

Human and Ecological Risk Assessment: An International Journal

ISSN: 1080-7039 (Print) 1549-7860 (Online) Journal homepage: <http://www.tandfonline.com/loi/bher20>

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To cite this article: Poonam Yadav, Balvinder Singh, V. K. Garg, Suman Mor & Vandana Pulhani (2017) Bioaccumulation and health risks of heavy metals associated with consumption of rice grains from croplands in Northern India, Human and Ecological Risk Assessment: An International Journal, 23:1, 14-27, DOI: [10.1080/10807039.2016.1218750](https://doi.org/10.1080/10807039.2016.1218750)

To link to this article: <https://doi.org/10.1080/10807039.2016.1218750>



Published online: 23 Sep 2016.



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Bioaccumulation and health risks of heavy metals associated with consumption of rice grains from croplands in Northern India

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ABSTRACT

The present study focused on micronutrients and nonessential toxic heavy metals quantification and their bio-concentration in soil–rice system in Haryana, India. Composite samples of rice grains and paddy fields' soil were collected, processed and analyzed for heavy metals. Nutrient and heavy metals' content of paddy fields' soil varied over a wide range, having highest Fe content followed by Cu > Cr > Pb > Ni > Zn > Co > Cd. Most of the metals viz. Fe, Zn, Cu and Co found profusely in rice grains were essential micronutrients. Bio-concentration factors (BCFs) in rice grains were in the following order: Zn (0.11) > Co (0.10) > Cu (0.08) = Cr (0.08) > Ni (0.02) > Pb (0.011) > Cd (0.009) > Fe (0.003), indicating higher accumulation of micronutrients than non-essential toxic heavy metals. Principal component analysis revealed three factors accounting for 70% of the total variance which supported correlation among electrical conductivity (EC)-Na, EC-cation exchange capacity (CEC), CEC-Na, pH-TOC, and BCF_{Cu}-BCF_{Zn}. Health risk indices anticipated that the grains are safe for consumption as non-carcinogenic hazard quotients were less than unity for all the studied heavy metals.

ARTICLE HISTORY

Received 10 June 2016
Revised manuscript
accepted 26 July 2016

KEYWORDS

bio-concentration factor;
hazard index; health risk;
heavy metals; reference oral
dose

Introduction

Soil acts as a natural buffer system, controlling the transport of chemical elements and substances into various components of biosphere as well as geochemical sink for contaminants (Smuc *et al.* 2012). Soil is a main source of nutrients to plants. It contains macronutrients and micronutrients that are essential for the growth of the plant and ultimately for sustenance as food for human beings and animals. Major nutrient metals such as Fe, Zn, Ni, Co and Cu are necessary up to a certain defined range of concentrations, below which they cause malnutrition and above which they become toxic. In the past few decades due to increased developmental activities, soil regularly gets inundated with various types of contaminants including heavy metals. Potential anthropogenic inputs of heavy metals include haphazard

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industrial emissions, improper agricultural practices like frequent fertilizer and pesticide applications, waste disposal, waste incineration, urban effluents, and vehicular exhausts (Singh *et al.* 2014).

Heavy metals usually find their way to agricultural soils through various irrigation practices (Chopra *et al.* 2007), and the bio-available fraction of elements in soil gets transferred to food and fodder crops, introducing risk to human and animal health. Transfer of heavy metals from agricultural soil to plant is the major pathway of human exposure to those metals, as soil is an important resource in food production. Factors such as local climate, atmospheric dry depositions, physicochemical properties of soil including pH, cation exchange capacity (CEC), soil texture, organic matter content, degree of maturity of plants at the time of harvest and metal species exhibit noticeable effect on bio-available fraction of heavy metals from soil to crops (Chen *et al.* 2016). Non-biodegradable, bio-accumulative and persistent nature of heavy metals make them extremely toxic even at trace concentrations because of their solubility in water and lack of proper mechanism in the body for their elimination (Alloway 2009). Dietary intake accounts for about 90% human exposure to environmental contaminants including heavy metals, as compared to inhalation and dermal routes of exposure (Fries 1995). Heavy metals intake in food diet may cause carcinogenic and non-carcinogenic health problems (Duruibe *et al.* 2007). Therefore, regarding food safety issues, maximum permitted levels of toxic heavy metals in foodstuffs are strictly defined by many regulatory agencies, e.g. the World Health Organization (WHO) and the Food and Agriculture Organization (FAO).

Rice is a staple food for nearly half of the world's seven billion people and more than 90% of this is consumed in Asia. About 55% of the total rice production is accounted by China, Taiwan, and India alone. Extensive studies have been undertaken in yester years by different researchers on quantification metals in rice (Table 1). Singh *et al.* (2014) investigated the uptake of heavy metals by rice grains and associated health risk due to heavy metals exposure via their ingestion to the people living around Narora Atomic Power Station, Narora, India. Results indicated the absence of non-carcinogenic health risks as non-carcinogenic hazard index (NHI) was less than unity. In another study in the same state conducted in the paddy fields of Ramgarh Lake, Gorakhpur, by Singh *et al.* (2011), heavy metals concentrations in

Table 1. Metal concentrations (mg kg^{-1}) in rice grains reported from different parts of the world.

