass spectrometry (ICP-MS).

Rice and flour samples

Exactly 0.5 g of each prepared dried rice or wheat flour powder sample was placed into a Parr Acid Digestion Vessel. Then, 5 mL of concentrated nitric acid (HNO₃, 70% wt ≥99.999% metal basis, Aladdin Reagent Co. Ltd, Shanghai, China) and 1 mL hydrogen peroxide (H₂O₂, 30% wt, guaranteed reagent grade, Aladdin Reagent Co. Ltd) was added. The samples were then set aside for 24 hours in order to allow the contents to thoroughly mix. After the initial reaction, the electric heating plate method (Chen et al., 2018) was used to individually digest the rice and wheat flour powder samples. After complete digestion, the sample mixture was filtered through a Whitman 0.45 µm filter paper. After cooling down to room temperature, the digested solutions were diluted with ultrapure water up to 25 mL and kept at 4 °C (Guo et al.,

2015). All digestions were carried out in duplicate, and a method blank was carried throughout the entire procedure for each batch of samples. Concentrations of the trace elements (Cd , Co , Pb , Ni , As , Se) were determined using Agilent 7700x inductively coupled plasma mass spectrometry (ICP-MS) and concentrations of the trace elements (Zn , Cu , Fe , Ca , Mg , Mn , Mo) were determined using the inductively coupled plasma optical emission spectroscopy (ICP-OES).

2.3.2 Quality control

All the laboratory apparatuses were soaked in 10% nitric acid solution for acid washing overnight before being rinsed with deionized water. ICP-OES and ICP-MS were calibrated before and after the analysis of elemental concentrations. An external calibration curve with corresponding standards was obtained before the analysis by using a stock standard solution (Chen et al., 2018). A blank sample containing 5% (v/v) nitric acid was run after every 10 samples were analyzed. Elements were analyzed in triplicate to assess the precision of the analytical process throughout the analysis. Standard deviation (SD) was used to confirm the precision of the analytical results (Guo et al., 2015). The accuracy of analyses was checked on samples of rice and wheat flour respectively by comparing their trace elements to the concentrations of the certified standard materials (GBW10010 and GBW10011, China National Center for Standard Materials).

2.4. Data analysis

This study used descriptive statistics to analyze the main features of the elements in drinking water, wheat flour, and rice. One-sample Kolmogorov-Smirnov (K-S) test was used to analyze the distribution of the concentration of the elements. Tukey HSD test and Non-parametric test were used to examine the differences in concentration of elements between

agricultural and pastoral areas. The study used the deterministic approach method to evaluate the hazard quotient and cancer risk index of elements. The study also used the 95th of the linear prediction method to calculate the 95th percentile of exposure level. All statistical analyses were performed using the IBM Statistical Package for the Social Sciences (SPSS) version 19.0, STATISTICA software.

2.5 Human health risk assessment

Health risk assessment is the process of estimating the probability of occurrence of events and the probable magnitude of adverse health effects over a specified time. It includes four main steps. These are hazard identification, dose response assessment, exposure assessment, and risk characterisation (U.S. EPA, 2004). This study used a deterministic risk to assess the risk index of drinking water, rice, and flour. Health risks from individual trace element exposure are insufficient to describe full health risks to residents (Praveena & Omar, 2017). Cumulative trace element exposures will increase the health risks more than individual trace element exposure (Zheng et al., 2013). Thus, in this study hazard identification and dose response assessment steps were identified by literature reviews and then exposure assessment and hazard identification were calculated with first hand data by the risk assessment model. The total cancer risk (TCR) and total non-cancer risk index (HI) of ten elements in drinking water, rice, and flour were calculated on this study. For the total cancer risk and non-cancer risk, we assumed that 10 trace elements have similar modes or mechanisms of action and there is no coordination or antagonist's effect.

2.5.1 Exposure assessment

Human exposure to trace elements mainly occurs in three pathways: direct ingestion, inhalation through the mouth and nose, and dermal absorption through exposed skin. For

trace elements in water and crops, ingestion is considered the main pathway. The average and 95% concentration doses are calculated with formula (1), which is adapted from the US Environmental Protection Agency. Calculations are performed for adults (as the general population).

$$ADD = \frac{C \cdot IR \cdot ED \cdot EF}{BW \cdot AT} \quad (1)$$

In the above formula, ADD is the average (50%) and 95% cumulative concentration value of the daily dose through ingestion (expressed in mg/ kg/day), C is the average (50%) and 95% cumulative concentration of the chemical pollutants in drinking water (mg/L), rice, and wheat flour (mg/kg); IR includes IR_w , IR_r and IR_f ; IR_w is the average daily water consumption, i.e., 2.810 L/day for adults; IR_r is rice average consumption, i.e., 80g/day for an adult; IR_f is wheat flour average consumption i.e., 456.30 g/day for an adult; ED is exposure duration (72.4 a); EF is exposure frequency (365 days); BW is an adult's body weight (62.5 kg); and AT is life time (365 days $\times$ 72.4 years). The above exposure variables were based on the results of the questionnaires, relevant research (Zheng et al., 2013), the standards of the United States Environmental Protection Agency (EPA, 2004), and the report by the Environmental Ministry of China (2010).

