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Research article

Impacts of long-term irrigation with coalmine effluent contaminated water on trace metal contamination of topsoil and potato tubers in Dinajpur area, Bangladesh

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ABSTRACT

Rapid depletion of groundwater and climate change mediated shifting precipitation pattern is forcing farmers to look for alternative irrigation options like wastewater. However, routine irrigation with trace metal contaminated wastewaters could potentially pollute soil as well as cause health risks through the consumption of food products grown in contaminated soil. Thus, the present study aimed to investigate the trace metals build-up status in topsoil and potato (*Solanum tuberosum* L.) tubers upon continuous irrigation with coalmine effluent contaminated wastewater compared to irrigation with groundwater and surface water over three consecutive years. Soil pollution status and human health risk associated with consumption of potato tubers grown on wastewater-irrigated soil was also assessed in this study. Three separate experimental sites differing in irrigation source (groundwater, surface water, and coalmine wastewater) were selected near Barapukuria Coal Mining Company Limited located at Parbatipur upazilla of Dinajpur district, Bangladesh. Nine trace metals namely arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb), and zinc (Zn) were estimated. Results showed significantly higher trace metal content in both soil and potato tubers due to wastewater irrigation. Wastewater suitability for irrigation regarding Cd, Cr, Cu, Fe, Ni and Pb were off the permissible level although the soil contamination with trace metals and their levels in potato tubers remained within the safety limit. Health risk assessment revealed that, consumption of potato tubers grown in wastewater-irrigated soil remained safe although health risk associated with Cr was almost at the border. The study exclusively highlighted the core massage that, trace metal contamination of both soil and potatoes cultivated in them was increasing alarmingly due to three years of wastewater-irrigation. Although the extent of contamination was below critical limit, it can potentially become hazardous in years to come unless wastewater-irrigation is checked. This study was successful to provide valuable insights regarding the potential environmental and human health threats that might arise due to unmindful irrigation of contaminated coalmine wastewater. Besides, this study should prove useful in strategizing safety measures for

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cropping under trace metal contaminated soils and for planning industrial effluent disposal to avoid agricultural soil contamination.

1. Introduction

The issue of food security and safety has become a burning matter in recent eras. Progressive public awareness towards a healthier lifestyle, coupled with the growing population is continuously creating massive pressure on the food production demand both in terms of quantity and quality [1,2]. Currently, such demands are being met via resource intensive and mechanized cultivation strategies that increase cropping area as well as per unit production [3–5]. However, these practices pose a significant risk of exploitation of natural resources, particularly groundwater. Lowered down groundwater table and frequent occurrence of acute droughts are one of the pieces of evidence [6–8]. Predictions indicate severe water scarcity by 2050 for more than two-thirds of the world's inhabitants [9–11]. Climate change impacts are also worsening the conditions [12]. Under such circumstances, increasing dependency on surface water resources for crop irrigation has been employed as a sustainable strategy in numerous countries worldwide for groundwater conservation and better water resource use efficiency [5,13–15]. However, using polluted surface water, also known as wastewater for irrigation purposes could potentially induce numerous risks for both human and environmental health [16,17]. Surface water polluted via direct and indirect effluent disposal originating from industrial sectors are termed as industrial wastewater that can contain large quantities of various trace metals like As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn [18,19]. Use of such wastewater for irrigation is only permittable till their quality parameters are maintained within the defined criteria by the Food and Agriculture Organization of the United Nations (FAO). However, in majority of the developing countries, knowingly or unknowingly wastewater irrigation is a very common practice that might pose potent ecological and health risks due to weaker policy and regulatory frameworks [5,20–22]. Long-term irrigation with contaminated water can cause trace metals accumulation in the topsoil at high concentrations as well as groundwater contamination through leaching [23,24]. Wastewater generated from coalmines are comparatively more hazardous as they contain suspended coal powder, stone dust, heavy metals, highly mineralized and/or acidic substances that are potent soil contaminants [24–26]. From the contaminated soils, trace metals find their way into the food products and reach the human body where they can cause health impairments even at low concentrations [27–29]. Pb and Cd, for example, can increase the risk of heart disease, weaken bones, and cause urologic disorders [30]. Excess Cr exposure can cause skin ulcers, dermatitis, bronchial carcinomas, and gastroenteritis [31,32]. Very few of the trace metals like Cu, Fe, Zn, Mn, and Ni are essential for metabolic activities but can become toxic at high concentrations [33]. For instance, excess Cu intake can cause liver damage and excess Mn may cause neural manganism, mitochondrial dysfunction and inflammation [34,35].