S. No.	Location	Cu	Pb	Co	Ni	Cd	Cr	Fe	Zn	Reference
1	Narora, India	1.12	0.24	0.09	—	0.14	0.03	63.24	—	Singh <i>et al.</i> 2014
2	Guqiao coal mine, China	8.0	0.38	—	—	—	—	—	—	Wang <i>et al.</i> 2013
3	Kota Maruda, Malaysia	0.31	ND	—	—	0.18	1.34	2.89	—	Yap <i>et al.</i> 2009
4	Pearl river estuary, China	8.31	0.28	—	2.34	0.46	2.40	—	20.9	Li <i>et al.</i> 2012
5	West Bengal, India	2.1–4.6	—	—	0.13–1.10	—	—	—	3.21–10.3	Roychowdhury <i>et al.</i> 2003
6	Yangtze delta, China	0.87–7.8	—	—	—	0.01–0.66	0.01–0.74	—	9.10–28.3	Hang <i>et al.</i> 2009
7	Australia	2.90	—	0.02	—	0.01	0.14	—	17.1	Rahman <i>et al.</i> 2014
8	Bangladesh	1.60	0.02	0.004	—	0.07	0.12	—	13.4	Rahman <i>et al.</i> 2014
9	China	3.30	0.35	0.17	0.48	0.03	0.19	—	—	Fu 2008
10	Yangtse Delta, China	1.3–1.8	—	—	—	0.002–0.71	—	—	—	He <i>et al.</i> 2013
11	GNPP site, India	1.9	0.62	0.68	0.40	0.05	0.57	17.2	11.6	Present study

rice grain were reported within the prescribed limits imposed by the Indian Prevention of Food Adulteration Act. Higher accumulation of metals (Zn, Mn, Cu, Fe, Pb, Cd and Sr) than their respective permissible residue limits in paddy straw from Panjab province, Pakistan, was reported (Abbas *et al.* 2007). In another study from Pakistan, different cultivars of rice from different sites in district Sheikhpura were assessed for the irrigation impact of heavy metal contaminated water (Nawaz *et al.* 2006). Irrespective of the site or the variety, accumulation of Cu in rice grains was significantly lower than that in the straw.

In the study area, rice is the second staple food after wheat. Therefore, in the present study the nutrient sufficiency of rice and non-carcinogenic health risk associated with the intake of any excess heavy metals through rice grain ingestion, grown under natural field conditions were determined. Health risk indices, viz. non-carcinogenic hazard quotient (NHQ) for individual metals and NHI for overall potential of all the metals together, were determined.

Methods and materials

Study area

The study area is an alluvial plain of the Indo-Gangetic basin with sandy, sandy loam and clay soil types located in Fatehabad district, Haryana, India. Climate of the area is semiarid with average rainfall of 354.5 mm, cultivated mainly for wheat, cotton and rice.

Rice sampling and analysis

Rice grain samples (*Oryza sativa*) with hulls were collected directly from paddy fields during its harvesting season (November–December) for 2 years, 2011 and 2012. The GPS coordinates of the rice grain sampling locations are given in Table 2. Composite rice grain samples of about 1 kg each were taken directly from the fields, stored in polyethylene bags and brought to the laboratory for further processing and analysis. The collected samples were de-hulled by mechanical means and then dried in a drying-oven at 90°C until a constant weight was achieved. The dried samples were then powdered, homogenized and ashed in a muffle furnace at 350–400°C till the ash was completely white. Each rice grain ash sample of 0.2 g was mixed with 5.0 ml diacid (HNO₃ and HClO₄) (Fisher scientific and Qualigens fine chemicals, Mumbai, India) mixture in the ratio of 9:1 (v/v). The sample mixture was then taken in pre-cleaned teflon vessels, left open overnight at room temperature and digested in a microwave digester (CEM MarsX, USA).

Table 2. Rice grain sampling locations with GPS coordinates for years 2011 and 2012.

Sampling location	Sample ID	Rice-2011*		Sample ID	Rice-2012*	
		Latitude	Longitude		Latitude	Longitude
Fatehabad	FTBR1	29°30'26.2" N	75°28'53.1" E	FTBR3	29°30'25.6" N	75°29'00.7" E
Fatehabad	FTBR2	29°30'11.9" N	75°29'26.5" E	FTBR4	29°30'15.1" N	75°29'09.1" E
Dhani Majra	DM R1	29°30'43.1" N	75°31'37.8" E	DM R3	29°30'39.5" N	75°31'43.1" E
Dhani Majra	DM R2	29°30'54.4" N	75°32'17.4" E	DM R4	29°30'47.2" N	75°31'33.2" E
Kajalheri	KAJLR1	29°26'49.0" N	75°36'19.8" E	KAJLR3	29°26'32.4" N	75°36'12.2" E
Kajalheri	KAJLR2	29°26'45.3" N	75°37'02.8" E	KAJLR4	29°26'43.4" N	75°36'31.4" E
Badopal	BDPLR1	29°24'49.2" N	75°33'21.6" E	BDPLR3	29°25'14.7" N	75°33'9.22" E
Badopal	BDPLR2	29°25'12.3" N	75°33'09.9" E	BDPLR4	29°25'06.5" N	75°33'27.9" E

*Composite soil samples from the same agricultural field were also collected.