2.5.2 Risk characterization

The deterministic approach is used for all the parameters. Cd, As, Ni, and Co are classified as carcinogenic elements (Table S1), while Co , Cu , Fe , Pb , Mn , Fe , Ni , Se , and Zn are non-carcinogenic (HQ) elements for humans, based on the ingestion reference dose reported by the U.S. EPA (EPA, 2004) (Table S1).

2.5.2.1 Hazard quotient

The potential non-carcinogenic human health risks that are caused by trace element exposure are usually characterized by the hazard quotient (HQ), which is the ratio of ADD of each contaminant for an individual ingestion exposure pathway to the corresponding reference dose (RFD) expressed in mg/ (kg day); and RFD is the oral reference dose, which is obtained from a database of risk-based concentrations (Table S1). The HQ is calculated using Equation (2). In this equation, HQ_w, HQ_r, and HQ_f in the text are the hazard quotients through ingestion of water, rice, and wheat flour (unitless).

$$HQ = \frac{ADD}{RFD} \quad (2)$$

The hazard index (HI) is introduced to evaluate the aggregate non-carcinogenic risks posed by the mixed trace elements using Equation (3).

$$HI = \sum_i^n HQ \quad (3)$$

In the text, HI_w, HI_f, and HI_r are the sum of the HQs from all the elements that induce hazard quotient in water, rice, and wheat flour respectively;; and n is an element that induces the HQ (hazard quotient). HI_w, HQ_r, and HQ_f in the text are the total hazard index of elements from water, rice, and wheat flour respectively (unitless). If HI or HQ is greater than 1, this indicates a potential for adverse effects on human health (Praveena & Omar, 2017).

2.5.2.2 Carcinogenic risk (CR)

The carcinogenic risk is the incremental probability of an individual developing cancer over a lifetime due to carcinogenic exposure in a specific scenario. The carcinogenic risk is

evaluated by the following Equation (4). The exposure metric used for carcinogenic risk assessment is the lifetime average daily dose (the average lifetime is 72.4 years in this study).

$$CR = ADD * CSF \quad (4)$$

To evaluate the total potential cancer risks (TCR) from all of these elements in each pathway for drinking water, rice, and flour, these parameters were calculated using Equation (5) (Praveena and Omar, 2017).

$$TCR = \sum^n CR \quad (5)$$

In this, CR is the individual carcinogenic risk of every element; n is an element that induces cancer risk; and CSF is the cancer slope factor (expressed in mg/kg/day). TCR_w, TCR_r, and TCR_f in the text are the total cancer risk index (TCR) of water, rice, and flour.

3. Results and discussion

3.1 Environmental trace element concentration

The descriptive statistical results for trace elements in drinking water and crops (wheat flour and rice), including minimum, maximum, mean, standard deviation (SD), coefficient of variation (CV), p value of the K-S test, and 95% cumulative concentration, are summarized in S2 and S3 respectively.

3.1.1 Trace element concentration in drinking water

The results obtained from these trace element analyses of drinking water in both agricultural and pastoral areas are shown in Figure 2a, and are also compared to the guideline values (China's drinking water standard (GB5749-2006) and those of WHO and USEPA). The results indicated that Cd, Cu, Mn, Ni, Pb, Zn, Fe, As, and Se in drinking water in the agricultural and pastoral areas are within the permissible limits of standards. This result

implies no obvious pollution resulted from these elements in drinking water, and it showed good to excellent water quality with respect to trace element concentrations in the agricultural and pastoral areas of Bay County, Xinjiang, China. The pH is a critical index for water quality, and it is useful for reflecting water chemistry (Hu et al., 2015). The pH levels of both areas were neutral to slightly alkaline, and within the limits of the World Health Organization recommended values.

