RESEARCH ARTICLE

Human health risk assessment due to dietary intake of heavy metals through rice in the mining areas of Singhbhum Copper Belt, India

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Abstract The study was intended to investigate heavy metal contamination levels in the rice grown in the vicinity of the mining areas of Singhbhum Copper Belt, India. The concentrations of the metals were below the Indian maximum allowable concentrations for food except for Pb, Ni, and Zn at some locations. Principal component analysis extracted three factors explaining 79.1% of the data variability. The extracted factors suggested that the sources of metals in the rice can be attributed to soil, irrigating water, and atmospheric dust deposition. High potential health risks of metal exposure from rice consumption were illustrated based on estimated daily intake (EDI) and target hazard quotient (THQ). The daily intakes of heavy metals for local adults were higher than the tolerable daily intakes provided by WHO in some samples for Cr, Fe, Ni, and V. Considering the geometric mean of the metals in rice samples of the study area, the hazard index (HI) for adult was above unity (3.09). Pb, Cu, and Cr were the key components contributing to potential non-carcinogenic risk. The HI varied from 2.24 to 12.7 among the locations indicating an appreciable heath risk to the consumers of the locally grown rice around the mining areas.

Keywords Rice \cdot Heavy metals \cdot Copper mining areas \cdot Principal component analysis . Risk assessment

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Introduction

A significant source of heavy metal contamination of the environment is mining and processing of metal ores (Dudka and Adriano [1997;](#page-9-0) Navarro et al. [2008](#page-10-0); Singh et al. [2005](#page-10-0)). Excessive buildup of heavy metals in agricultural soils around mining areas either by aerial depositions or by irrigation is likely to induce a corresponding elevation of metals in the harvested crops (Nan et al. [2002](#page-10-0)). The accumulation of the metals in the crops either can be from the contaminated soils and water via plant roots or can be through direct deposition of contaminants from the atmosphere onto plant surfaces (McBride [2003\)](#page-10-0). Since a major route for heavy metal exposure to human is through soil–crop–food pathway, cultivation of crops on contaminated soil can potentially lead to the uptake and accumulation of trace metals in the edible plant parts and then exert potential risk to the local inhabitants (Gupta and Gupta [1998](#page-10-0); Adriano [2001](#page-9-0); McLaughlin et al. [1999;](#page-10-0) Järup [2003;](#page-10-0) Monika and Katarzyna [2004;](#page-10-0) McBride [2007\)](#page-10-0).

Earlier studies show that heavy metal pollution of mined areas caused health damage to the local inhabitants (Kachenko and Singh [2006;](#page-10-0) Liu et al. [2005](#page-10-0); Pruvot et al. [2006\)](#page-10-0). It is known that serious systemic health problems like malfunction of organs and chronic syndromes may be caused by ingestion of relatively low doses of toxic heavy metals over a long period (Oliver [1997\)](#page-10-0). Thus, information about heavy metal concentrations in food products and their dietary intake is very important for assessing their risk to human health (Zhuang et al. [2009a](#page-11-0), [b](#page-11-0)).

As per the statistical data provided by the Food and Agriculture Organization (FAO) ([2004](#page-9-0)), approximately 30% of the dietary energy supply and 20% of the dietary protein intake around the world are provided by rice. As rice is the main staple food in India, especially in the region of our study area, heavy metals in rice may have a major contribution to the

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total metal intake. Therefore, there is an essentiality for the study of heavy metal levels in rice from the copper mining areas of East Singhbhum. The objectives of this study were to investigate the concentrations of metals in rice (Oryza sativa L.) samples in the mining areas of Singhbhum Copper Belt in India and to assess the associated potential health risk to local inhabitants through consumption of this crop. The hazard quotient (HQ) and hazard index (HI) were used to assess noncarcinogenic health risks from individual metals and the combined health risk from all the studied metals, respectively.

Materials and methods

Site description

The study was carried out in the vicinity of mining and processing industries in the Singhbhum Copper Belt of Jharkhand, which is rich in mineral resources and copper, iron, uranium, gold, and kyanite.

Sampling and analysis

Samples were collected from 14 locations in the environs of the copper mining and processing facilities (Fig. [1\)](#page-2-0). At each site, approximately 500 g of rice was collected in April 2016. All the samples were collected in triplicates. Rice grain samples were washed with deionized water. The rice grain without hull was oven-dried at 70 °C for 72 h and then ground with an agate mortar to fine powder. A microwave-assisted digestion procedure was used (Schuhmacher et al. [1993\)](#page-10-0). Of homogenized samples, 0.5 g was digested under pressure in Teflon vessels with 6 mL of nitric acid and 2 mL of hydrogen peroxide. On completion of the digestion and after adequate cooling, solutions were filtered and made up to 50 mL with 1% nitric acid. Each rice sample was collected along with the corresponding soil and water samples for correlation purposes. Each soil sample with about 1 kg was taken from the depths of 0–15 cm, which represented the plow layer. Then, the soil samples were packed and sealed in polyethylene bags and brought to the laboratory, where they were air-dried at room temperature (25–30 $^{\circ}$ C). The coning and quartering method was employed for mixing the samples, and about 100 g of the sample was taken. The samples were then transferred into a porcelain dish and dried in an oven at 110 °C for 24 h. The samples were powdered in a dry mortar–pestle and sieved through standard sieve of 200 mesh size. The samples were then subjected to digestion in microwave by the method 3052 as given by USEPA ([1996](#page-11-0)) for total metal concentration. Aliquots were preserved for the analysis of metals. Water samples for metal analysis were collected, filtered with a Millipore filter paper (pore size 0.45 lm), and preserved by

lowering the pH to >2 by addition of 6 N ultrapure nitric acid (Radojevic and Bashkin [1999](#page-10-0)).

Determination of metals

Concentrations of As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Se, V, and Zn were determined in rice, water, and soil samples using inductively coupled plasma-mass spectrometry (ICP-MS, PerkinElmer Elan DRC-e). A calibration blank and an independent calibration verification standard were analyzed every 15 samples to confirm the calibration status of the ICP-MS. Matrix interference (blank) was <1% for all elements. Triplicates of sample analysis yielded relative percent differences of <5%.