When digestion was complete, the digests were evaporated to dryness on a hot plate and the residue was dissolved in double distilled water to make the desired volume. The extracted solutions were then transferred to polypropylene bottles and refrigerated until analysis.

Soil sampling and analysis

In order to evaluate the bio-concentration factor (BCF) of metals from soil to rice grains, the corresponding paddy fields' sub-surface soil samples were also collected from the rooting zone. From a depth of 5–10 cm, four sub-samples of soil from a rectangular grid of 0.5 m² area were collected and then mixed together to obtain a representative sample. The soil samples were collected using a non-metallic spade so as to avoid metal contamination. Samples were sealed in clean plastic bags after removing foreign bodies. Soil samples were oven-dried, ground and then passed through a 2.0-mm sieve. The ground soil samples (0.5 g) were extracted following the same procedure adopted for the grain samples.

All working standards (CDH, New Delhi, India) used for analysis were prepared by diluting 1000 mg/L certified standard solutions. Acetylene gas was used as fuel and air as support in Flame atomic absorption spectrophotometer (FAAS). An oxidizing flame was used in all cases except chromium, where nitrous oxide flame was used for reducing metal quantification. The extracts were analyzed for eight metals, viz. Fe, Cu, Cd, Ni, Zn, Cr, Co and Pb using Flame Atomic Absorption Spectrophotometer (GBC-SensAA, Australia) and concentrations were finally expressed in mg kg⁻¹ on dry wt. basis.

During processing and analysis of samples, quality assurance and control measures were adopted to ensure the reliability of results. To avoid sample contamination, high purity deionized water was used for rinsing glassware, preparation of standards and dilution of samples. Reagent blanks were analyzed and the data were subsequently blank corrected to remove analytical bias. Relative standard deviations of replicate measurements were <10% metals. The calibration curves were linear within the concentration range, with the regression coefficients (R^2) > 0.999. To ensure the reliability of results, standards of respective metals were run after every 10 samples analyzed.

Soil physicochemical analysis

Rice is a critical staple food in Asian countries produced under flooded conditions; thus, the uptake of metals by rice is affected by soil conditions. Soil pH and electrical conductivity (EC) were determined in an aqueous suspension of 10 g dry soil and 100 mL distilled water, stirred and allowed to stand for 30 min by pH (Cyberscan pH tutor, Eutech Instruments, Singapore) and EC (EC meter: CM183, Elico, India) meters. The total organic carbon of the soils was analyzed by the Walkley and Black (1984) method, CEC by HESSE (1971) method and available Na, K contents by ion exchange method using Flame photometer (Flame photometer: CL378, Elico, India). Calcium contents of the soil samples were analyzed by EDTA titration method.

Bio-concentration factor of metals from soil to rice grains

The BCF was determined to evaluate the metals' uptake efficiency of rice grains as the ratio of concentration of the metal in grain to their concentration in respective rhizospheric zone

soils using Eq. (1) (Singh *et al.* 2014):

$$\text{BCF}_{\text{Grain}} = \frac{C_{\text{Grain}}}{C_{\text{Soil}}} \dots\dots\dots (1)$$

where C_{Grain} and C_{soil} are the concentration of metal in rice grains and respective root zone soils on dry weight (DW) basis, respectively.

Health risk due to ingestion of metals via rice grains

Exposure to individual metal termed as chronic daily intake (CDI) is expressed as the mass of the metal per unit body weight per unit time, averaged over lifetime (U.S. Environmental Protection Agency (USEPA) 2010)

$$\text{CDI}_{\text{ingestion}} = \frac{[C_M \times F_{\text{IR}} \times E_F \times E_D]}{[W_{AB} \times T_A]} \dots\dots\dots (2)$$

where C_M is the concentration of a metal in rice grain (mg kg^{-1} DW); F_{IR} is the food ingestion rate ($0.178 \text{ kg day}^{-1}$) of rice for an adult person living in the study area (IRRI 2013); E_F is the exposure frequency, i.e. how often a person is exposed to the contaminated medium over the course of a typical year ($365 \text{ days year}^{-1}$); E_D is the exposure duration, i.e. how long a person is exposed to the contaminated medium over the course of their lifetime (70 years for adults); W_{AB} is the average body weight of the exposed person (60 kg for Indian adults) (NNMB 2002) and T_A is the average exposure time for non-carcinogenic effects which specifies the length of time over which the average dose is calculated ($ED \times 365 \text{ days year}^{-1}$).