The trend of mean metal concentrations (mg L^{-1}) (Table S2) in drinking water in the agricultural and pastoral areas respectively followed a decreasing order:

Agricultural areas: $\text{Fe} > \text{Zn} > \text{As} > \text{Mn} > \text{Ni} > \text{Se} > \text{Cu} > \text{Co} > \text{Pb} > \text{Cd}$

Pastoral areas: $\text{Fe} > \text{Zn} > \text{Se} > \text{Ni} > \text{As} > \text{Mn} > \text{Cu} > \text{Co} > \text{Cd} > \text{Pb}$.

In both areas, large values of CV ($> 50\%$) were found for Cd, Cu, Mn, Pb, Zn, As, and Se in drinking water in the agricultural areas and for Cu, Mn, Pb, and Zn in drinking water in the pastoral areas. This represents wide variations in element concentrations. The variety of the water source is a main reason for the wide variations in element concentrations. Water samples were collected from the residents' containers and households used different containers, so this container variety is another possible reason for the wide variations in element concentrations. The highest concentrations were observed for Fe and Zn, and these two elements accounted for almost 90% of the total concentrations, while the lowest were for Pb and Cd. Trace elements reach water systems in natural and anthropogenic ways. Regardless of origin, many physicochemical and biochemical processes affect their distribution in the water system (Kostić et al., 2016). The distribution trends in metal concentrations found in this study are similar to trends observed in previous studies (Magesh et al., 2017, Zhang et al., 2017, Saha et al., 2017a). The source of Fe and Zn is mainly

associated with natural weathering of source rocks or aquifer materials (Magesh et al., 2017, Saha et al., 2017a). Thus, the relatively high levels of Zn and Fe found in this area may be from natural occurrences, including composition of the earth's crust and weathering, erosion of bedrocks, volcanic activities, and ore deposits. The average contents of Fe and Zn in this study were lower than those in the previous studies, which were collected in the Tamiraparani river basin, South India (Magesh et al., 2017, He, et al ., 2016).

T test was used to analyze the difference in contamination resulting from Cd , Cu , Mn, Pb , Zn , Co , and As (these elemental concentrations were disturbed normally in both areas) in drinking water between the agricultural and pastoral areas. A non-parametric test (Kolmogorov-Smirnov two sample test) was used to analyze the difference in contamination by Ni , and Fe , (these elemental concentrations were non-normally distributed) in the drinking water between the agricultural and pastoral areas (Figure 3). Figure 3 shows that there were significant differences in the drinking water between the agricultural and the pastoral areas in the levels of all elements except Zn. The levels of As , Pb , Mn , Cd , Cu , and Co in the drinking water were higher in the agricultural areas than in the pastoral areas, and the mean values of Fe , Se , and Ni were higher in the pastoral areas than those in the agricultural areas. The levels of most of the elements, especially those with relatively high negative impacts in the drinking water were higher, and their concentration variations were larger in the agricultural areas than in the pastoral areas. This could be attributed to human activities during the land cultivation, including agricultural fertilizer and pesticide leaching, and using wastewater for irrigation (Yang et al., 2018) , which increased the concentrations of trace elements in the drinking water in agricultural areas. The levels of elements Ni, Se, and Fe in the drinking water were higher in the pastoral areas than in the agriculture areas.

The trace elements Ni, Se, and Fe may be related with the geological background of the pastoral areas.

3.1.2 Trace element concentration in crop (wheat flour and rice)

The results obtained from these element analyses of crops in both agricultural and pastoral areas are shown in Figure 2 (b and c) and are also compared to the guideline/s. The Chinese food hygiene standard (GB2762-2012) was initially selected as the evaluation criterion in this study. However, Cu and Zn are not specified in this standard. Thus, the Agricultural Standard in China (NY861-2004) and the standard of the Agriculture Organization and World Health Organization (FAO/WHO 1984) were chosen as the evaluation criteria for Cu and Zn. The results indicated that the mean contents of Pb, As, Ni, Cd, Cu, and Zn in wheat flour and rice from the agricultural and pastoral areas were significantly lower than the tolerance limits of the national standard. This implies that obvious pollution was not observed in all sites in agricultural and pastoral areas of Bay Xinjiang, China.

The mean trace element concentrations (mg/kg) in wheat flour powder in agricultural and pastoral areas followed a decreasing order: Fe > Zn > Mn > Cu > As > Ni > Pb = Se > Cd > Co (agricultural areas); and Fe > Zn > Mn > Cu > As > Ni > Se > Pb > Cd > Co (pastoral areas). Large values of CV (>50%) were found for As, Pb, and Co in flour samples in the agricultural areas, representing wide variations in trace element concentration (Table S3). In both cases, the highest concentrations in wheat flour were observed for Fe and Zn while the lowest were for Cd and Co. The distribution trends in metal concentrations found in this study were similar to those in the previous study (Lei et al., 2015). The mean contents of these elements in wheat flour were compared to those in the previous studies and the results indicated that the average concentration levels of the trace elements in wheat flour in both

cases were lower than those for these elements from the wheat flour collected in Jinhui Shaanxi (Lei et al., 2015) and the Kunshan (Huang et al., 2008).