Quality assurance and quality control

Standard reference material (SRM) of rice flour (SRM 1568b) supplied by the National Institute of Standards and Technology (NIST), USA, was used for validation of the analytical procedure. The heavy metal contents found in the SRMs were in good agreement with the certificate values, which confirmed the feasibility of the analytical protocols in the determination of heavy metals in rice (Table [1\)](#page-3-0). The accuracy and precision were checked by analyzing the certified reference material under the same conditions. The percentage of recovery varied from 92.5 to 105.9.

Statistical analysis of the data

All statistical analyses were done using SPSS 16.0 for Windows software package. Prior to statistical analyses, all datasets were checked for normality of distribution using the curve of accumulated frequency (Miller and Miller [1989](#page-10-0)). As recurrently encountered in environmental samples, the distributions of the concentrations of the metals were not elucidated by normal distribution and were better represented by the lognormal distribution. Thus, the central tendency is represented by the geometric mean (Wayne [1990](#page-11-0)). The correlation analysis was conducted by Pearson correlation. Correlation is significant at the 0.01 level (two tailed).

Principal component analysis

Principal component analysis (PCA) is a variation reduction technique in which a number of measured parameters can be transformed into a small number of artificial variables known as principal components (PC). The number of extracted PC is equal to the number of variables included in the analysis; however, normally, PCs having eigenvalues >1 are taken into consideration as they account for the maximum variance in the observed parameter (Huang et al. [2014](#page-10-0); Kolsi et al. [2013\)](#page-10-0). Varimax rotation is applied to all the extracted PC to reduce

Fig. 1 Map of the study area with sampling locations

a Reference values

the contribution of the variables which are not significant (Closs and Nichol [1975](#page-9-0)). Each variable considered for PCA has a weight factor associated with it. This weight factor, also referred to as PC score, is the correlation between the original variable and a factor. A PC score close to ± 1 indicates a strong correlation between the given variable and the factor.

Metal pollution index

To examine the overall heavy metal concentrations in the rice samples of a location a, metal pollution index (MPI) was computed (Eq. 1). This index was obtained by calculating the geometrical mean of concentrations of all the metals in the rice (Usero et al. [1997\)](#page-11-0).

$$
MPI (mg/kg) = (Cf1 × Cf2 ×Cfn)1/n
$$
 (1)

where Cf_n = concentration of metal *n* in the sample.

Estimated daily intake of heavy metals through rice consumption

The daily intake of metals depends on both the metal concentration in food and the daily food consumption. In addition, the body weight of the human can influence the tolerance of pollutants. The estimated daily intake (EDI) is a concept introduced to take into account these factors. The EDI value for an adult was calculated with the Eq. (2) according to Song et al. [2009](#page-11-0) as follows:

$$
EDI = (C_{\text{metal}} \times W_{\text{rice}}) / B_{\text{w}}
$$
 (2)

where C_{metal} is the concentration of the heavy metals in the rice samples, W_{rice} stands for the daily average consumption of rice in this region, and B_w represents the body weight. Based on the National Sample Survey of India (NSSO [2014\)](#page-10-0), the

average daily intake of rice was 199 g per day. The average value of B_w used was 52 kg for Indian man (Jain et al. [1995;](#page-10-0) Dang et al. [1996](#page-9-0)).

Risk from the intake of metals through ingestion: target hazard quotient

Non-carcinogenic health risks from consumption of rice by the local inhabitants were assessed based on the target hazard quotient (THQ) (Eq. 3). THQ values were calculated by comparing the estimated daily intake (EDI) dose (EDI) (milligrams per kilogram body weight per day) of a chemical to a reference dose $(R_fD,$ milligrams per kilogram per day).

$$
THQ = EDI/R_fD \tag{3}
$$

If the ratio is less than 1, the exposed population is unlikely to experience obvious adverse effects. The method of estimating risk using THQ was provided in the US EPA Region III risk-based concentration table (USEPA [2007](#page-11-0)).

Health risk assessment of multiple metals

The HI was used to estimate total non-carcinogenic health risks of multiple metals on the assumption of dose additivity (Eq. 4).

$$
HI = THQ1 + THQ2 + \dots + THQn \tag{4}
$$

where THQi is the target hazard quotient of an individual element of metals, HI is the total hazard index for all the 12 metals studied in the present study, and n is 12.

Although dose additivity usually requires that all components act by the same mechanism, the HI is commonly also used as a screening tool for components with the same critical target without regard to mechanism and even for components with different target organs. The HI method does not take into account interactions among the components of the mixture and thus may underestimate the health hazard if the interactions are greater than additive or overestimate the health hazard if the interactions are less than additive (Wilbur et al. [2004\)](#page-11-0).

Results and discussion

Distribution of metals in rice

A wide range of metal concentrations were observed in the rice samples collected from the 14 locations around the copper mining areas of East Singhbhum (Table [2](#page-4-0)). The average concentration of triplicate samples of each location is given in the table. The geometric mean concentration (mg kg⁻¹, dry weight basis) of As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Se, V, and Zn was 0.065, 0.008, 0.011, 0.281, 4.56, 49.8, 7.3, 0.461, 0.792, 0.033, 0.045,

Table 2 Heavy metal concentrations of rice samples in milligram per kilogram collected from mining areas of Singhbhum Copper Belt (each value is average of 3 samples)