Non-carcinogenic health risk associated with individual metals exposure through ingestion of rice grain was calculated as a quotient between the estimated exposure (CDI) and reference dose oral (RfDo) metal. If $\sum \text{NHQ}$ exceeds unity, potential non-cancer effects may be a concern (USEPA 2010)

$$\text{NHQ} = \frac{\text{CDI}}{\text{RfDo}} \dots\dots\dots (3)$$

Results and discussion

Metal concentration in rice field soil samples

Metals may become part of the soil in due course of pedogenesis, and their distribution in the soil-plant system is governed by physical processes including capillary action, runoff, leaching and root uptake (Singh *et al.* 2014). The results of metals in paddy fields' soil (mg kg^{-1}) are given in Table 3. It was observed that Zn and Fe contents in the soil were higher as compared to other metals. Much lower concentrations of Cd (0.87 mg kg^{-1}), Pb (19.6 mg kg^{-1}) and Zn (65.1 mg kg^{-1}) were reported in paddy fields' soils in China (Feng *et al.* 2012). Lower Cu, Cd, Pb and Zn (20.70 , 0.15 , 10.0 , and 31.40 mg kg^{-1}) content than the present study was reported from Chinese paddy fields' soils around the Guqiao coal mine area (Wang *et al.* 2013). Similarly Yap *et al.* (2009) also reported Malaysian paddy fields' soils having lower concentrations of Cd, Zn and Cr (0.78 , 21.1 , and 2.08 mg kg^{-1}). In a study

Table 3. Metals concentrations in rice grains, corresponding fields' soil and their BCF.

Matrices	Statistics	Fe	Cu	Cd	Pb	Ni	Zn	Co	Cr
Rice grain (mg kg ⁻¹)	Min	5.1	0.91	<0.01	<0.06	<0.04	5.5	<0.05	<0.05
	Max	58.8	5.7	0.09	0.99	0.41	20.2	0.83	0.87
	Mean	17.2	1.9	0.05	0.62	0.40	11.6	0.68	0.57
	GM	13.8	1.6	0.05	0.60	0.39	10.9	0.7	0.5
	SD	13.7	1.3	0.02	0.17	0.02	4.1	0.11	0.18
Rice fields' soil (mg kg ⁻¹)	Min	3017	10.8	1.0	7.5	<0.04	71.5	5.1	<0.05
	Max	5687	37.0	7.7	148.2	23.5	146.7	12.9	9.7
	Mean	4770	21.8	5.1	54.4	10.4	106.0	7.0	6.5
	GM	4723	20.7	4.8	36.9	7.8	104.2	6.7	6.4
	SD	651	6.9	1.7	44.1	7.1	20.2	2.1	1.4
BCF rice grain	Min	0.001	0.04	NC	NC	NC	0.04	NC	NC
	Max	0.012	0.37	0.02	0.07	0.03	0.19	0.16	0.09
	Mean	0.004	0.10	0.01	0.02	0.02	0.11	0.11	0.08
	GM	0.003	0.08	0.009	0.011	0.02	0.11	0.10	0.08
	SD	0.001	0.03	0.004	0.02	0.01	0.04	0.03	0.02

NC, not countable due to one or the other matrix value being BDL.

conducted by Fu (2008) on metals content in soil–rice system at Zhejiang, China, much higher content of Cu (98.81 mg kg⁻¹), Pb (55.81 mg kg⁻¹), Cr (61.21 mg kg⁻¹) and several-fold lower content of Cd (1.19 mg kg⁻¹) in soil was reported.

Results showed that metals concentration in the soil samples collected from the study area varied considerably. Differences in soil metal contamination may be due to the use of different agricultural applications such as addition of manures, fertilizers and pesticides, and different irrigation practices. Higher iron concentration in soils may be due to its relative abundance in the Earth crust and that of Zn might be due to the use of zinc fertilizers in the fields to enhance crop productivity. Further, movement of metals from the soil solution depends upon its physico-chemical characteristics. Hence paddy fields' soil quality parameters were also analyzed. pH of the paddy fields' soil varied from slightly neutral (6.9) to alkaline (8.9); EC (77.7–1021.5 μ s/cm), available Na (126–1754 mg kg⁻¹), available K (114–812 mg kg⁻¹), total Ca (0.9–6.4 g kg⁻¹), CEC (14.5–109.8 meq/100 g of soil) and TOC (1.1–3.2%).

Metal concentration in rice grain samples

Results showed the concentration spectra of various metals such as Fe, Cu, Cd, Ni, Zn, Cr, Co and Pb in rice grains gathered from Haryana, India (Table 3). Concentration of Zn in rice grains collected from study area varied from 5.50 to 20.24 mg kg⁻¹. Zinc is required to maintain the functioning of the immune system but dietary intake above a safe limit of 12–15 mg day⁻¹ may cause vomiting, renal damage, sideroblastic anemia and cramps (ATSDR 1994a). Copper concentrations in rice samples varied from 0.91 to 5.70 mg kg⁻¹ and were well within the maximum permissible limit of 10 mg kg⁻¹ given by WHO (1998). Rice grain Fe content was reported in the range of 5.10 to 58.80 mg kg⁻¹. Iron can be found in meat, whole meal products and vegetables. Iron may cause conjunctivitis, choroiditis, and retinitis if it contacts and remains in the tissues. Chronic inhalation of excessive concentrations of iron oxide fumes or dusts may result in development of a benign pneumoconiosis. Inhalation of excessive concentrations of iron oxide may enhance the risk of lung cancer development in workers exposed to pulmonary carcinogens. A more common problem for humans

is iron deficiency, which leads to anemia. A man needs an average daily intake of 7 mg of iron and a woman 11 mg.

