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RESEARCH ARTICLE

Potential carcinogenic and non-carcinogenic health hazards of metal(loid)s in food grains

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Abstract

Metal(loid) contamination of vital food grains such as wheat and rice is a very serious problem throughout the world because consumption of such contaminated food can lead to severe health effects in humans. Metal(loid) contamination of food crops can occur from different sources such as contaminated soil, irrigation water, and aerial deposition. Therefore, the present study was conducted to analyze potential non-carcinogenic and carcinogenic health impacts posed by different metal(loid)s (As Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Se, and Zn) via consumption of wheat and rice grown on metal(loid)-contaminated soils in areas around rivers (Beas and Sutlej) of Punjab, India. Among the metal(loid)s analyzed in wheat and rice samples, contents of As, Cd, Cr, Ni, and Pb were found to be above the international (FAO/WHO and EU) maximum permissible limits. The non-carcinogenic and carcinogenic health risk assessment of individual metal(loid)s revealed that As posed highest risk followed by Cd, Cu, Fe, Mn, and Pb. The values of indices calculated for analysis of combined non-carcinogenic, i.e., (hazard index; range 3.49–15.94) and carcinogenic (total carcinogenic risk index; range 8.30×10^{-4}–131.62×10^{-4}) risks for both crops were found to be many fold higher than the prescribed limits of 1.0 and 1.0×10^{-4}, respectively. Thus, the analysis of combined risks posed by metal(loid)s indicated that human population consuming wheat and rice from the study area faced both non-carcinogenic and carcinogenic health risks. Therefore, immediate steps must be taken to reduce the levels of metal(loid)s in wheat and rice from the study area.

Keywords Arsenic · Estimated daily intake · Food grains · Metal(loid)s · Risk analysis

Introduction

Metal(loid) contamination of different environmental components (soil, water, vegetation etc.) is becoming a grave cause of concern throughout the globe. Metal(loid)s in different environmental components ultimately affect human health via different routes. Among different metal(loid)s, non-essential metal(loid)s such as arsenic (As), cadmium (Cd), chromium (Cr), and lead (Pb) are well recognized to be toxic to humans and animals even at low concentrations causing mortality, carcinogenic, and reproductive effects (Chandra et al. 2009; Ma et al. 2016). In contrast, metals such as copper (Cu), iron (Fe), and zinc (Zn) are essentially required by humans and animals for various physiological and metabolic processes at relatively low concentrations (Giri and Singh 2017). However, at elevated concentrations, these essential metals can be toxic resulting in symptoms such as gastrointestinal bleeding, vomiting, diarrhea, damage to internal organs, immunodeficiency, and reproductive effects (Duruibe et al. 2007; Jaishankar et al. 2014). Humans can be exposed to

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metal(loid)s contamination through a number of mediums, including water, soil, air, and food, and the toxicity of metal(loid)s depends on several factor such as exposure concentration, chemical speciation, bioavailability, age, gender, genetics, and nutritional status of the exposed individual (Tchounwou et al. 2012; Fan et al. 2017).

The contamination of water sources (surface and groundwater) and soils by metal(loid)s due to anthropogenic activities (industrial, agricultural, mining, etc.) is well recognized problem in India (Pandey et al. 2016; Sharma et al. 2017). But, less attention has been given to food contamination (e.g., cereal grains) from pollutants and its implications to human health. Metal(loid) contamination of vital food grains (wheat and rice) due to different factors (use of polluted irrigation water, cultivation in contaminated soils, dust deposition, etc.) can lead to direct effects (short- and long-term) on human health following consumption (Antoniadis et al. 2017; Hu et al. 2017). In India, wheat is the most important food grain consumed in the northern regions, while rice is the most important cereal grain consumed in the southern parts of the country. India is the second largest producer of wheat grains with an annual production of approximately 90 million tons in 2016 (Gupta et al. 2016; Patowary et al. 2017). India is also one of the most prominent countries in rice production with an annual production of 109.5 million tons and production target of 140 million tons by year 2025 (Tiwari et al. 2018). The state of Punjab in northern India produces 16–18% of the wheat and 10–12% of rice in India (Government of India 2016). The shoots and husk of wheat and rice plants are also used as fodder for animals in Punjab, so these are also important crops for animal productivity. But, there are increasing community concerns throughout the globe including Punjab about the metal(loid) contamination in these food grains arising from anthropogenic activities and its impact on human health (short- and long-term). Cereal crops such as rice and wheat are known to uptake and bioaccumulate metal(loid)s from contaminated waters and soils (Ogunkunle et al. 2016). The potential non-carcinogenic and carcinogenic impacts to human health posed by metal(loid) concentrations in foods (e.g., cereals) can be assessed by calculating the estimated daily intake (EDI), target hazard quotient (THQ) (non-carcinogenic health hazard characterization), and carcinogenic risk index (CRI) (USEPA 2010; FAO/WHO 2011; Chamannejadian et al. 2013). The THQ and CRI provide an assessment of the potential non-carcinogenic and carcinogenic health risks posed by metal(loid)s concentrations individually and mixtures in foods (Bermudez et al. 2011; Moradi et al. 2015; Kumar et al. 2016; Liao et al. 2016; Khan et al. 2017; Yadav et al. 2017).

The major anthropogenic factors leading to metal(loid)s contamination in soils, water and food crops in Punjab include intensive agriculture (use of chemical fertilizers, pesticides, herbicides, and irrigation with contaminated river and groundwater), industrial activities (smoke, dust, and aerosol pollution and contamination of surface and ground water), and urbanization (Kaur et al. 2014a; Bhatti et al. 2016). The problem of metal(loid)s contamination of food crops is highly complex in areas around the main rivers of Punjab (Beas and Sutlej) due to the complexity of different point and non-point sources. In a previous study, we observed that metal(loid) contents in agricultural soils under wheat-rice cultivation collected from six villages around Harike wetland in Punjab, India, posed ecological and genotoxic risks (Bhatti et al. 2018). Since, wheat and rice are most consumed cereals in Punjab and other parts of country, further need was observed to analyze the metal(loid)s concentrations in wheat and rice growing in these contaminated soils.