The mean trace element concentrations (mg/kg) in rice in the agriculture and pastoral areas, separately, followed a decreasing order:

$Fe > Zn > Mn > Cu > As > Ni > Se > Pb > Co > Cd$ (agricultural areas);

$Fe > Zn > Mn > Cu > As > Ni > Pb = Se > Co > Cd$ (pastoral areas). Large values of CV (>50%) were found for As and Fe in rice samples in the agricultural areas, representing a wide variation in trace element concentrations (Table S3). In both cases, the highest concentrations in rice were observed for Fe and Zn, while the lowest were for Cd and Co.

The distribution trends in metal concentrations found in this study were similar to those of the previous study (Liang et al., 2017). The average concentration levels of these elements in rice were compared to those of the previous studies and the results indicated that the concentration levels of trace elements in rice in both cases were lower than those of these elements from the rice collected in Taiwan (Tsai et al., 2010), Iran (Fakhri et al., 2018), and Guangdong (Ma et al., 2017).

In both cases, the levels of Fe and Zn in rice and wheat flour were in the highest concentrations. Iron is identified as an essential natural element in plants, and it plays an important role in plant growth, productivity, and leaf and grain yield, and it can be found naturally in different kind of plants (Praveena & Omar, 2017), but if it exceeds the limited amount it leads to some adverse health effects. Although Zn is an essential element for human health, the level of Zn that produces adverse health effects is much higher than the dietary reference intake (DRI) for the essential nutrient (Tsai et al., 2010). Elements including As, Cd, Pb, and Co are the most toxic elements because As, Pb, and Cd are nonessential

elements with toxicity effects on plants and humans (Saha et al., 2017b). Although the levels of these elements are below the danger threshold in this study, residents still need to be aware of long or excessive exposure to these non-essential and essential elements, which pose dangers to their health. The trace elements in the soil and irrigation water, and pollution from surrounding industry and agriculture practices significantly influence the level of trace elements in rice and wheat (Abbas et al., 2017). The toxic elements can be accumulated but are non-biodegradable; therefore, the presence of toxic elements in a marketed crop is largely unavoidable (Mastilović et al., 2010). Mastilović et al. (2010) also reported that trace element contamination in marketed rice and flour can also come from processing and packaging operations, such as the equipment and sanitary control preparations, as well as from packaging materials. In this study we collected rice and wheat flour samples from residents' own containers. Different households used different types of containers, so the containers may also be one of the sources of the difference in trace element contamination in a crop. The different containers may be the other reason for the wide variation of the coefficients of trace element concentration in agriculture and pastoral areas. The variety of crop cultivars, soil properties, and cultivation practices on element uptake of crops (Ma et al., 2017) are the other reason for the distribution pattern of trace elements and the wide variation of the coefficients of trace element concentration in rice and wheat flour.

Non-parametric test and Tukey HSD test were used to analyze the difference in concentration levels of trace elements in the wheat flour from agricultural areas, rice from agricultural areas, wheat flour from pastoral areas, and rice from pastoral areas (Figure. 4). The results indicated that there were significant differences in the levels of all elements except for Fe, Pb, and As in the rice samples between the agricultural areas and the pastoral areas, while the levels of these elements in the rice samples in the agricultural areas were higher than those in

the pastoral areas. Although the rice was imported to both the agricultural and pastoral areas, the levels of the elements in the agricultural area were higher than those in the pastoral area. This difference is because the rice was imported from various sources nationwide, which reflected the different degrees of local environmental contamination by these elements.

The results showed that there were no significant differences in the levels of Fe , Zn , Cu and Co in the wheat flour samples between the agriculture areas and the pastoral areas, whereas there were significant differences in the levels of Mg , Mn , Pb , As , Se , Cd , and Ni. The levels of Mg , Mn , Pb , As , Se , Cd, and Ni in the wheat flour samples were higher in the pastoral areas than in the agricultural areas. The wheat flour in the agricultural areas was produced in the local environment but it was imported from elsewhere in the pastoral areas, so the levels of these elements in the flour in pastoral areas were different from those in the agricultural areas. The result also indicated that the wheat flour in these agricultural areas is /less contaminated than those imported from outside.