Locations	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Se	V	Zn
Bhatin	0.008	0.019	0.082	0.139	0.67	27.9	5.23	0.75	0.208	0.013	0.001	20.0
Kuldiha	0.096	0.077	0.506	1.309	0.82	157.3	59.03	2.89	0.012	0.006	0.038	8.7
Darisai	0.071	0.001	0.034	0.434	9.93	97.0	15.74	0.77	1.298	0.041	0.076	45.6
Bodamdera	0.017	0.003	0.028	0.332	0.43	38.4	7.71	0.01	1.751	0.002	1.821	7.4
Rakha	0.080	0.003	0.003	0.218	5.03	286.2	2.63	0.22	0.784	0.154	3.002	18.4
Jagganathpur	0.033	0.072	0.002	0.230	3.30	12.3	6.45	1.33	0.472	0.046	0.008	10.5
Kendadih	0.112	0.003	0.003	0.238	12.52	215.4	3.51	0.23	1.072	0.065	1.028	29.2
Terenga	0.142	0.043	0.069	0.438	16.45	166.5	11.30	1.29	5.177	0.084	0.326	114.6
Benashol	0.142	0.050	0.018	0.244	7.96	47.6	6.77	1.37	2.211	0.091	0.059	68.7
Kutludih	0.056	0.003	0.001	0.339	9.09	24.0	2.24	0.65	1.229	0.113	0.003	57.9
Chirugoda	0.162	0.010	0.003	0.172	9.93	32.5	10.59	0.44	2.339	0.046	0.004	87.8
Pathergoda	0.067	0.002	0.025	0.270	3.73	21.1	9.19	0.58	0.698	0.063	0.050	17.1
Latia	0.064	0.002	0.001	0.194	4.38	8.8	7.46	0.29	0.606	0.145	0.004	17.9
Badia	0.127	0.010	0.014	0.186	18.08	41.0	8.98	0.36	1.264	0.002	0.026	30.8

and 27.3, respectively. The metals generally ranked in the order $Fe > Zn > Mn > Cu > PB > Ni > Cr > As > V > Se > Co > Cd.$ The concentrations of metals in the rice samples exceeded the limits of Indian standards for food (Awasthi [2000\)](#page-9-0) for Ni and Pb (1.5 and 2.5 mg kg^{-1} , respectively) in one sample each. In 29% of the samples, the concentration of Zn exceeded the Indian limit of 50 mg kg−¹ . All the other metal concentrations in rice samples were well within the limits of Indian standards for food.

Table [3](#page-5-0) provides the concentrations of metals in the rice samples of the present study compared with those found in some previous studies. The studies represented the areas with strong anthropogenic influence like industrial and mining activities. The metal concentrations in the rice samples varied largely among the different studies, which may be attributed to geology of the area, concentration of metals in the soil and irrigating water, and the anthropogenic activities of the area. However, the metal concentrations of the rice samples in the present study were in range of the other studies.

On analyzing the data, it can be observed that the concentration of the metals varied spatially. The highest concentration of Cd, Co, Cr, Mn, and Ni was observed at Kuldiha, which is located in close proximity to a railway line and a busy roadway. Terenga, which is under both copper mining and vehicular influences, recorded the highest levels of Pb and Zn. Cu concentration was found to be highest at Badia, which is very near to a closed copper mine.

Heavy metal concentrations in water and soil

Water and soil samples were collected from the same locations as that of rice samples for correlation studies. The statistical data along with the standards are presented in Table [4.](#page-6-0) The comparison of the metal concentrations with the average concentration of metals in the shale (Turekian and Wedepohl [1961\)](#page-11-0) depicted that the levels of almost all the metals exceeded the values. As the study area is characterized by the presence of metal-bearing formations and mining and industrial operations, it is expected to have elevated levels of metals in the soil (Forstner and Wiltman [1983](#page-9-0); Preston and Chester [1996](#page-10-0)). However, the concentrations of metals do not exceed the irrigation water standards for metals as given by FAO ([1985](#page-9-0)).

Correlation between heavy metal levels in soil and water with rice

To assess the association between the different matrices, i.e., rice, water, and soil, correlations of the metals were determined and Table [5](#page-6-0) presents the coefficients between the different matrices. The results suggested a strong association of As, Cu, Pb, and Zn between the rice samples and corresponding soil samples while a strong correlation for Fe, Se, and V was observed between the rice and water samples. Positive correlations of metals in crops with soils and irrigating water have been reported by many workers (Adams et al. [2004;](#page-9-0) Liu et al. [2016;](#page-10-0) Wang et al. [2003](#page-11-0); Khan et al. [2008;](#page-10-0) Singh and Kumar [2006\)](#page-10-0). Since the uptake mechanism of metals includes both adsorption (from soil) and absorption (from water) and takes place from the roots, the uptake of metals through roots depends on the presence of metal concentration in water as well as in the soil. However, the other studied metals, Cd, Co, Cr, Mn, and Ni, were correlated with neither water nor soil.

Principal component analysis

Principal component analysis (PCA) was carried out on the heavy metal data of the rice samples in order to better

Table 4 Concentration of heavy metals in water and soil (total metals) collected from mining areas of Singhbhum Copper Belt

	As	C _d	Co	Cr	Cu	Fe	Mn	Ni	Pb	Se	V	Zn
Water (unit μ g/l)												
Minimum	0.14	N.D.	0.24	0.04	0.42	316.0	28.8	2.0	N.D.	0.08	0.44	9.7
Maximum	4.18		13.74	3.14	41.4	2676.0	3537.9	40.8		2.66	11.1	1139.2
Geometric mean	1.02		2.29	0.36	3.06	928.9	273.9	9.9		0.67	2.13	71.6
Irrigation standard FAO ^a	100			100	200		200	200				2000
Soil (unit mg kg^{-1})												
Minimum	31.2	0.08	12.1	65.2	54.3	20,147	111.7	35.2	21.3	ND	65.3	52.9
Maximum	182.5	0.84	63.8	751.0	3314.4	64,695	1045.0	293.1	102.6		200.8	621.4
Geometric mean	74.5	0.32	31.0	150.3	190.3	37,187	489.4	88.4	45.5		110.4	191.5
Average shale ^b	13	0.3	19	90	45	47,200	850	68	20	0.6	130	95

ND not detected

a FAO [\(1985\)](#page-9-0)

^b Turekian and Wedepohl [\(1961\)](#page-11-0)

understand their interrelationships and to explore the reduction of the experimental variables. The purpose of PCA is to reduce the dimensionality of the dataset, since a few of the new components explain the major part of the variation of the data. It also helps in assigning source identity to each one of the PCs (Miller and Miller [2000;](#page-10-0) Yuan et al. [2013,](#page-11-0) [2014](#page-11-0)).