Therefore, the present study was carried out with an aim to determine metal(loid)s concentrations in wheat and rice (grains) crops growing in the metal(loid)s contaminated agricultural soils (analyzed in the previous study) from Punjab, India, and assess potential impacts on human health (non-carcinogenic and carcinogenic health risk) via wheat and rice consumption.

Material and methods

Study area

The study area comprised of six villages situated along the rivers Beas and Sutlej in Punjab (two on the banks of river Beas upstream to Harike wetland, two on the banks of river Sutlej upstream to Harike wetland, and two downstream to the Harike wetland) (Fig. 1). These villages have fertile lands with sandy loam and calcisolic soils. The annual rainfall in this area is 435.6 mm and has semiarid to sub-humid climate. Agriculture is the main occupation of this area and has two main crop seasons Rabi (wheat) and Kharif (rice). Extensive use of agrochemicals such as fertilizers, pesticides, and herbicides (e.g., acetamiprid, carbendazim, glyphosate, clodinafop-propargyl) is widespread in this area. The upstream region of the study area consists of large cities like Ludhiana, Jalandhar, and Kapurthala having significant industrial (electroplating, dying, leather tanning, alloys, paints, etc.) and urban activities which causes contamination of rivers Beas and Sutlej by various industries and sewage (Kaur et al. 2014b; Bhatti et al. 2018).

Sampling and preparation

Grain samples were collected from agricultural farms in the six villages at a time when the wheat and rice crops were ready for harvesting, i.e., in the months of April 2013 (wheat) and October 2013 (rice). It was ensured that grains samples were unpolished and taken directly from farms so that no

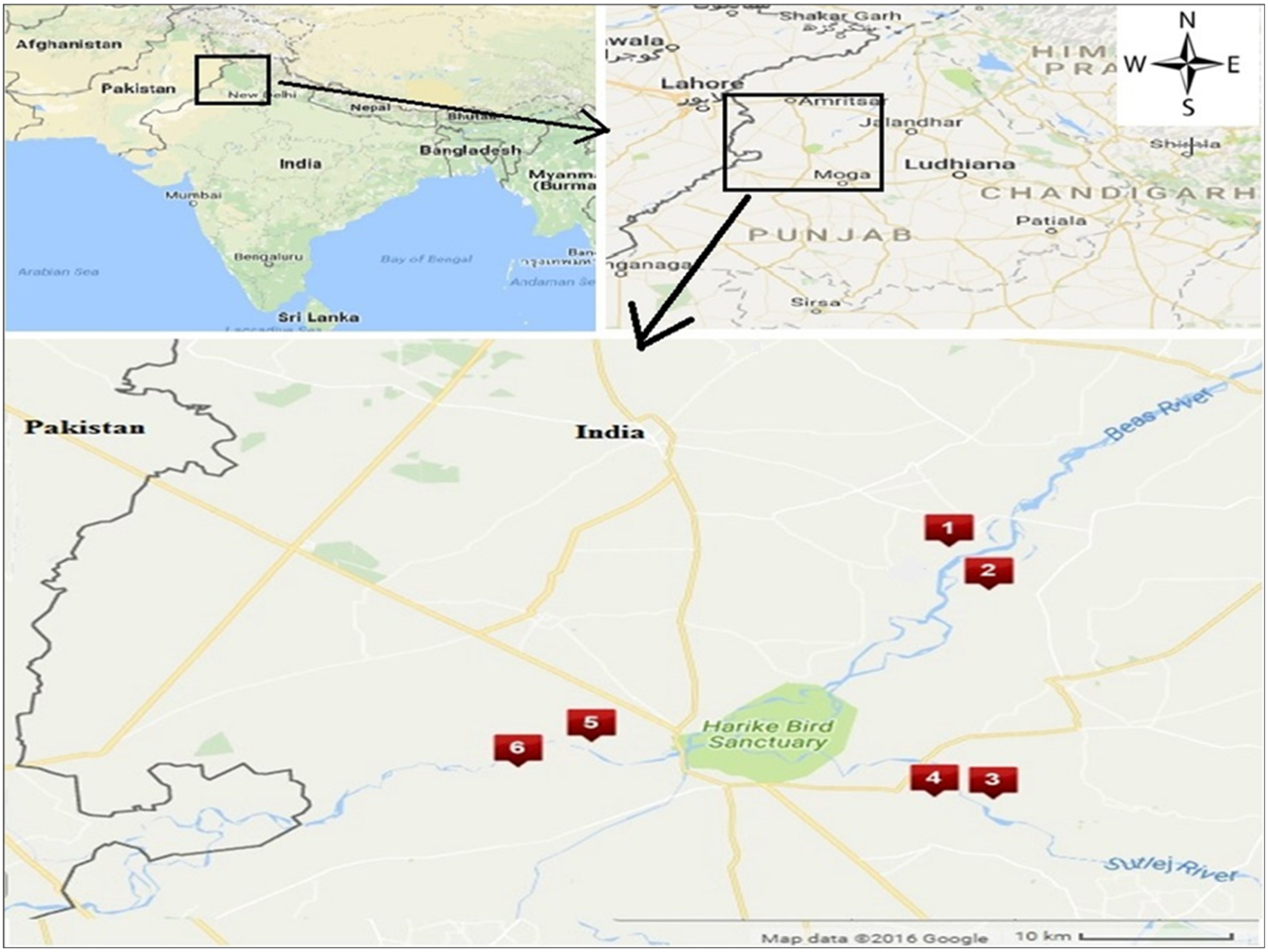


Fig. 1 Map of the sampling villages around Beas and Sutlej rivers, Punjab, India

discrepancy could occur in data. Wheat and rice grain samples were collected in triplicate from corresponding farms in villages where agricultural soil samples were collected. A composite grain sample at each farm was collected by randomly harvesting plants corresponding to approximately 50 g of grains (wheat and rice). The grains were separated from the ears in laboratory by hand thrashing, washed with ultrapure deionized water, oven-dried at 70 °C to a constant mass, and then ground to fine powder with pestle and mortar (Bermudez et al. 2011).