In the present study, rice grains' Ni concentrations were below the permissible limit of 0.5 mg kg^{-1} for food crops (FAO/WHO 1984) and ranged from <0.04 to 0.41 mg kg^{-1} . Deficiency of nickel has been linked with hyperglycemias, depression, sinus congestion, fatigue, reproductive failure and growth problems in humans, while hyperintake of nickel above a permissible safe limit of $3.0\text{--}7.0 \text{ mg day}^{-1}$ may lead to hypoglycemia, asthma, nausea, and headache (ATSDR 1999). Deficiency of Co in diet results in pernicious anemia, severe fatigue, shortness of breath and hypothyroidism, while overdose may lead to angina, asthma, cardiomyopathy, polycythemia and dermatitis. The safety limit for human consumption of Co is $0.05\text{--}1.0 \text{ mg day}^{-1}$ in humans (ATSDR 1994b). Thus, the recorded range of $<0.05\text{--}0.83 \text{ mg kg}^{-1}$ for cobalt concentration in rice grains falls within the safety limit in the present study. Cadmium concentrations in rice grain samples varied from <0.01 to 0.09 mg kg^{-1} and were well within the maximum permitted level of 0.4 mg kg^{-1} in rice ((FAO/WHO 2014). Gastrointestinal, hematological, musculoskeletal, renal, neurological and reproductive effects may result from low-level chronic exposure to Cd and kidney being the main target organ (ATSDR 1999). Intake of Cd can double if one smokes cigarette because each cigarette contains about 2 mg of Cd. Recorded Pb and Cr concentrations in rice grains in the present study ranged from <0.06 to 0.99 and <0.05 to 0.87 mg kg^{-1} , respectively. Dietary intake of Pb may cause neurological, musculoskeletal and developmental problems.

In the present study, geometric mean concentrations (mg kg^{-1}) of the studied metals occupied the following decreasing order in rice grains: Fe (13.8) > Zn (10.9) > Cu (1.6) > Co (0.70) > Pb (0.60) > Cr (0.50) > Ni (0.39) > Cd (0.05). Zn, Fe and Cu are essential elements required for various enzyme activities; they play important roles in photosynthesis and growth of a plant (Tripathi *et al.* 1997). Their abundance was comparatively higher than the non-essential metals in rice grains. Copper, Ni, Co, Cd content in rice grain samples were well within the respective maximum permissible limit of 10 mg kg^{-1} (WHO 1998), 0.5 mg kg^{-1} (FAO/WHO 1984), 1.0 mg kg^{-1} (ATSDR 1994a) and 0.4 mg kg^{-1} (FAO/WHO 2014). Comparable Cu contents in rice grains ranging between 0.8 and 5.8 mg kg^{-1} were reported by Smuc *et al.* (2012) from the Kocani field plant system. Much higher concentrations of Cd, Zn and Ni (0.44, 33.1 and 20.3 mg kg^{-1}) in rice grains were reported by Zheng *et al.* (2013) in South China. Several-fold higher concentration of Pb (61.17 and 55.81 mg kg^{-1}) than the present study was reported by Orisakwe *et al.* (2012) and Fu (2008) in rice grains from Nigerian and Chinese paddy fields, respectively. The rice grain Fe content in the present study was higher than that reported by Abbas *et al.* (2007) but lower than that reported by Antoine *et al.* (2012). Li *et al.* (2012) reported comparatively higher Cd, Cr, Zn, and Cu ($0.46, 2.4, 20.9$, and 8.3 mg kg^{-1}) and lower Pb and Ni (0.28 and 2.3 mg kg^{-1}) content in rice grains collected from China. Rice grains collected from Jiangsu province, China, were reported having higher Cr, Cu and Zn ($0.75, 2.6$, and 12.0 mg kg^{-1}) content by Cao *et al.* (2010). Nickel concentration in rice grains was lower than some studies reported globally (Fu 2008; Li *et al.* 2012; Roychowdhury *et al.* 2003) but higher than Cuadrado *et al.* (2000). Hang *et al.* (2009) have reported higher Cu, Pb, Zn and Cd ($0.87\text{--}7.77, 0.26\text{--}2.60, 9.10\text{--}28.3$, and $0.012\text{--}0.66 \text{ mg kg}^{-1}$) but lower Cr ($0.01\text{--}0.74 \text{ mg kg}^{-1}$) content in rice grains grown in the Yangtze River Delta region, China.

Bio-concentration factor of metals from soil to rice grains

Soil-to-plant transfer of metals is the most important pathway of human exposure to them through the food chain. Bioavailability of metals from corresponding rooted soils to plants can easily be assessed by their BCF values, as higher the BCF more available the metal is and vary greatly with the metal and plant species (Cui *et al.* 2004). BCF of metals in soil to rice grains is reported in Table 3 and Figure 1. The mean BCF of metals in rice grains was in the following order: Zn (0.11) > Co (0.10) > Cu (0.08) = Cr (0.08) > Ni (0.02) > Pb (0.011) > Cd (0.009) > Fe (0.003).