The PC loadings, the cumulative percentages for rotated matrix, and the factor scores are presented in Table [6.](#page-7-0) There were three components with eigenvalues >1, which accounted for 79.1% of the total variance. The loading factors indicate that the first principal component (PC1) was associated with Cd, Co, Cr, Mn, and Ni explaining 36.5% of variance. The factor seems to be associated with air deposition. A number of studies have reported the increase of metals in the cereals and vegetables due to the atmospheric dust deposition (Harrison and Chirgawi [1989;](#page-10-0) Camazano et al. [1994](#page-9-0); Voutsa et al. [1996](#page-11-0); Pandey and Pandey [2009;](#page-10-0) Feng et al. [2011](#page-9-0); Sharma et al. [2008\)](#page-10-0). The second component explained 26.6% of variation in the data and is underlined by significant factor loading for As, Cu, Pb, and Zn while the third factor having high loadings for Fe, V, and Se explicated 16% of the variance. The PC2 and PC3 can be related to the concentration of the metals in the soil and irrigating water, respectively. This suggests that the water is more conducive for Fe, V, and Se for their occurrence in the rice samples while for As, Cu, Pb, and Zn, soil environment is more contributing for their uptake in the rice samples of the study area. This is in also correspondence to the correlation

studies which deduced similar inference. However, the concentration of metals in soil, water, or air of an area in turn may be dependent on geogenic sources and anthropogenic sources like mining, industrial, and vehicular activities (Yuan et al. [2013;](#page-11-0) Giri and Singh [2016\)](#page-10-0).

Metal pollution index and human exposure to metals through rice

MPI is suggested to be a reliable and precise method for metal pollution monitoring in dietary components. It has been used to study the difference of total metals between the locations or species (Singh et al. [2010;](#page-10-0) Usero et al. [1997\)](#page-11-0). In the present study, MPI is used to evaluate the variation of total metals among the locations (Fig. [2](#page-7-0)). The highest MPI was calculated at Terenga followed by Benashol. Both the locations are in the close proximity to copper mining and processing industries and a highway.

Estimation of daily intake of heavy metals by consuming rice

To evaluate the health risk associated with heavy metal contamination of rice grown in the vicinity of copper mining areas of East Singhbhum, estimated dietary intake (EDI) and target hazard quotients (THQs) were calculated with an assumption that local population consumes the local rice. The comparison

Table 5 Pearson correlation coefficients of heavy metals between rice and different matrices

	As	Cd	Co.	Cr	Cu.	Fe	Mn	Ni	P _b	Se		Zn
Soil (total metal)	$0.73^{\rm a}$	-0.01	-0.31	0.06	$0.72^{\rm a}$	0.17	0.34	-0.26	0.88 ^a	$\overline{}$	-0.16	0.96°
Water	0.17	$\hspace{0.1mm}$	-0.24	-0.31	-0.15	$0.63^{\rm a}$	-0.23	-0.18	$\overline{}$	0.81 ^a	$0.73^{\rm a}$	0.42

a Correlation significant at the 0.01 level (two tailed)

Table 6 Principal component loadings (varimax normalized) for the metals in the rice samples collected from mining areas of Singhbhum Copper Belt

	PC1	PC ₂	PC ₃
As	0.184	0.852	0.117
Cd	0.755	0.059	-0.178
Co	0.951	-0.188	0.057
Cr	0.942	-0.093	0.127
Cu	-0.194	0.837	0.013
Fe	0.231	0.174	0.913
Mn	0.900	0.016	-0.114
Ni	0.933	0.054	-0.141
Pb	-0.066	0.859	0.053
Se	-0.381	0.175	0.502
V	-0.251	-0.219	0.869
Zn	-0.043	0.931	-0.089
Eigenvalues	4.476	3.148	1.866
% Total variance	36.5	26.6	16.0
Cumulative variance	36.5	63.1	79.1

of EDI with provisional tolerable daily intake (PTDI) as provided by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) ([1999](#page-10-0), [2003](#page-10-0)) for all the studied metals is shown in Table [7.](#page-8-0) In the present study, it is to be noted that the R_fD of 3, 5, and 5 µg/kg day⁻¹ for Cr, Se, and V, respectively, established by USEPA [\(2011](#page-11-0)) is taken as the PTDI value because of their unavailable PTDI data. The PTDI and RfD values for arsenic as established by WHO and EPA are for inorganic arsenic. However, the arsenic content estimated in the rice samples was total arsenic, which includes both organic and inorganic types. It is estimated that 35% of the total arsenic in rice is inorganic (WHO Environmental Health Criteria 224 [2001\)](#page-11-0). Since there is no standard value for total arsenic intake for an adult, we have calculated the EDI and HQ for As taking this factor into consideration.

From the EDI of the studied heavy metals through the consumption of rice, it can be suggested that the consumption

Fig. 2 Metal pollution Index in different locations

of average amounts of the rice from the study area does not pose a health risk for the consumers as the values obtained are below the FAO/WHO (JECFA [1999](#page-10-0)) limits for heavy metal intake based on the body weight of an average adult. The present study showed that the contributions of the local rice consumed in the area to daily intake of As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Se, V, and Zn were 4.1, 3.06, 0.008, 35.8, 3.5, 2.8, 19.9, 35.3, 84.9, 2.51, 3.42, and 10.4%, respectively, of PTDI as suggested by JECFA. Lower dietary intake was observed for Cd, Co, Cu, Se, and V as compared to other metals. Maximum EDI was depicted for Pb followed by Cr and Ni. The EDI value of Pb is high in all the locations studied, and highest EDI was calculated for Terenga, which was about 5.5 times more than the tolerable intake. The other metals whose EDI was exceeding the PTDI at some of the locations were Cr, Fe, Ni, and V. Thus, perennial intake of these contaminated cereals is likely to induce adverse health effects arising largely from Pb, Ni, and Cr exposure at some of the locations.