Total metal(loid) contents in crops samples

Total metal(loid) (As, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Se, and Zn) concentrations in crop samples were determined using a concentrated acid closed vessel microwave digestion procedure (US EPA method 3052, 1996). Approximately 0.5 g of plant material was weighed in a 50-ml PFA digestion vessel, and 10 ml of nitric acid was added. The samples were digested in a microwave oven (Milestone) using the following temperature and time program: 180 °C ramped over 10 min and then kept constant at 180 °C for 10 min. The samples were then cooled for 20 min, diluted to 50 ml using ultrapure deionized water (Milli-Q, Millipore), filtered to < 0.45 μm (Sartorius), and stored in a refrigerator until instrumental analysis for metal(loid) concentrations. The digested plant solutions were used to determine concentrations of selected metal(loid)s, i.e., arsenic (As), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), molybdenum (Mo), nickel (Ni), lead (Pb), selenium (Se), and zinc (Zn) using an inductively coupled plasma-optical emission spectrometer (ICP-OES) (iCAP 6000 series, Thermo Fisher Scientific) and inductively coupled plasma-mass spectrometer (ICP-MS) (Agilent 7700) at CSIRO, Australia. For quality assurance and quality control, a certified reference material (NIST 1567a), in-house standards, and duplicate samples were analyzed. Duplicate analysis was done for 10% samples for which the relative standard deviations for these replicate samples were less than 5% (Bhatti et al. 2018).

Potential effects of metal(loid)s in grains on human health

Estimated daily intake of metal(loid)s

The potential human health effects (non-carcinogenic and carcinogenic) posed by the consumption of wheat and rice grains contaminated with metal(loid)s were assessed by calculation of estimated daily intake (EDI, mg kg^{-1} $person^{-1}$ day^{-1}) (Giri and Singh 2017):

$$\mathrm{EDI} = C_{\mathrm{crop}} \times \mathrm{CI}/\mathrm{BW} \tag{1}$$

where, C_{crop} is the metal(loid) concentration in grains (wheat or rice) in mg/kg dry mass, CI is the daily intake rate of crop in kg/person/day, and BW is the average body weight in kg. The average BW was taken to be 60 kg for adult in the Punjab State (Yadav et al. 2017). The daily intake of wheat for adults was assumed to be 0.345 kg $person^{-1}$ day^{-1}, and for rice, it is 0.178 kg $person^{-1}$ day^{-1} (Bhatti et al. 2017; Yadav et al. 2017). The higher consumption rate of wheat is due to the fact that people in northern India, especially Punjab consume more wheat than rice on daily basis.

Non-carcinogenic health hazard characterization

Non-carcinogenic health hazard characterization of individual and mixture of metal(loid)s in the wheat or rice grains in Punjab were analyzed using a THQ which was calculated using the following equation (FAO/WHO 2011; Liao et al. 2016):

$$\mathrm{THQ} = (\mathrm{EDI} \times \mathrm{EF} \times \mathrm{ED})/(\mathrm{AT} \times \mathrm{RfD}) \tag{2}$$

where, EDI is the daily metal ingestion in mg/person/day (calculated in Eq. (1)), EF is the exposure frequency (365 days/year), ED is the average exposure duration (60 years), AT is the average exposure time for non-carcinogens (365 days per year × ED), RfD is the oral chronic reference dose in mg/kg body weight/day. The RfD values defined by USEPA/IRIS (2010) for As, Cd, Cu, Co, Cr, Fe, Mn, Mo, Ni, Pb, Se, and Zn are 0.0003, 0.001, 0.04, 0.02, 1.5, 0.7, 0.14, 0.005, 0.02, 0.0035, 0.004, and 0.3, respectively (Liao et al. 2016; Kumar et al. 2016; Fan et al. 2017).

In order to assess the non-carcinogenic health hazard of the mixture of metal(loid)s in wheat or rice grains, a hazard index (HI) was calculated according to following equation (Ogunkunle et al. 2016):

$$\mathrm{HI} = \mathrm{THQ}_{\mathrm{M1}} + \mathrm{THQ}_{\mathrm{M2}} + \mathrm{THQ}_{\mathrm{M3}}\mathrm{THQ}_{\mathrm{Mn}} \tag{3}$$

where, $\mathrm{THQ}_{\mathrm{M}}$ represents the target hazard quotient value of individual metal(loid) in the grain samples (i.e., separately for wheat and rice samples). A HI value > 1 suggests a greater probability of non-carcinogenic health effects occurring over the life time of exposure (Ogunkunle et al. 2016).