Singh *et al.* (2014) reported a BCF of Cu (0.07) from soil to rice grains in highly fertile Gangetic plains in India. Consistent to the results of the present study, stronger capacity to bio-accumulate micronutrient Zn as compared to toxic metals was presented by rice grown in south Indian paddy fields (Satpathy *et al.* 2014). Higher mean BCF values of 0.37 for Zn and 0.15 for Cd in rice grains were reported by Smuc *et al.* (2012) in Kocani Field plant system, Republic of Macedonia. Similarly, Cao *et al.* (2010) reported higher BCF of Zn from soil to rice grains followed by Cu > Cd > Hg > Cr > Pb. Ali and Al-Qahtani (2012) reported higher BCF of Cu and Zn in rice grains collected from Egyptian open market. Comparatively, higher BCFs of Cu, Pb, Cd and Zn (0.25, 0.05, 0.44 and 0.58) were estimated in rice grains grown around Guqiao coal mine area, China (Wang *et al.* 2013). Kibria *et al.* (2012) compared to the present study also reported higher BCF of Pb and Cd (0.21 and 0.36) but lower of Cu, Fe and Zn (0.05, 0.02 and 0.008) in rice grains grown in Bangladesh.

Further, to identify any relationships between the BCF of metals in rice grains and corresponding fields' soil quality parameters, Pearson's correlation analysis was performed; the results are listed in Table 4. But no statistically significant correlations among the BCF of the studied metals in rice grains and the corresponding fields' soil physico-chemical properties were observed. However, correlation analysis revealed associations among different soil quality parameters in rice fields (Table 4). Significant positive correlations were found between EC and Na ($r = 0.86^{**}$;

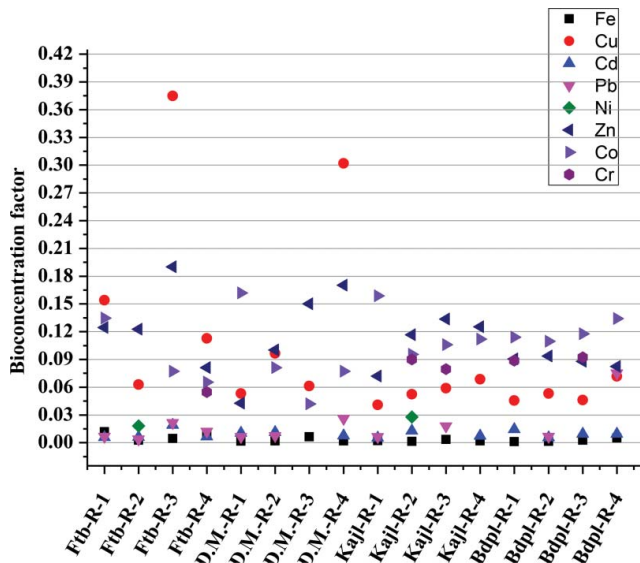


Figure 1. Bio-concentration factor of metals from soil to rice grains cultivated in the study area.

Table 4. Correlation among the bio-concentration factor of metals in rice grain and corresponding fields' rhizospheric soils.

	BCF-Fe	BCF-Cu	BCF-Cd	BCF-Pb	BCF-Zn	BCF-Co	BCF-Cr	pH	EC	Na	K	Ca	TOC	CEC
BCF-Fe	1													
BCF-Cu	0.199	1												
BCF-Cd	-0.238	0.419	1											
BCF-Pb	0.06	0.123	0.223	1										
BCF-Zn	0.115	0.722*	0.346	0.059	1									
BCF-Co	-0.199	-0.373	-0.303	0.016	-0.637*	1								
BCF-Cr	-0.97**	-0.98**	0.745	1.00*	0.281	0.899*	1							
pH	0.021	-0.07	-0.014	0.046	0.031	0.151	0.261	1						
EC	0.24	-0.211	-0.282	0.454	-0.378	-0.054	-0.862	-0.379	1					
Na	0.113	-0.263	-0.23	0.705*	-0.341	0.07	-0.608	-0.125	0.863**	1				
K	-0.005	-0.419	-0.071	0.455	-0.425	0.34	0.718	0.176	0.211	0.482	1			
Ca	-0.221	0.014	0.086	0.02	0.183	-0.233	0.372	0.502*	-0.236	-0.2	-0.132	1		
TOC	0.081	0.172	0.034	0.147	-0.029	-0.183	-0.628	-0.93**	0.484	0.229	-0.14	-0.538*	1	
CEC	-0.005	-0.229	0.061	0.552	-0.258	-0.073	-0.568	-0.183	0.790**	0.846**	0.247	-0.186	0.243	1

*Correlation is significant at the 0.05 level (two-tailed).