Risk assessment of heavy metals by consuming rice

THQ has been recognized as a useful parameter for evaluation of risk associated with the consumption of metalcontaminated food crops (Rupert et al. [2004](#page-10-0); Sridhara Chary et al. [2008](#page-11-0); Zhuang et al. [2009a](#page-11-0), [b\)](#page-11-0). The THQ values of heavy metals by consuming rice for an individual adult are shown in Table [8](#page-8-0). The results show that the mean THQ values for the metals are below 1, highest being for the Pb (0.87). The THQ for the metals from rice consumption is in decreasing order of $Pb > Cu > Cr > Zn > As > Fe > Mn > Co > Ni > V > Cd > Se.$ However, at some of the locations, the THQ values for Co, Cr, Cu, Fe, Pb, V, and Zn are higher than 1, which showed that the inhabitants around the copper mining areas are experiencing relatively high health risk. The local consumers may also be at risk due to the combination of several heavy metals. The HI values for all the locations varied from 2.24 to 12.7 with a geometric mean value of 3.09 indicating an appreciable risk on the human body. Pb, Cu, and Cr are the key components contributing to potential non-carcinogenic risk while V, Cd,

Table 7 Estimated dietary intake (EDI) of heavy metals by consuming locally grown rice from mining areas of Singhbhum Copper Belt

Locations	Heavy metals											
	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Se	V	Zn
Bhatin	0.010	0.073	0.314	0.532	2.58	106.6	20.0	2.87	0.80	0.048	0.004	76.5
Kuldiha	0.128	0.295	1.937	5.009	3.13	601.9	111.1	11.07	0.04	0.022	0.144	33.5
Darisai	0.095	0.002	0.128	1.661	38.0	371.3	60.2	2.95	4.97	0.157	0.290	174.6
Bodamdera	0.022	0.011	0.105	1.271	1.63	146.8	29.5	0.02	6.70	0.009	6.968	28.4
Rakha	0.107	0.012	0.011	0.832	19.3	1095.3	10.0	0.83	3.00	0.588	11.487	70.5
Jagganathpur	0.044	0.276	0.007	0.880	12.6	47.0	24.7	5.08	1.80	0.177	0.032	40.1
Kendadih	0.150	0.011	0.011	0.910	47.9	824.1	13.4	0.88	4.10	0.247	3.934	111.6
Terenga	0.191	0.164	0.263	1.676	63.0	637.0	43.2	4.94	19.81	0.321	1.246	438.7
Benashol	0.190	0.190	0.069	0.932	30.5	182.0	25.9	5.23	8.46	0.349	0.225	262.8
Kutludih	0.075	0.012	0.004	1.295	34.8	91.7	8.6	2.48	4.70	0.432	0.011	221.5
Chirugoda	0.217	0.038	0.013	0.658	38.0	124.5	40.5	1.69	8.95	0.174	0.014	335.9
Pathergoda	0.089	0.009	0.096	1.034	14.3	80.7	35.2	2.21	2.67	0.240	0.192	65.4
Latia	0.086	0.008	0.001	0.743	16.7	33.6	28.6	1.11	2.32	0.556	0.014	68.4
Badia	0.170	0.037	0.055	0.711	69.2	157.0	34.4	1.39	4.84	0.006	0.098	117.7
Mean EDI ^a	0.088	0.031	0.04	1.074	17.46	190.7	27.9	1.764	3.03	0.125	0.171	104.3
PTDI ^b	2.14	1	500	3	500	800	140	5	3.57	5	5	1000

a EDI values (in μg/kg body wt/day)

^b PTDI values (in μg/kg body wt/day) of all the metals were based on the data suggested by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) ([1999](#page-10-0), [2003](#page-10-0)). The PTDI value of Cr, Se, and V was based on the reference dose (RfD) established by US Environmental Protection Agency ([2011\)](#page-11-0)

and Se are the least contributing. Heavy metals have a toxic impact, but detrimental impacts become apparent only when long-term consumption of contaminated food occurs. Thus, a prolonged consumption of unsafe concentrations of heavy metals through cereals may lead to the chronic accumulation of heavy metals in the kidney and liver of humans causing disruption of numerous biochemical processes, leading to cardiovascular, nervous, kidney, and bone diseases. It is therefore

Table 8 Target hazard quotients (THQs) of heavy metals by consuming locally grown rice from mining areas of Singhbhum Copper Belt

Locations	THQ												H1
	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Se	V	Zn	
Bhatin	0.035	0.073	1.0460	0.177	0.064	0.152	0.143	0.144	0.228	0.010	0.001	0.255	2.33
Kuldiha	0.426	0.295	6.4579	1.670	0.078	0.860	0.794	0.554	0.013	0.004	0.029	0.112	11.29
Darisai	0.318	0.002	0.4273	0.554	0.950	0.530	0.430	0.147	1.419	0.031	0.058	0.582	5.45
Bodamdera	0.075	0.011	0.3508	0.424	0.041	0.210	0.211	0.001	1.915	0.002	1.394	0.095	4.73
Rakha	0.356	0.012	0.0351	0.277	0.481	1.565	0.072	0.041	0.857	0.118	2.297	0.235	6.35
Jagganathpur	0.145	0.276	0.0223	0.293	0.316	0.067	0.176	0.254	0.516	0.035	0.006	0.134	2.24
Kendadih	0.500	0.011	0.0383	0.303	1.198	1.177	0.096	0.044	1.172	0.049	0.787	0.372	5.75
Terenga	0.635	0.164	0.8770	0.559	1.574	0.910	0.309	0.247	5.660	0.064	0.249	1.462	12.71
Benashol	0.633	0.190	0.2296	0.311	0.761	0.260	0.185	0.261	2.418	0.070	0.045	0.876	6.24
Kutludih	0.249	0.012	0.0128	0.432	0.870	0.131	0.061	0.124	1.344	0.086	0.002	0.738	4.06
Chirugoda	0.722	0.038	0.0446	0.219	0.950	0.178	0.289	0.084	2.558	0.035	0.003	1.120	6.24
Pathergoda	0.297	0.009	0.3189	0.345	0.357	0.115	0.251	0.110	0.763	0.048	0.038	0.218	2.87
Latia	0.286	0.008	0.0032	0.248	0.419	0.048	0.204	0.055	0.662	0.111	0.003	0.228	2.27
Badia	0.566	0.037	0.1818	0.237	1.730	0.224	0.246	0.070	1.382	0.001	0.020	0.392	5.09
Mean HQ	0.291	0.031	0.135	0.358	0.436	0.272	0.200	0.088	0.866	0.025	0.034	0.348	3.09
R_fD values	0.0003	0.001	0.0003	0.003	0.04	0.7	0.14	0.02	0.0035	0.005	0.005	0.3	

suggested that regular monitoring of heavy metals in dietary components should be performed in order to prevent excessive buildup of these heavy metals in the human food chain.