Carcinogenic health risk analysis

The potential human health risk of known carcinogens such as As, Cd, and Pb in grains (wheat or rice) was assessed using a CRI. The CRI was calculated according to the following equation (Islam et al. 2017):

$$\mathrm{CRI} = \mathrm{EDI} \times \mathrm{SF} \tag{4}$$

where, EDI is estimated daily intake of individual metal(loid) with grains (mg $kg^{-1}person^{-1}$ day^{-1}) as calculated in Eq. (1), and SF is the carcinogenic slope factor (upper bound approximating a 95% confidence limit, on the increased cancer risk from a lifetime exposure to an agent by ingestion or inhalation). The CRI was calculated only for As, Cd, and Pb as SF for other metals was not available. The values of SF for As, Cd, and Pb as defined by USEPA (2010) are 1.5, 6.1, and 0.0085, respectively (Liao et al. 2016; Islam et al. 2017).

The carcinogenic risk posed by mixture of metal(loid)s (As, Cd, and Pb) in grains was assessed by calculating total carcinogenic risk index (TCRI) according to following equation (Fan et al. 2017):

$$\mathrm{TCRI} = \sum \mathrm{CRI} \tag{5}$$

The acceptable or tolerable range for TCRI for regulatory purposes is within 10^{-6} to 10^{-4} (USEPA 2010; FAO/WHO 2011; Liao et al. 2016). If the value of TCRI exceeds the tolerable limit, the metal(loid)s in the food grains can pose unacceptable carcinogenic risk to human health.

Statistical analysis

The descriptive and other statistics of metal(loid) concentrations in wheat and rice grains was done using IBM SPSS (Statistical Package for the Social Sciences) software version 16.0 (New York, USA) and PAST (Paleontological Statistics) software version 3.06 (Hammer et al. 2001). One way ANOVA followed by Tukey's HSD test as post hoc was used to compare the means of metal(loid) contents in grains (wheat and rice) collected from the six villages. The difference between metal(loid)s contents in wheat and rice grains collected from a village was analyzed by Student's independent *t* test. Differences at $p < 0.05$ were considered statistically significant in both Tukey's HSD test and Student's *t* test. Pearson's correlation coefficients were calculated to analyze the correlation between metal(loid)s in wheat and rice grains samples. In case of metal(loid)s below detectable limit, a value of zero was assigned for correlation analysis.

Results and discussion

Metal(loid) contents in wheat and rice grains

The descriptive statistics of different metal(loid)s analyzed in the grain samples from the different villages in Punjab can be found in Table 1. In the present study, order of metal(loid)s on the basis of their average concentrations in wheat and rice grain samples was Fe > Mn > Zn > Cu > Cr > Ni > Pb > Mo > Co > Se > As > Cd. However, the order of metal(loid) concentrations in soil samples collected from farms where the wheat and rice samples were growing was different, i.e., Fe > Mn > Zn > Cr > Ni > Cu > Pb > Co > As > Se > Mo > Cd which was published in an earlier study (Bhatti et al. 2018). The order in crops was slightly different from the concentrations found in soil samples, which might be attributed to the selective uptake of these metal(loid)s by crop plants based upon their metabolic requirements and toxicity levels of metal(loid)s (Hu et al. 2017). The variation of most metal(loid) contents among the six villages were found to be statistically significant (ANOVA + Tukey's test) for both wheat and rice grains (Supplementary Table 1). This spatial variation of metal(loid) contents among the six villages can be attributed to the variation among the sources of the metal(loid)s in the six villages. In case of villages along Beas river (villages 1 and 2), the effects of industrial pollutants is slightly less in comparison to villages on the banks of river Sutlej (villages 3 and 4). The largest industrial city of Punjab, i.e., Ludhiana, is situated upstream to villages 3 and 4 on the bank of river Sutlej, and there are several reports regarding release of untreated industrial effluents from Ludhiana into Sutlej river and its tributaries which causes severe water pollution (Kaur et al. 2014b; Sharma and Walia 2018). Therefore, the effect of contamination of irrigation sources an farming practices can lead to the spatial variation of metal(loid) contents in these six villages (Supplementary Table 1). Statistically significant temporal variation (Student's t test; $p \leq 0.05$) was also observed among contents of most metal(loid)s in the grains of these two crops (wheat and rice) collected from the six villages (Supplementary Table 1). Among the different metal(loid)s, contents of As, Cr, and Mn were found to be higher in rice grains in comparison with wheat grains in most villages. Such variation in contents of metal(loid)s in wheat and rice grains collected from the same villages can be attributed to variations in metal(loid)s uptake capability of wheat and rice plants, quality of irrigation water in different seasons, application of different agrochemicals in these crops, differences in irrigation practices and variation in other point, and non-point sources of metal(loid)s in different seasons (Nagajyoti et al. 2010; Ogunkunle et al. 2016; Sharma et al. 2017). In case of rice, higher amount of irrigation water is required, and water stagnant conditions are provided in initial time of growth. So the exposure of rice to irrigation water is higher in comparison with wheat which increase the chances of uptake of metal(loid)s. Also, the present study area is prone to floods in monsoon season, and rice is grown in the times of monsoon to post-monsoon season. The flood water brings metal(loid)s from different point and non-point sources which further enhances the chances of metal(loid) exposure and accumulation in rice plants. Thus the spatial and temporal variation in the contents of metal(loid)s in grains of these two crops can be attributed to combination of various factors in the present study area. Pearson's correlation analysis of metal(loid)s in the grain samples indicated statistically significant positive correlation among the different metal(loid)s (Supplementary Table 2) in both crops which can be attributed to common sources of metal(loid)s, similar pathways of metal(loid) absorption from soils and interdependency of different metal(loid)s for different metabolic processes (Bhatti et al. 2017). Kumar et al. (2016) and Xing et al. (2016) also observed

Table 1 Descriptive statistics of metal(loid) contents (mg/kg) in grain (wheat and rice) samples collected from the six villages