The results also indicated that the levels of most of the elements except Mg, Cu , and Cd in the rice were higher than those of the wheat flour. The distribution patterns of contamination of the trace elements in crops are different, and such differences almost depend to some degree of both the kinds of crops and the regions where they are grown (Xiao et al., 2018). Therefore, the trace element levels in different crops in the agricultural and pastoral areas were different. The other reasons for the difference are absorption characteristics of elements in rice and wheat flour, which greatly vary due to different elements and different varieties of crops (Silins and Högberg, 2011), and this is the other main reason for the differences in levels of elements in the crops in the agricultural and pastoral areas. Toxic elements in rice studies involving paddy fields and marketed rice samples have attracted worldwide concern

(Praveena and Omar, 2017, Ma et al., 2017). Rice is able to absorb toxic elements from soil and water in paddy fields more effectively than other crops (Praveena and Omar, 2017), so the levels of most trace elements in rice in both areas are higher than those in flour.

3.2 Health risk assessment

3.2.1 Exposure assessment

The deterministic estimated ADD values (average and 95%) for adults through ingestion of drinking water, wheat flour, and rice are summarized in Tables S4, S5, and S6 respectively. In this study, 95% cumulative concentration was considered as the high-end estimate/s for risk calculations (Saha et al., 2017a). The differences in the 95% of the ADD were more significant than the average values.

3.2.2 Risk characterization

According to the evaluation of health risk assessment models recommended United States Environmental Protection Agency by the U.S. EPA (2010), the level of health hazard risks (cancer and hazard quotient) caused by trace elements ingested orally through drinking water, wheat flour, and rice **were** assessed for the population in agricultural and pastoral areas of Bay County.

3.2.2.1 Cancer risk assessment

To date from previous studies, oral slope factors have been derived only for Cd , Ni , Co , and As (Fakhri et al., 2018). Consequently, the risk of cancer for adults due to ingestion exposure to water, wheat flour, and rice were only estimated for Cd , Ni , Co , and As by the deterministic approach and graphically shown in Figures 5a, Figure 5b and Figure 5c and also

in Tables S7, S8, and S9.

Drinking water

The result of the cancer risk assessment of the drinking water is shown in Figure 5a and Table S7. In both areas, the average (50%) of the risk from Cd, Ni, Co and As from drinking water were higher than the acceptable risk level (1×10^{-6}) but lower than the safety risk level (1×10^{-4}). The 95% of the cancer risk from Cd, Ni, Co and As from drinking water in pastoral areas and the 95% of the cancer risk from Cd, Ni, Co from drinking water in agricultural areas were higher than the acceptable risk level (1×10^{-6}) but lower than the safety risk level (1×10^{-4}). The 95% cancer risk of the As in agriculture areas exceed the safety level (1×10^{-4}). In both areas, the average and 95% total cancer risk (TCR) from drinking water exceeded the safety reference level (1×10^{-4}). The 95% of and the total cancer risk (TCR) in the drinking water were almost 4 times higher in the agricultural areas and almost 2 times higher in the pastoral areas than the safety reference level. In both areas, the risk index of drinking water followed this order: As > Ni > Co > Cd; thus, for the exposed population As contributed the most to the total cancer risk. The population exposed to As via drinking water is causally related to cancer of the lung, kidney, bladder, and skin, and the risk increases with increasing exposure (Zheng et al., 2013). It was indicated that ingesting drinking water from the agricultural and pastoral areas in Bay County over a lifetime could increase the probability of cancers closely related to high As intake. These results are similar to most of the previous studies (Zhang et al., 2017, Zeng et al., 2015, Liang et al., 2017). Moreover, previous studies indicated that the traditional drinking water purification processes including coagulation–sedimentation, sand filtration, and disinfection are applied to predicate water in China, but the As removal efficiency is almost zero. Thus, the cancer risk that was induced by trace elements and especially by As in water in both areas should arouse attention.

The average cancer risk indexes of Cd , As , and Co and the average value of total cancer risk from drinking water in the agricultural areas were higher than those in the pastoral areas. The risk differences indicated that cultivation and land reclamation (Guo et al., 2014) from pastoral to agricultural areas could increase trace element concentrations and their risks in drinking water, and cause many more health problems.