It must be noted that our assessment only considered the intake of toxic elements via rice consumption. In fact, humans are also exposed to heavy metals through the dietary intake of other food items such as wheat, vegetables, fruit, fish, meat, eggs, milk, and water (Wang et al. [2005](#page-11-0); Zheng et al. [2007](#page-11-0); Chary et al. 2008; Sipter et al. [2008\)](#page-10-0). Furthermore, there are other pathways for exposure to metals such as dust inhalation, dermal contact, and ingestion (for children) of metalcontaminated soils (Grasmück and Sholz [2005;](#page-10-0) Hellström et al. [2007](#page-10-0)). Therefore, the overall potential health risks due to heavy metals to local residents are far beyond the scope of our present study.

Conclusion

From the estimation of metal concentration in the rice samples collected from this study, we obtained a better knowledge regarding the impact of the copper mining and processing operations on the environment and the potential risk to human health. The concentrations of all the metals in the rice grains were below the national MAL except for Pb, Ni, and Zn at some locations. The metal concentration in the corresponding soil samples exceeded the average shale values for almost all the metals. The results furthermore suggested a strong association of the As, Cu, Pb, and Zn between the soil and the rice samples and Fe, Se, and V between water and soil samples. PCA suggests that the source of metals in the rice can be attributed to soil, irrigating water, and atmospheric dust deposition. High potential health risks of heavy metal exposure from rice consumption were depicted based on the estimated daily intake (EDI) of heavy metals and the target hazard quotient (THQ). The estimations of daily intakes of heavy metals for local adults were higher than the tolerable daily intakes provided by WHO at some of the locations for Cr, Fe, Ni, and V indicating potential health risk for the local inhabitants through consumption of contaminated food crops. The local consumers are also likely to be at risk due to the synergistic effect of the metals. The HI values for all the locations varied from 2.24 to 12.7 with a geometric mean value of 3.09 indicating an appreciable risk on the human body. Thus, the local inhabitants due to long-term consumption of the rice grown in vicinity of the copper mining area may bear a potentially serious health risk, which would further elevate if other locally grown food items are also considered. Relevant data for this are still limited, and the implications for human health should be identified by a further detailed study.

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Compliance with ethical standards