			As	Cd	Co	Cr	Cu	Fe × 10^3	Mn	Mo	Ni	Pb	Se	Zn
Grains	Wheat	Min	< 0.08	< 0.02	0.09	1.50	7.20	0.083	18.13	0.29	0.30	0.30	0.07	19.30
		Max	0.22	0.04	0.35	8.09	19.50	0.424	37.53	0.91	2.27	1.82	0.32	42.47
		Mean	0.14	0.02	0.21	3.57	11.97	0.295	28.65	0.61	1.16	0.91	0.16	28.59
		SD	0.07	0.01	0.11	2.88	4.38	0.161	6.75	0.25	0.87	0.60	0.08	9.18
	Rice	Min	0.15	< 0.02	0.09	2.95	5.00	0.045	23.70	0.89	1.17	0.12	< 0.04	12.97
		Max	0.74	0.74	0.33	10.62	22.23	0.312	107.43	3.23	3.80	0.55	0.13	32.50
		Mean	0.41	0.02	0.18	6.51	11.27	0.135	50.37	1.75	2.07	0.29	0.07	21.44
		SD	0.23	0.01	0.09	2.92	6.65	0.098	30.23	0.82	1.05	0.19	0.04	7.24
Indian Limit of metal (mg/kg)[a]			1.1	1.5	–	20.0	30.0	–	–	–	1.5	2.5	–	50.0
International limits of metals (mg/kg)			0.7[b]	0.24[c]	50[c]	2.3[c]	73[c]	–	500[c]	–	67.90[c]	0.3[c]	–	99.40[c]

[a] Awashthi (2000)

[b] European Union (2002)

[c] FAO/WHO (2011)

statistically significant positive correlation among different metal(loid)s (As, Cd, Co, Cr, Cu, Ni, and Zn) in wheat and rice grains which was attributed to similar sources.

The concentrations of As, Cd, Cr, Ni, and Pb found in the several wheat and rice grains in the present study were above the international maximum permissible limits for food crops (FAO/WHO 2011) described in Table 1. These elevated metal(loid) concentrations found in grains may pose a significant health risk to the human population consuming these grains. The consumption of elevated As via food chain can lead to health effects such as inhibition in production of ATPs, increased risk of carcinogenicity, immunogenic disorders leading to nerve inflations and weakening of peripheral nervous system (Kantor 2006; Duruibe et al. 2007). Cadmium exposure from food can cause various toxic symptoms including bone defects such as osteomalacia and osteoporosis, increased blood pressure, myocardic dysfunctions, and different pulmonary and renal effects (Young 2005; Islam et al. 2017). The excessive exposure to Cr in food can lead to pathological conditions such as (i) inhibition of erythrocyte glutathione reductase which affects the capacity to reduce methemoglobin to hemoglobin, (ii) formation of ulcers in nasal septum, and (iii) defects in DNA and chromosomes (Jaishankar et al. 2014). Nickel in high amounts in foods can cause various metabolic dysfunctions in humans such as hypoglycemia, asthma, nausea, and headaches (ATSDR 1999; Yadav et al. 2017).

The concentrations of As, Cd, Cu, Ni, and Pb observed in grains of wheat and rice from Punjab in the present study were found to be higher in comparison with their contents observed at other locations in India and abroad (Supplementary Table 3). These locations in India and other countries include highly contaminated areas having activities such as mining (Liao et al. 2016; Giri and Singh 2017), lead smelting (Xing et al. 2016), and sewage water irrigation (Chandra et al. 2009). This study shows that different anthropogenic sources of contamination such as extensive agriculture using agrochemicals, pollution of irrigation sources by industrial and urban activities, and aerial deposition from various point and non-point sources are resulting in soil and grain contamination.

Potential effects of metal(loid)s in grains on human heath

Estimated daily intakes

For the analysis of the non-carcinogenic and carcinogenic health risks posed by metal(loid)s in the food crops, the EDI of metal(loid)s must be analyzed which describes the intake of individual metal(loid)s by humans via food consumption (Ogunkunle et al. 2016). These values can be compared with the maximum tolerable daily intake (TDI) values, issued by various organizations such as EU, WHO, and FAO. If the EDI values are above the TDI values for a metal(loid), it indicates excessive ingestion of that metal(loid) by humans, which can severely affect the human health (Antoniadis et al. 2017).

The EDI and TDI of the metal(loid)s for wheat and rice grain samples are presented in Table 2. The EDI values for As via rice consumption were found to be higher as compared with the wheat which can be attributed to higher accumulation of As by rice in comparison to other cereals (Fan et al. 2017). This higher accumulation can be due to easier uptake of As by rice plants from irrigation water in the flooded soils in which rice is grown (Lomax et al. 2012). Overall, the values of EDI for most metal(loid)s were found to be higher in case of wheat in comparison with rice in the six villages (Supplementary Table 4), which can be attributed to higher values of metal(loid)s in the grain samples and higher consumption of wheat (0.345 kg person^{-1} day^{-1}) in the study area in comparison with rice (0.178 kg person^{-1} day^{-1}). The average EDI values of Cr, Fe, Mn, and Ni were found to be higher than the provisional TDI values for both wheat and rice. The average EDI value of Pb was found to be above the provisional TDI value for wheat only. Thus, the contents of Cr, Fe, Mn, Ni, and Pb may pose health risk to the populations consuming the food grains from the study area.