**Correlation is significant at the 0.01 level (two-tailed).

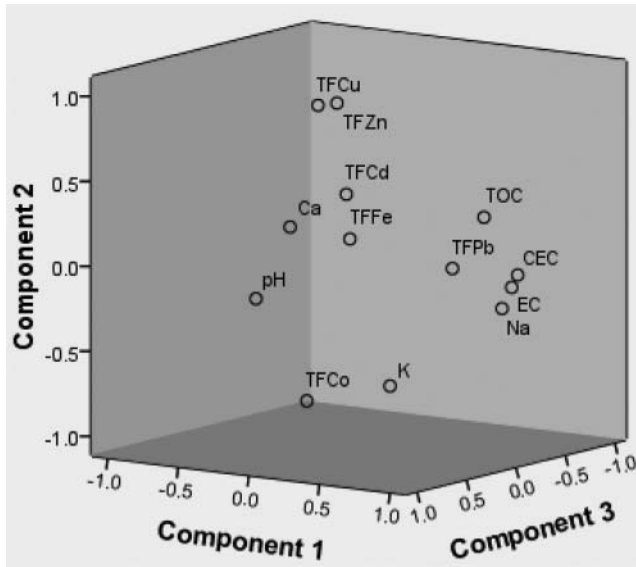


Figure 2. Principal component plot in a rotated space (PCA) for metals bio-concentration factor in rice grain and soil physicochemical properties.

$p < 0.01$), EC and CEC ($r = 0.79^{**}$; $p < 0.01$), CEC and Na ($r = 0.84^{**}$; $p < 0.01$), pH and Ca ($r = 0.50^{*}$; $p < 0.05$) content of soil. Total organic carbon was found negatively correlated with pH ($r = -0.92^{**}$; $p < 0.01$) and Ca ($r = -0.53^{**}$; $p < 0.01$) content of soil. Decrease in pH of the soil might be due to the formation of organic acids, thus lowering the pH of the soil solution with increasing organic carbon contents.

BCF_{Cr} has been reported to be perfectly correlated with BCF_{Pb} ($r = 1.00^{**}$; $p < 0.01$). In another study conducted on soil–rice system at Narora India, similar observations were made for the BCF of two metals in rice grains with correlation coefficient value of 0.62. In some yester studies rice has been acknowledged as one of the major sources of Cd and Pb for human beings in Asian continent (Fangmin *et al.* 2006; Zhang and Ke 2004). Probable sources of these toxic metals in paddy crops may be the indiscriminate application of agrochemicals in the Asian fields.

Principal component analysis (PCA), a multivariate statistical technique allows simplified graphical representations, observation and interpretation of information. Principal component analysis with Varimax normalization (PCA-V) was applied for the qualitative evaluation of correlation matrix results. Three factors loaded with BCF of different metals and soil quality parameters, explaining a total variance of 70%, were obtained (Figure 2). Factor-1 contributed 26.3% to

Table 5. Chronic daily intake and non-carcinogenic hazard quotient (NHQ) of the studied metals to the target population via ingestion of rice grains.

	Statistics	Fe	Cu	Cd	Pb	Ni	Zn	Co	Cr
CDI (mg kg ⁻¹ day ⁻¹)	Min	1.51E-02	2.70E-03	<2.97E-05	<1.78E-04	<1.00E-04	1.63E-02	<1.00E-04	<1.00E-04
	Max	1.74E-01	1.69E-02	2.52E-04	2.94E-03	1.22E-03	6.00E-02	2.46E-03	2.58E-03
	Mean	4.09E-02	4.89E-03	1.34E-04	1.78E-03	1.17E-03	3.25E-02	1.99E-03	1.63E-03
NHQ	Min	2.16E-02	6.75E-02	<3.00E-02	<4.50E-02	<6.00E-03	5.44E-02	NC	<6.60E-05
	Max	2.49E-01	4.23E-01	2.52E-01	7.34E-01	6.08E-02	2.00E-01	NC	1.72E-03
	Mean	5.84E-02	1.22E-01	1.34E-01	4.45E-01	5.85E-02	1.08E-01	NC	1.09E-03

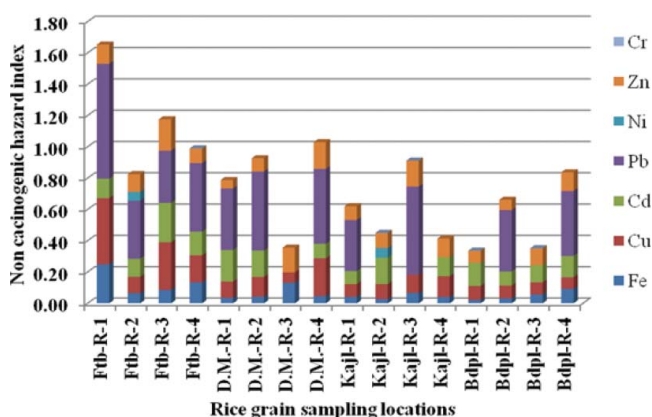


Figure 3. Non-carcinogenic hazard index (NHI) of metals through rice grains ingestion for the adult population residing in the study area.

the total variance with a high loading on Na ($r = 0.961$), EC ($r = 0.851$), CEC ($r = 0.885$), thus explaining associations observed among these soil quality parameters. Factor-2 (23.4%) with a high loading on BCF_{Cu} ($r = 0.904$) and BCF_{Zn} ($r = 0.898$) supported a correlation between BCF of these metals. Factor-3 with a variance of 20.2% explained lowering of pH ($r = 0.947$) with increasing organic carbon contents ($r = -0.952$) of the soil.