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

References

- Abbas ST, Sarfraz M, Mehdi SM, Hassan G, Rehman O (2007) Trace elements accumulation in soil and rice plants irrigated with the contaminated water. Soil Tillage Res 94(2):503–509
- Adams M, Zhao FJ, McGrath SP, Nicholson FA, Chambers BJ (2004) Predicting cadmium concentrations in wheat and barley grain using soil properties. J Environ Qual 33:532–541
- Adriano DC (2001) Trace elements in terrestrial environments: biogeochemistry, bioavailability and risks of metals, Second edn. Springer-Verlag, Berlin
- Al-Saleh I, Shinwari N (2001) Report on the levels of cadmium, lead, and mercury in imported rice grain samples. Biol Trace Elem Res 83:91– 96
- Awasthi SK (2000) Prevention of Food Adulteration Act No. 37 of, 1954 Central and State rules as amended for 1999. Ashoka Law House, New Delhi
- Camazano MS, Martin MJS, Lorenzo LF (1994) Lead and cadmium in soils and vegetables from urban gardens of Salamanca (Spain). Sci Total Environ 146(147):163–168
- Cao H, Chen J, Zhang J, Zhang H, Qiao L, Men Y (2010) Heavy metals in rice and garden vegetables and their potential health risks to inhabitants in the vicinity of an industrial zone in Jiangsu, China. J Environ Sci 22(11):1792–1799
- Chary NS, Kamala CT, Raj DSS (2008) Assessing risk of heavy metals from consuming food grown on sewage irrigated soils and food chain transfer. Ecotox Environ Safe 69:513–524
- Closs LG, Nichol I (1975) The role of factor and regression analysis in the interpretation of geochemical reconnaissance data. Can J Earth Sci 12(8):1316–1330
- Dang HS, Jaiswal DD, Parameswaran M, Deodhar KP, Krishnamony S (1996) Age dependent physical and anatomical Indian data for application in internal dosimetry. Radiat Prot Dosim 63:217–222
- Dudka S, Adriano DC (1997) Environmental impacts of metal ore mining and processing: a review. J Environ Qual 26:590–602
- Fangmin C, Ningchun Z, Haiming X, Yi L, Wenfang Z, Zhiwei Z, Mingxue C (2006) Cadmium and lead contamination in japonica rice grains and its variation among the different locations in southeast China. Sci Total Environ 359:156–166
- FAO (1985) Food and Agriculture Organization. Water quality for agriculture. Irrigation and Drainage Paper No. 29, Rev. 1. Rome, United Nations
- FAO (2004) Food and Agriculture Organisation, FAOSTAT, FAO Statistical Databases. <http://apps.fao.org/>
- Feng JF, Wang YX, Zhao J, Zhu LQ, Bian XM, Zhang WJ (2011) Source attributions of heavy metals in rice plant along highway in Eastern China. J Environ Sci 23(7):1158–1164
- Forstner U, Wiltman GTW (1983) Metal pollution in aquatic environment. Springer, Berlin
- Fu J, Zhou Q, Liu J, Liu W, Wang T, Zhang Q, Jiang G (2008) High levels of heavy metals in rice (Oryza sativa L.) from a typical E-waste recycling area in southeast China and its potential risk to human health. Chemosphere 71:1269–1275
- Giri S, Singh AK (2016) Spatial distribution of metal(loid)s in groundwater of a mining dominated area: recognising metal(loid) sources and assessing carcinogenic and non-carcinogenic human health risk. Int J Environ Anal Chem 96:1313–1330
- Grasmück D, Sholz RW (2005) Risk perception of heavy metal soil contamination by high-exposed and low-exposed inhabitants: the role of knowledge and emotional concerns. Risk Anal 25:611–622
- Gupta U, Gupta S (1998) Trace element toxicity relationships to crop production and livestock and human health: implications for management. Commun Soil Sci Plant Anal 29:1491–1522
- Hang X, Wang H, Zhou J, Ma C, Du C, Chen X (2009) Risk assessment of potentially toxic element pollution in soils and rice (Oryza sativa) in a typical area of the Yangtze River Delta. Environ Poll 157:2542– 2549
- Harrison RM, Chirgawi MB (1989) The assessment of air and soil as contributors of some trace metals to vegetable plants I. Use of a filtered air growth cabinet. Sci Total Environ 83:13–34
- Hellström L, Persson B, Brudin L, Petersson Grawé K, Öborn I, Järup L (2007) Cadmium exposure pathways in a population living near a battery plant. Sci Total Environ 373:447–455
- Huang G, Chen Z, Sun J (2014) Water quality assessment and hydrochemical characteristics of groundwater on the aspect of metals in an old town, Foshan, South China. J Earth Syst Sci 123(1):91–100
- Jain SC, Mehta SC, Kumar B, Reddy AR, Nagaratnam A (1995) Formulation of the reference Indian adult: anatomical and physiological data. Health Phys 68:509–522
- Järup L (2003) Hazards of heavy metal contamination. Brit Med Bull 68: 167–182
- JECFA (1999) Joint FAO/WHO Expert Committee on Food Additives,. Reports of the 53rd meeting of the Joint FAO/WHO Expert Committee on Food Additives, JECFA/53/TRS. Rome, Italy
- JECFA (2003) Joint FAO/WHO Expert Committee on Food Additives, Summary and conclusions of the 61st meeting of the Joint FAO/ WHO Expert Committee on Food Additives. JECFA/61/SC. Rome, Italy
- Kachenko AG, Singh B (2006) Heavy metals contamination in vegetables grown in urban and metal smelter contaminated sites in Australia. Water Air Soil Pollut 169:101–123
- Khan S, Cao Q, Zheng YM, Huang YZ, Zhu YG (2008) Health risks of heavy metals in contaminated soils and food crops irrigated with wastewater in Beijing, China. Environ Poll 152:686–692
- Kolsi SH, Bouri S, Hachicha W, Dhia HB (2013) Implementation and evaluation of multivariate analysis for groundwater hydrochemistry assessment in arid environments: a case study of Hajeb Elyoun-Jelma, Central Tunisia. Environ Earth Sci 70:2215–2224
- Lin HT, Wong SS, Li GC (2004) Heavy metal content of rice and shellfish in Taiwan. J Food Drug Anal 12(2):167–174
- Liu HY, Probst A, Liao BH (2005) Metal contamination of soils and crops affected by the Chenzhou lead zinc mine spill (Hunan, China). Sci Total Environ 339:153–166
- Liu J, Zhang X, Tran H, Wang D, Zhu Y (2011) Heavy metal contamination and risk assessment in water, paddy soil, and rice around an electroplating plant. Environ Sci Pollut Res 18:1623–1632
- Liu Z, Zhang Q, Han T, Ding Y, Sun J, Wang F, Zhu C (2016) Heavy metal pollution in a soil-Rice system in the Yangtze River region of China. Int J Environ Res Public Health 13:63. doi:[10.3390/](http://dx.doi.org/10.3390/ijerph13010063) [ijerph13010063](http://dx.doi.org/10.3390/ijerph13010063)
- McBride MB (2003) Toxic metals in sewage sludge-amended soils: has promotion of beneficial use discounted the risks? Adv Environ Res 8:5–19
- McBride MB (2007) Trace metals and sulfur in soils and forage of a chronic wasting disease locus. Environ Chem 4:134–139
- McLaughlin MJ, Parker DR, Clarke JM (1999) Metals and micronutrients-food safety issues. Field Crops Res 60:143–163
- Miller JC, Miller JN (1989) Statistics for analytical chemistry, Second edn. Ellis, Horwood
- Miller NJ, Miller JC (2000) Statistics and chemometrics for analytical chemistry, Fourth edn. NJ7 Pearson Education, Englewood Cliffs
- Monika DP, Katarzyna SK (2004) Histopathological changes in the liver, kidneys, and testes of bank voles environmentally exposed to heavy metal emissions from the steelworks and zinc smelter in Poland. Environ Res 96:72–78
- Nan Z, Zhao C, Li J, Chen F, Sun W (2002) Relations between soil properties and elected heavy metal concentrations in spring wheat (Triticum aestivum L.) grown in contaminated soils. Water Air Soil Pollut 133:205–213
- Navarro MC, Perez-Sirvent C, Martinez-Sanchez MJ, Vidal J, Tovar PJ, Bech J (2008) Abandoned mine sites as a source of contamination by heavy metals: a case study in a semi-arid zone. J Geochem Explor 96:183–193
- NSSO (2014) National Sample Survey Office, Household Consumption of various goods and services in India 2011–12, NSS 68th Round, Ministry of Statistics and Programme Implementation, Government of India
- Oliver MA (1997) Soil and human health: a review. Eur J Soil Sci 48: 573–592
- Pandey J, Pandey U (2009) Accumulation of heavy metals in dietary vegetables and cultivated soil horizon in organic farming system in relation to atmospheric deposition in a seasonally dry tropical region of India. Environ Monit Assess 148:61–74
- Preston MR, Chester R (1996) Chemistry and pollution of the marine environment. In: Harison RM (ed) Pollution: causes, effects and control, Third edn. Royal Society of Chemistry, UK, pp 26–51
- Pruvot C, Douay F, Herve F, Waterlot C (2006) Heavy metals in soil, crops and grass as a source of human exposure in the former mining areas. J Soils Sediments 6:215–220
- Radojevic M, Bashkin VN (1999) Practical environmental analysis. Royal Society of Chemistry, London
- Rupert LH, Neil B, Scott DY, Neil MJC, Andrew MT, Ann MM et al (2004) Assessing potential risk of heavy metal exposure from consumption of home-produced vegetables by urban populations. Environ Health Perspect 112:215–221
- Schuhmacher M, Domingo JL, Llobet JM, Corbella J (1993) Dietary intake of copper, chromium and zinc in Tarragona Province, Spain. Sci Total Environ 132:3–10
- Sharma RK, Agrawal M, Marshall FM (2008) Heavy metal (Cu, Zn, Cd and Pb) contamination of vegetables in urban India: a case study in Varanasi. Environ Poll 154(2):254–263
- Singh S, Kumar M (2006) Trace metal load of soil, water and vegetables in peri-urban Delhi. Environ Monit Assess 120:79–91
- Singh AN, Zeng DH, Chen FS (2005) Heavy metal concentrations in redeveloping soil of mine spoil under plantations of certain native woody species in dry tropical environment, India. J Environ Sci 1: 168–174
- Singh A, Sharma RK, Agrawal M, Marshall FM (2010) Health risk assessment of heavy metals via dietary intake of foodstuffs from the wastewater irrigated site of a dry tropical area of India. Food Chem Toxicol 48:611–619
- Singh J, Upadhyay SK, Pathak RK, Gupta V (2011) Accumulation of heavy metals in soil and paddy crop (Oryza sativa), irrigated with water of Ramgarh Lake, Gorakhpur, UP, India. Toxicol Environ Chem 93(3):462–473
- Sipter E, Rózsa E, Gruiz K, Tátrai E, Morvai V (2008) Site-specific risk assessment in contaminated vegetable gardens. Chemosphere 71: 1301–1307