Crop (wheat flour and rice)

The result of the cancer risk assessment of flour is shown in Table S8 and Figure 5b. In both areas, the average and 95% of the risk induced by As , Cd , Ni , Co , and total cancer risk from flour exceeded the safety reference limit ((1×10^{-4})). The average risk of As and total cancer risk in the agricultural areas were 15 and 30 times higher than the safety reference levels. The 95% risk of As and total cancer risk in the agricultural areas were 50 and 80 times higher than the safety reference limit. The average risks of the As and total cancer risk in pastoral areas were 28 and 47 times higher than the safety levels. The 95% risk of As and TCR in pastoral areas were 50 and 80 times higher than the safety levels. In both areas, the average risk index of the flour follows this order: As > Co > Ni = Cd ; thus, for the exposed population, As contributed the most to the total cancer risk of the flour.

The result of the cancer risk assessment of the rice is shown in Table S9 and Figure. 5c. In both areas, the average and 95% risk indexes of As, Co, Ni and total cancer risk from the rice exceeded the safety reference level (1×10^{-4}). The average and 95% risk indexes of Cd in the rice were higher than the acceptable risk level (1×10^{-6}) but lower than the safety reference level (1×10^{-4}). The average risk of As and total cancer risk from rice in the agricultural areas were 7 times and 11 times higher than the safety level. The 95 % risk of the As and total cancer risk from rice in agriculture areas were 20 and 28 times higher than the safety

level. The average risk of As and total cancer risk from rice in the pastoral areas were 5 times and 7 times higher than the safety level. The 95% risk indexes of As and total cancer risk from the rice in the pastoral areas were 10 times and 15 times higher than the safety level. In both areas, the cancer risk index from the rice followed this order: $As > Ni > Co > Cd$; thus, As contributed the most to total cancer risk from the rice and the result was similar to most of the previous studies (Ma et al., 2017, Liang et al., 2017). Arsenic contamination in crops mainly come from two sources: natural process and anthropogenic activities, such as ore mining, smelting, coal combustion, and arsenic-based pesticides and herbicides in agriculture (Ma et al., 2016). Arsenic is intaken by humans via diet could be absorbed by the gastrointestinal tract and distributed into the body to impair normal functions. Thus, the cancer risk induced by trace elements, especially As in from flour and rice in both areas, should arouse great attention.

The average and 95 % cancer risk index from Cd and Ni and the average total cancer risk and As from flour in the pastoral areas were higher than those in the agricultural areas (Figure. 5b). The average and 95% cancer risk index from Cd, As, Ni, Co and total cancer risk of these elements from rice in agricultural areas were higher than those in pastoral areas (Figure. 5c). The different source of the wheat flour and rice are the main reason for the risk difference. The differences in the 95% risk index between the agricultural areas and the pastoral areas from these elements from flour and rice were more significant than the average risk index from these elements, and this result also reported in the previous study (Saha et al., 2017a).

3.3.2.2 Hazard Quotient (HQ)

Deterministic estimates of HQ for eight trace elements in water, wheat flour, and rice through

exposure to ingestion by adults in the agricultural and pastoral areas of Bay, Xinjiang, China, are displayed in Figure. 6 (a, b and c) and Tables S10, S11, and S12.

Drinking water

The result of the non-cancer risk assessment of drinking water (HQ) is shown in Figure. 6 a and Table S10. The average and 95% risk index of HQ of the non-cancer induced trace elements (Cu , Mn , Ni , Pb , Zn , Fe , Co , and Se) in drinking water and the average (50%) and 95% risk index of the total hazard quotient (HI) of these elements in both agricultural and pastoral areas did not exceed the respective safe reference doses (HQ or HI = 1) . So, there was small concern for potential non-carcinogenic health risks associated with overexposure in the drinking water in the present study areas. In both areas, the risk index of drinking water follows this order: Fe > Se > Co > Ni > Zn > Cu > Mn > Pb. For the exposed population in both areas, Fe, Co, and Se contributed the most to HI_w, but the rest of the metals (Cu , Mn , Ni , Pb , and Zn) made a minimum contribution to HI_w. Although Zn was characterized with the highest ADD value, it posed a least-noncarcinogenic risk due to its relatively high reference dose value, and the result was identical to those of previous studies (Zeng et al., 2015, Zhang et al., 2017).

The average and 95% non-cancer risk index (HQ) of Cu , Mn , and Pb from drinking water in the agricultural areas were higher than those in the pastoral areas, but the average and 95% non-cancer risk index (HQ) of Zn and Se, and the average and 95% non-cancer risk (HI) from drinking water (HZ_w) in the pastoral areas, were higher than those in the agricultural areas (Figure. 6a). The differences in the concentration levels of trace elements in the agricultural and pastoral areas were the main reasons for the risk difference. The differences

between the agricultural areas and pastoral areas in the 95% non-cancer risk of these elements were more significant than the average risk value of these elements.