Non-carcinogenic health risk analysis

The THQ analysis is a very informative tool to analyze the non-carcinogenic health risk posed by consumption of metal(loid) contaminated food crops (Khan et al. 2017). If the THQ values are above 1 for any metal(loid), it indicates very high risk due to that metal(loid) (Giri and Singh 2017). In the present study, average THQ values above 1 were observed for As and Mn for both wheat and rice grain samples (Table 2), while THQ values > 1 were observed for Mo in rice and Cu, Fe, and Pb in wheat samples. The highest THQ values were found to be for As in rice and wheat, which suggests that As poses highest non-carcinogenic risk due to consumption of grains sampled from the study area. Liao et al. (2016) also found maximum non-carcinogenic risk index values for As among the studied metal(loid)s in rice in Guangdong Province, China. Various other researchers from India and other countries also found THQ values above 1 for As, Cu, Fe, Mn, and Pb in wheat and rice samples (Bermudez et al. 2011; Moradi et al. 2015; Kumar et al. 2016; Giri and Singh 2017).

The HI provides an assessment of combined non-carcinogenic health risks due to metal(loid)s via food consumption (Huang et al. 2008). In the present study, the HI values (Fig. 2) for all six villages were found to be > 1 for both wheat and rice grain samples. The HI values for wheat grain samples ranged from 5.66 to 15.06, while for rice grain samples they ranged from 3.49 to 14.82. The HI was found to be lowest for villages 1 and 2 which can be attributed to lower levels of metal(loid)s in grain samples in these villages.

Table 2 Descriptive statistics of estimated daily intakes (EDI) and target hazard quotient (THQ) values for metal(loid)s in wheat and rice grains

			As	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	Se	Zn
EDI ($\times 10^{-3}$)	Wheat	Min	0.345	0.058	0.489	8.644	41.400	475.525	104.267	1.687	1.725	1.706	0.374	110.975
		Max	1.265	0.230	2.013	46.518	112.125	2436.083	215.817	5.233	13.033	10.465	1.821	244.183
		Mean	0.795	0.131	1.231	20.521	68.844	1696.410	164.738	3.527	6.676	5.242	0.925	164.418
		SD	0.401	0.056	0.644	16.562	25.179	924.154	38.793	1.462	4.996	3.451	0.484	52.793
	Rice	Min	0.455	0.024	0.267	8.752	11.778	133.698	70.310	2.630	3.461	0.346	0.059	38.468
		Max	2.195	0.158	0.969	31.496	65.959	925.600	318.719	9.592	11.273	1.622	0.396	96.417
		Mean	1.215	0.055	0.524	19.303	33.430	401.538	149.421	5.183	6.131	0.852	0.209	63.602
		SD	0.681	0.052	0.253	8.677	19.719	290.231	89.688	2.438	3.121	0.573	0.123	21.492
TDI ($\times 10^{-3}$)			2.1	1.0	500	3.0	500	800	140	600	5.0	3.6	5.0	1000
THQ	Wheat	Min	1.150	0.058	0.024	0.006	1.035	0.679	0.745	0.337	0.086	0.487	0.075	0.370
		Max	4.217	0.230	0.101	0.031	2.803	3.480	1.542	1.047	0.652	2.990	0.364	0.814
		Mean	2.651	0.131	0.062	0.014	1.721	2.423	1.177	0.705	0.334	1.498	0.185	0.548
		SD	1.338	0.056	0.032	0.011	0.630	1.320	0.277	0.292	0.250	0.986	0.097	0.176
	Rice	Min	1.516	0.024	0.013	0.006	0.294	0.191	0.502	0.526	0.173	0.099	0.012	0.128
		Max	7.318	0.158	0.048	0.021	1.649	1.322	2.277	1.918	0.564	0.463	0.079	0.321
		Mean	4.049	0.055	0.026	0.013	0.836	0.574	1.067	1.037	0.307	0.243	0.042	0.212
		SD	2.269	0.052	0.013	0.006	0.493	0.415	0.641	0.488	0.156	0.164	0.025	0.072

EDI and TDI units- mg kg^{-1} person^{-1} day^{-1}

TDI Tolerable daily intake

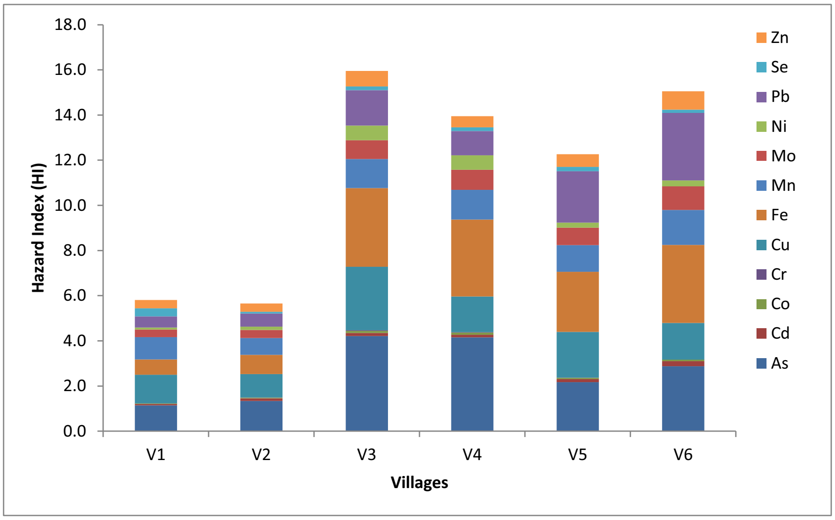
Among the metal(loid)s analyzed in the present study, maximum contribution to HI was from As in both the crops. Arsenic is a well-known toxic metalloid with carcinogenic and neurotoxic effects on human systems (Ma et al. 2016). Long-term exposure to As can cause formation of skin lesions, neurological problems, internal cancers, pulmonary diseases, hypertension, cardiovascular disease, and diabetes mellitus (Duruibe et al. 2007; Kumar et al. 2016). Huang et al. (2008) and Ma et al. (2016) from China and Kumar et al. (2016) and Giri and Singh (2017) from India have also found As to be a significant contributor to the total non-carcinogenic health risks posed by metal(loid)s in food grains (wheat and rice). The other metals having significant contributions (> 1) to HI were Fe, Mn, Cu, and Pb in wheat grains and Mn, Mo, and Cu in rice grains. Pb is also one of the most toxic metal which can cause several gastrointestinal, renal, and neurological disorders in humans (Duruibe et al. 2007). But, it was surprising to note that among the studied metal(loid)s, Cu, Fe, Mn, and Mo were significant contributors to the non-carcinogenic health risks due to food consumption in the present study. These metals are essential for human health for structural and catalytic components of proteins and have important roles in vital physiological activities (Huang et al. 2008; Khan et al. 2017). But at high concentrations, these metals can lead to toxic effects such as gastrointestinal bleeding, vomiting, diarrhea, and hepatic necrosis especially in children (Jaishankar et al. 2014). These metals are mainly of geogenic origins and have very less human contribution. Thus, while analyzing the anthropogenic influences on metal(loid) contents in food crops, the geogenic factors must also be taken into consideration during the contamination analysis as metal(loid)s having mainly geogenic origins can also contribute significantly to non-carcinogenic health risks. Different researchers from India and abroad also recorded significant contributions of these essential metals to non-carcinogenic health risks in previous studies on wheat and rice (Bermudez et al. 2011; Moradi et al. 2015; Liao et al. 2016; Ogunkunle et al. 2016).