Song B, Lei M, Chen T, Zheng YM, Xie YF, Li XY, Gao D (2009) Assessing the health risk of heavy metals in vegetables to the general population in Beijing, China. J Environ Sci (China) 21:1702–1709

Sridhara Chary N, Kamala CT, Samuel Suman RD (2008) Assessing risk of heavy metals from consuming food grown on sewage irrigated soils and food chain transfer. Ecotoxicol Environ Saf 69(3):513–524

- Turekian KK, Wedepohl KH (1961) Distribution of the elements in some major units of the earth's crust. Am Geol Soc Bull 72:175–182
- USEPA (1996) United States Environmental Protection Agency, EPA Method 3052: microwave assisted acid digestion of siliceous and organically based matrices. U.S. Environmental Protection Agency, Office of Solid Waste and Emergency Response, U.S. Government Printing Office, Washington, USA
- USEPA (2007) United States Environmental Protection Agency, Integrated Risk Information System database; Philadelphia PA; Washington, USA
- USEPA (2011) United States Environmental Protection Agency, Riskbased concentration table, Washington, USA
- Usero J, Gonzalez-Regalado E, Gracia I (1997) Trace metals in the bivalve mollusks Ruditapes decussates and Ruditapes philippinarum from the Atlantic Coast of southern Spain. Environ Int 23(3):291– 298
- Voutsa D, Grimanis A, Samara C (1996) Trace elements in vegetables grown in an industrial area in relation to soil and air particulate matter. Sci Total Environ 94:325–335
- Wang Q, Cui Y, Liu X, Dong Y, Christie P (2003) Soil contamination and plant uptake of heavy metals at polluted sites in China. J Environ Sci Health, Part A 38(5):823–838
- Wang XL, Sato T, Xing BS, Tao S (2005) Health risks of heavy metals to the general public in Tianjin, China via consumption of vegetables and fish. Sci Total Environ 350:28–37
- Wayne RO (1990) A physical explanation of the lognormality of pollutant concentrations. J Air Waste Manag Assoc 40:1378–1383
- WHO (2001) World Health Organization, Environmental health criteria 224: arsenic and arsenic compounds: chapter 5. Environment levels and human exposure, 2nd edn. Geneva
- Wilbur SB, Hansen H, Pohl H, Colman J, McClure P (2004) Using the ATSDR guidance manual for the assessment of joint toxic action of chemical mixtures. Environ Toxicol Pharmacol 18:223–230
- Yang QW, Shu WS, Qiu JW, Wang HB, Lan CY (2004) Lead in paddy soils and rice plants and its potential health risk around Lechang lead/zinc mine, Guangdong, China. Env Int 30(7):883–889
- Yang QW, Lan CY, Wang HB, Zhuang P, Shu WS (2006) Cadmium in soil–rice system and health risk associated with the use of untreated mining wastewater for irrigation in Lechang. China Agri Water Manag 84:147–152
- Yap DW, Adezrian J, Khairiah J, Ismail BS, Mahir RA (2009) The uptake of heavy metals by paddy plants (Oryza sativa) in Kota Marudu, Sabah, Malaysia. American-Eurasian J Agric Environ Sci 6(1):16–19
- Yuan GL, Sun TH, Han P, Li J (2013) Environmental geochemical mapping and multivariate geostatistical analysis of heavy metals in topsoils of a closed steel smelter: Capital Iron & Steel Factory, Beijing, China. J Geochem Explor 130:15–21
- Yuan GL, Wu HZ, Fu S, Han P, Lang XX (2014) Persistent organic pollutants (POPs) in the topsoil of typical urban renewal area in Beijing, China: status, sources and potential risk. J Geochem Explor 138:94–103
- Zeng F, Ali S, Zhang H, Ouyang Y, Qiu B, Wu F, Zhang G (2011) The influence of pH and organic matter content in paddy soil on heavy metal availability and their uptake by rice plants. Environ Poll 159: 84–91
- Zheng N, Wang QC, Zhang XW, Zheng DM, Zhang ZS, Zhang SQ (2007) Population health risk due to dietary intake of heavy metals in the industrial area of Huludao City, China. Sci Total Environ 387: 96–104
- Zhuang P, McBride BB, Xia HP, Li NY, Li ZA (2009a) Health risk from heavy metals via consumption of food crops in the vicinity of Dabaoshan mine, South China. Sci Total Environ 407:1551–1561
- Zhuang P, Zou B, Li NY, Li ZA (2009b) Heavy metal contamination in soils and food crops around Dabaoshan mine in Guangdong, China: implication for human health. Environ Geochem Health 31:707–715