Crop (wheat flour and rice)

The results of non-cancer risk (HQ) assessment of wheat flour is shown in Figure. 6b and Table S11. Although the average (50%) and 95% of the HQ index of trace elements (Cu , Mn , Ni , Pb , Zn , Fe , Co, and Se) exposure from flour in both agricultural and pastoral areas were less than 1 ($HQ < 1$), the average (50%) and 95% of the (total non-cancer risk)of these elements from flour (HIf) in both agricultural and pastoral areas exceeded the respective safe reference doses ($HQ > 1$). So there might be a concern for potential non-carcinogenic health risks associated with the total overexposure from the flour wheat. For the exposed population in both areas, Cu , Fe , and Mn contributed the most to the total HIf, followed by Zn , Co , and Pb. The rest of element such as the Se and Ni made a minimum contribution to HIf.

The result of HQ risk assessment of the rice is shown in Figure. 6c and Table S12. The result indicted that the average (50%) and 95% HQ index from trace elements (Cu , Mn , Ni , Pb , Zn , Fe , Co , and Se) exposure from rice in both the agricultural and pastoral areas were less than 1 ($HQ < 1$). The average (50%) and 95% HI of these elements from rice (HIr) in both the agricultural and pastoral areas were less than 1 ($HIr < 1$), so there was a less concern for potential non-carcinogenic health risks associated with the overexposure from the rice in this study area. For the exposed population in both areas, Cu , Fe , and Co contributed the most to HIr, followed by Zn , Mn , and Pb. The rest of Se and Ni made a minimum contribution to HIr.

The average (50%) of HQ from Mn , Pb , Se , and Ni from flour were higher in the pastoral

areas than those in the agricultural areas and the result were shown in Figure. 6b. The average (50%) and 95% HQ risk from Zn , Cu , Mn , Ni , Se , Co , and the average (50%) and 95% risk index from the total HQ index of the eight elements from rice (HI_r) were higher in the agricultural areas than those in the pastoral areas (Figure. 6c). The differences in levels of the trace elements between the agricultural and pastoral areas and the different sources of the rice and flour are the main reason for the risk difference.

3.3 Sensitivity analysis

Method of Percentage which calculates the percentage contribution of exposure pathways is used to determine which variables and pathways most strongly influence the risk estimate. Percentage contribution of exposure pathways to total risk is calculated by the following equation:

$$HI_{total} = \sum_{i=1}^n (HI_i) \quad (6)$$

$$\text{Precent contribution}_i = \frac{HI_i}{HI_{total}} * 100\%$$

The result indicated that in both areas, the risk index of flour made up the highest percentage both in the total cancer risk and the total non-cancer risk, followed by the risk index of rice and then the risk index of water (Table1).

The consumption of foods may vary considerably from one individual to another, and is dependent on culture and the availability of food species. The daily intake of elements from food consumption is dependent on the concentration of the element in food as well as the amount or frequency of consumption. From the survey, it was clear that most of the communities in both the agricultural and pastoral areas consumed much more wheat flour

than rice. Although most of the trace element concentration levels in the rice were higher than that of the wheat flour in both areas, the consumption of the flour was higher than that of the rice; thus, flour was the main pathway for the exposure to trace elements and their human health risks in this study area.

The residents in agricultural areas produced flour themselves but imported rice from outside the area, so the human health risk in the agricultural areas comes from both local and external sources. However, the residents in the pastoral areas imported both rice and flour from the outside regions, so the human health risk in pastoral areas came from an external source.

3.4 Source of Uncertainty

There are some uncertainties that may limit the validity of the case presented: (1) the concentrations of trace elements across the seasons are different, but seasonal variation of trace metals were not investigated; (2) CSF and RFD were treated as constants for all members of population, but they could vary from person to person; (3) the concentrations of total trace elements found in the water, rice, and flour samples were considered as bioavailable concentrations in the human body that could somewhat exaggerate the risk assessment; (4) in this study, we did not consider the metal speciation, but this speciation is one of the important factors for a more robust health risk estimation because the metal may perform in a number of species that exhibit different levels of toxicity.

Although the risk assessments in this study may not provide an absolutely accurate scenario of human health hazards, this study has provided useful information from a preliminary investigation of the health risk of trace elements in three pathways to adult populations in agricultural and pastoral areas. It also provides the basic evidence for [the] sustainable

agricultural development and management in Xinjiang, China.