The very high values of HI observed in the present study suggests that people consuming the wheat and rice grains grown in the study area have a high risk of non-carcinogenic health effects. Thus, prolonged consumption of these food grains from the study area can lead to severe implications to vital organ systems such as the renal, liver, cardiovascular, and nervous systems (Giri and Singh 2017).

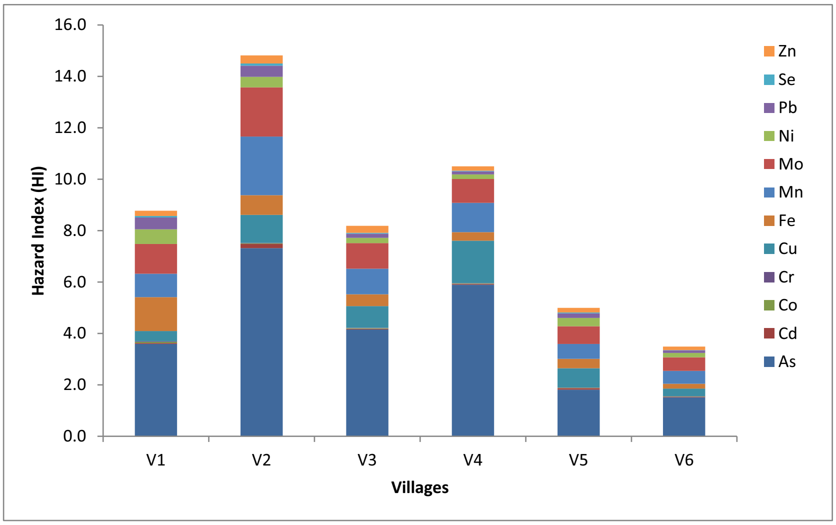
Carcinogenic health risk analysis

The CRI values calculated to assess the carcinogenic risk to human adults due to exposure to As, Cd, and Pb in wheat and rice grain samples are presented in Table 3. TCRI due to consumption of grains by human population of the six villages is presented in Fig. 3. The average CRI values of As, Cd, and Pb for wheat grains were found to be 1.193 × 10^{-3}, 0.799 × 10^{-3}, and 1.774 × 10^{-3}, respectively. For rice grains, the average CRI values for As, Cd, and Pb was found to be 1.822 × 10^{-3}, 0.333 × 10^{-3}, and 0.007 × 10^{-3}, respectively. The values of CRI for the three metal(loid)s were above the acceptable

Fig. 2 Hazard index (HI) for potential non-carcinogenic risk to humans due to metal(loid)s intake via consumption of food grains (wheat and rice) collected from six villages



a) Wheat grains



b) Rice grains

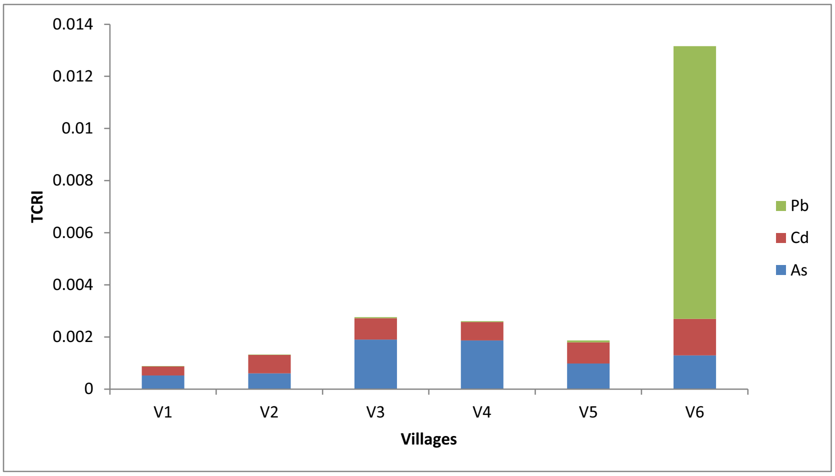
risk limit of 0.001×10^{-3} (USEPA 2010). Similar to the non-carcinogenic health risk, the highest carcinogenic risk in the present study is posed by As concentration in the wheat and rice grains from the study area.