Health risk due to dietary intake of metals through rice grain ingestion

Metals usually find their way to agricultural soils through various irrigation practices (Chopra *et al.* 2009), and from their soil bio-available fraction gets transferred to food crops, introducing risk to human health. Heavy metals intake in food diet may cause carcinogenic and non-carcinogenic health problems (Duruibe *et al.* 2007). Major non-carcinogenic health problems caused by metals include hepatic, renal, neurological, developmental, gastrointestinal, respiratory, cardiovascular, hematological, reproductive and immunological disorders, etc. In the present study non-carcinogenic health risk assessment indices viz. CDI and NHQ were calculated using standard equations given by USEPA (encapsulated in Table 5).

Chronic daily intake of metals via rice grains ingestion

Tracing the exposure level of contaminants and its possible routes to the target organisms are of great concern in health risk assessment studies. By consuming rice grown at the studied sampling locations, an adult will have higher average CDI of Co, Cr and Pb than the threshold CDI values given by WHO/ EC (Table 5). These values were several-fold lower than the CDIs of Cu, Cr, Ni, Pb, and Cd (33.99, 6.74, 13.33, 1.50, and 0.18 $\text{mg kg}^{-1} \text{day}^{-1}$), respectively, reported by Liu *et al.* (2011) for adults through rice grain ingestion grown in electroplating wastewater contaminated fields in Guangxi, China. Oral intake of Zn, Pb, Cd and Cu through ingestion of rice grains grown in Yangtze River Delta, China, was 0.09, 0.002, 0.0003, and 0.02 $\text{mg kg}^{-1} \text{day}^{-1}$, respectively (Hang *et al.* 2009). Zhuang *et al.* (2009) observed CDIs of Cd and Pb through rice ingestion exceeding the FAO/WHO (1984) permissible limits. Huang *et al.* (2013) in their study on rice from Zhejiang, China, reported CDI ($\text{mg kg}^{-1} \text{day}^{-1}$) of 0.49 of As, 0.20 of Cd, 0.03 of Hg and 0.37 of Pb through rice grain ingestion.

Non-carcinogenic hazard quotient of metals via rice grains ingestion

The NHQ of individual metal to the target population via ingestion of rice grains is enlisted in Table 5. The NHQ value was less than unity for all the studied metals which indicates that the daily intake of these metals through rice grains ingestion is unlikely to cause adverse health effects to target population.

For the target population ingesting the rice grains produced at most of the sampling locations except *Fathebad* site-1 and *Fathebad* site-3, there was no cause of concern relating to non-carcinogenic risks. Ingestion of rice grown at the rest of the studied sampling locations was safe as NHQ for individual metal and NHI for $\sum_{\text{all studied metals}}$ was <1.0 (Figure 3). The NHI indicated that poor non-carcinogenic risks were associated with the ingestion of rice grains grown in *Fathebad* site-1 (NHI = 1.65) and also, but negligible, from the ingestion of that grown in *Badopal* site-1 (NHI = 0.34). Rice grown in the vicinity of Narora Atomic Power Station, India, was reported safe for consumption by the local population as NHQ for all studied metals was less than 1.0 (Singh *et al.* 2014). Because rice is the main staple food in south India, adult populace of Kalpakkam might experience some adverse health effects if they consume locally grown rice, as the reported HRI values exceed the safe standards of unity (Satpathy *et al.* 2014). Various authors have reported comparatively higher NHI values via dietary intake of locally grown rice grains by the Chinese populace in different provinces of China, but still it is allowed for consumption (Hang *et al.* 2009; Liu *et al.* 2011; Qian *et al.* 2010; Zhuang *et al.* 2009).

Conclusion

Rice being the second staple food grain in the study area makes it liable to be examined for nutrients and contaminants like metals. The grain collected from the study sites had most of the studied metals within the threshold levels permitted by various regulatory agencies. It can be concluded from the study that essential elements, i.e. Zn, Co and Cu, had higher bio-concentration in rice grains as compared to other toxic metals. No statistically significant dependence relating to metals BCFs in rice grains with that of paddy fields' soil properties was observed. However, there existed significant correlations among different soil quality parameters, which was further supported by factor analysis. Health risk indices revealed ingestion of rice grown at most of the studied locations safe, as NHI was less than unity and there was no cause of concern for non-carcinogenic health risks. However, long-term assessment studies are recommended as considerable seasonal variations in metal uptake by crops may be observed under natural field conditions.

Funding

The work was financially supported by the Board of Research in Nuclear Sciences, Mumbai. The authors gratefully acknowledge and are thankful to the funding agency.

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