4. Conclusions

All of the average concentrations of trace elements in the drinking water, rice, and flour in both the agricultural and pastoral areas in the present study were below the thresholds recommended by China's national guide. Most of the concentration levels of pollution elements and their variations and health risk in both the drinking water and rice in the agricultural areas were higher than those in the pastoral areas, while the element concentration levels and the health risk in the flour in the pastoral areas were higher than those in the agricultural areas. Most of the element concentration levels in the rice were higher than in the wheat flour but the health risk of the flour is higher than rice.

Although all the average concentration levels of the trace elements in the drinking water, rice, and flour were significantly lower than the tolerance limits of the national standard, the mean and 95% total cancer risk from the drinking water, rice, and flour, remained high and exceeded the safety reference level. For the exposed population, As contributed the most to the total cancer risk from the water, rice, and flour. There was little concern for potential non-carcinogenic health risks associated with overexposure to the drinking water and rice, but there might be a concern for potential non-carcinogenic health risks associated with total overexposure in the flour in the present study. For the exposed population, Cu, Fe, and Mn contributed the most to the total Hf. The risk index of flour took up the highest percentage, both in the total cancer risk and the total non-cancer risk, followed by the risk index of the rice and then the risk index of water.

The land reclamation from the pastoral areas to agriculture areas increase the trace elements

from drinking water and its health risk but may not influence the trace element concentration from crops and its health risk. The human health risk in the agricultural areas mainly came from the local environment, whereas the human health risk in the pastoral areas was mainly attributed to external regions.

Acknowledgments: I am grateful to Binggan Wei, Jing Wang, my father Tudi Aisha, my mother Gulibahaer Kuzi and my younger brother Mardanjan Tudi who helped with my field work. I thank Jiangping Yu and Hong-Zhi Zhang who gave some suggestions for my experiments. I also thank Dr. Anwar Aziz for his advice and assistance with technical issues and language correction during the course of research.

This study was supported by Key programme of national natural Science foundation of China (2016YFD0801004) and China's Agriculture Special Fund for Agro-Scientific Research for the Public Interest (201203012-6).

Author Contributions: Muyesser Turdi and Linsheng Yang conceived and designed the experiments; Muyesser Turdi performed the experiments; Muyesser Turdi analyzed the data; Muyesser Turdi Dung Tri Phung, Huada Daniel RUAN, Lin-sheng Yang, Hai-Jun Guo, Des Connell, Ross Sadler and Cordia Chu contributed reagents/materials/analysis tools; Muyesser Turdi wrote the paper.

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Fig1 Map of the study area

Fig.2 Box-whisker diagrams showing metal concentrations in drinking water, rice, flour and their comparison with national standards. (a) Represents for the drinking water; (b) represents for wheat flour; (c) represents for rice.

Drinking water national standards: WHO (WHO, 2006), USEPA (USEPA, 2012) and China drinking water standards (GB5749-2006).

Rice and flour national standards : (GB 2762-2012), (NY861-2004) and (FAO/WHO 1984)

Fig. 3 Concentrations of trace elements in drinking water in agricultural and pastoral areas of Bay country. Boxes with different letters in significance determined by T test and Non-parametric comparison (Kruskal –Wallis test).

Fig. 4 Concentrations of trace elements of rice and flour in agricultural and pastoral areas of Bay country. Boxes with different letters in significance determined by Non-parametric comparison Kruskal –Wallis test and Tukey HSD test

Fig. 5 Deterministic estimates of the cancer risk for adults through ingesting water, rice and flour in the agricultural and pastoral areas of Bay County, (a) represents drinking water, (b) represents wheat flour; and (c) represents rice

Fig. 6 Deterministic estimates of hazard quotients of drinking water, rice and flour through ingestion by adults in agricultural and pastoral areas, (a) representing drinking water, (b) representing wheat flour, and (c) representing rice.

Table 1 Sensitivity analysis result of identification of relative pathway contributed to total cancer risk and total non-cancer for adult group

Percent contribution of exposure pathway to TCR and HI				
Agricultural area	TCR point estimate		HI point estimate	
	Average TCR	% of total	HI	% of total
Exposure pathway				
Drinking water	1.84E-04	4.18%	4.94E-02	2.29
Rice	1.12E-03	25.40%	3.78E-01	17.52
Flour	3.10E-03	70.39%	1.73E+00	80.19
Total	4.40E-03	100	2.16E+00	100%
Pastoral area	TCR point estimate		HI point estimate	
	TCR	% of total	HI	% of total
Exposure pathway				
Drinking water	1.11E-04	1.98%	6.40E-02	2.91
Rice	7.92E-04	14.13%	2.67E-01	12.15
Flour	4.70E-03	83.99%	1.87E+00	84.93
Total	5.60E-03	100	2.20E+00	100