Table 3 Descriptive statistics of carcinogenic risk index (CRI) for metal(loid)s in wheat and rice grains

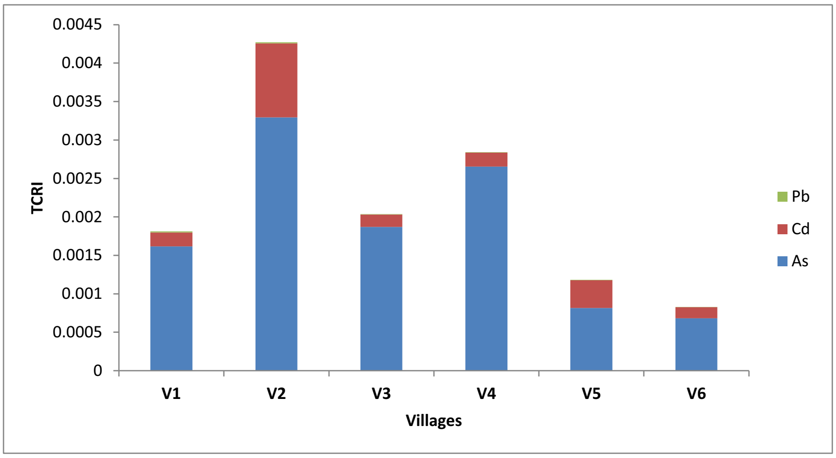
			As	Cd	Pb
CRI ($\times 10^{-3}$)	Wheat	Min	0.518	0.351	0.014
		Max	1.898	1.403	10.465
		Mean	1.193	0.799	1.774
		SD	0.602	0.342	4.258
	Rice	Min	0.682	0.145	0.003
		Max	3.293	0.965	0.014
		Mean	1.822	0.333	0.007
		SD	1.021	0.320	0.005

TCRI values which were calculated to assess the combined carcinogenic health risk of metal(loid)s due to consumption of wheat grains (8.83×10^{-4}–131.62×10^{-4}) and rice grains (8.30×10^{-4}–42.71×10^{-4}) were found to be significantly higher than the acceptable range of 1.0×10^{-6} to 1.0×10^{-4} (USEPA 2010). Such high values of TCRI indicated that the human population consuming wheat and rice grains from the study area was at a significantly high carcinogenic risk. Such high carcinogenic risk in the study area becomes far more dangerous considering the history of cancer cases from Malwa belt of Punjab, which is the neighboring region of the study area in the south of Punjab (Kumar et al. 2015). Hence, high values of TCRI observed in the present study suggest that further monitoring need to be done to determine the scale of metal(loid) contamination of food in the region

Fig. 3 Total carcinogenic risk index (TCRI) for potential carcinogenic risk to humans due to metal(loid)s intake via consumption of food grains (wheat and rice) collected from six villages



a) Wheat grains



b) Rice grains

and evaluate the risks posed by metal(loid)s due to the consumption of other food crops. Similar high TCRI values have been reported from other regions of the world receiving anthropogenic inputs of pollution (Liao et al. 2016; Ma et al. 2016; Islam et al. 2017) which indicate the imminent danger of contamination of these vital food sources with carcinogens.

Overall, the present study revealed that the wheat and rice grains analyzed from the areas around the two most prominent rivers of Punjab were contaminated with metal(loid)s such as As and Pb, which can pose significant non-carcinogenic and carcinogenic health risks to the human population consuming these crops. Hence, further monitoring and assessment of grains and other foods in this area need to be undertaken to determine if this was an isolated occurrence, its scale, and potential impact on human and animal health (short- and long-term). Furthermore, various industries and urban municipalities contaminating the irrigation (river water and groundwater) water with their effluents and sewage must be regulated. It must be ensured that only treated unpolluted water should be discharged in rivers Beas and Sutlej. There must also be monitoring and regulation of the levels of agrochemicals used by farmers in these areas, and organic manures must be promoted which have very less metal contents in them. Different metal(loid) removal techniques must be adopted for removing metal(loid)s from already contaminated soils of the study area. Among the different techniques for metal(loid) removal, phytoremediation is one of the most cost-friendly and effective technique for this purpose (Ali et al. 2013). Thus, economic schemes must be launched by the government to inspire farmers to plant those species which can efficiently remove metal(loid)s from soils for once in a year. Lastly, further analysis must be done to find the level of metal(loid) contamination in other crops such as vegetables which accumulate the metals at higher rate in comparison with wheat and rice.

Conclusions

In the present study, the concentrations of As, Cd, Cr, Ni, and Pb in grains (wheat and rice) were found to be above the international permissible limits for food. The EDI values for Cr, Fe, Mn, and Ni for both wheat and rice grains were found to be above the TDI which indicated that consumption of these food grains can lead to ingestion of these metals above tolerable limits. The THQ values for As, Cu, Fe, Mn, Mo, and Pb were found to be > 1 which mean that these metal(loid)s in grain samples posed a significant non-carcinogenic health risks to humans individually. The CRI values for As, Cd, and Pb were found to be above the permissible limits indicating a high carcinogenic risk posed by these metal(loid)s to humans individually. Arsenic was found to be the most prominent metal(loid) which could cause health related issues in humans consuming food grains from the study area. The analysis of combined non-carcinogenic (HI) and carcinogenic (TCRI) risks posed by metal(loid)s in the present study indicated that the human population consuming the wheat and rice grains from the study area was under very high carcinogenic and non-carcinogenic health risks, which can lead to severe health problems. Hence, immediate and effective steps must be taken to reduce the concentrations of these metal(loid)s and further monitoring must be carried out to determine the scale of food contamination by metal(loid)s and evaluate the risks posed by metal(loid)s due to the consumption of other food crops.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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