Bangladesh is one of the most densely populated developing countries in the world, accommodating around 160 million people within an area of 1,47,570 square kilometers [36,37]. The presence of numerous trace metals in Bangladeshi foodstuffs are quite concerning. Previous reports highlighted this fact and also suggested that the availability of trace metals on food products are in such quantities which are more than enough for creating various health issues [36,38,39]. Topsoil pollution via anthropogenic means as well as human activities such as rapid industrialization, mining activities, land spreading of industrial and municipal waste, wastewater irrigation of crops, application of metal contaminated agrochemicals, and improper post-harvest handling of food products were reported as majorly responsible for metal contamination in topsoil and subsequent accumulation in foodstuffs in Bangladesh [40–45]. Potato (*Solanum tuberosum* L.) is the most consumed vegetable of Bangladesh (64.8 g person^{-1} day^{-1}) and the fourth most important carbohydrate delivering crop of the world [46–48]. Bangladesh remained world's 4th highest potato producing country and it is one of the most promising crops for domestic as well as international food processing industries [49,50]. However, recent studies outlined that potato is very prone to trace metal accumulation as it remains directly in contact with soil [51,52]. Metals and metalloids are naturally available in the soil system, but careless human activities like unplanned effluent management and long-term irrigation with contaminated wastewaters are bound to fill up the trace metal bearing capacity of the soil, after which these trace elements find their way towards contaminating groundwater, surface water and eventually the plants and crops [53,54]. As the most popular vegetable, high trace metal accumulation in potatoes would pose a greater health risk because food chain contamination is one of the major routes for trace metals' entry into the human body [23,33,55]. The massive population of Bangladesh creates a huge food demand resulting in mostly year-round cultivation of available arable lands. She is also a riverine country, and her water bodies provide a significant portion of total irrigation water. Thus, pollution of surface water with trace metals and metalloids could prove highly hazardous for both environment and human health. In recent years, numerous reports relating to food and environment safety issues of wastewater irrigation have been reported mostly focusing on municipal and industrial wastewater [10,21,23,56–59]. However, research advances exploring the potentiality of human health and ecological risks associated with coalmine wastewater irrigation are very scarce, even though coalmines are quite widespread throughout countries of the world including Bangladesh. Coal is one of the biggest natural resources of Bangladesh that majorly supports the countries electrical power plants and cement industries and thus coalmines are an important industry for the country [60]. Barapukuria Coal Mining Company Limited (active since 1985) is one of the biggest coalmine factories of the country located at Northern Bangladesh [61]. Adjacent to the factory there is a canal that provides irrigation water for a large amount of potato cropping lands. Coalmine generated effluents can easily find their way to contaminate surface water both actively and passively. Our hypothesis is that long term irrigation with coalmine effluent contaminated wastewater can result in permanent soil pollution and crop contamination with harmful metals and metalloids that could possibly be extremely risky for human health. Potato is the dominant vegetable crop of the target region and thus the objectives of the current study was (i) to assess and compare different trace metals (As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) contamination level in irrigation water, topsoil and potato tuber harvested from three different irrigation regimes (groundwater, surface water, and coalmine wastewater) and (ii) to assess the associated human health risk upon oral consumption of potato tubers produced through irrigation from these water sources.

2. Materials and methods

2.1. Experimental location

The current study location was in Parbatipur upazilla of Dinajpur district, Bangladesh. The study area falls under a tropical wet and dry climate featuring cold and dry winter followed by warm and humid summer. The annual average temperature is about 25.8 °C with total annual rainfall of approximately 1800 mm. Three different sites of (200 × 200) m^2 each within the selected location were designated as experimental sites which are distinctive from each other based on their irrigation source. Site-I (Ichabpur) received groundwater irrigation (GWI) [control]; site-II (Laharpur) received surface water irrigation (SWI) from the nearby waterbodies; and site-III (Majidpur) received wastewater irrigation (WWI) from the adjacent canal connecting to Barapukuria Coal Mining Company Limited (BCMCL) (Fig. 1) that directly or indirectly pollutes the canal water via effluent discharge (Fig. 1S). Soil of the experimental site is categorized as light-textured, and the dominant crop cultivated during winter is potato (*Solanum tuberosum* L.). Potato cultivar, Cardinal was selected as the plant material because of its highest popularity among farmers of the selected area. Water, soil and potato tuber samples were drawn from each of the designated sites for monitoring trace metal contamination during the time period from 2019 to 2021. Samples were collected between mid-January and early-February each year.

2.2. Determination of soil characteristics

Soil properties of the three experimental sites were analyzed according to the procedures as described by Estefan et al. [62] and have been presented in Table 1S. Briefly, the textural constituents of the soil (sand, silt and clay) were determined using a Hydrometer with Bouyoucos scale in g L^{-1}. Forty grams of oven-dried soil was suspended into an immediately prepared dispersing solution (a solution containing 40 g sodium hexametaphosphate and 10 g sodium carbonate per liter) and kept overnight in room temperature. Then the soil was transferred to a soil-stirring container, filled with water up to three quarters and stirred with a high-speed electric stirrer for 3 min. Afterwards the soil was transferred to a cylindrical hydrometer jar and volumed to 1 L. the suspension was thoroughly mixed, allowed to sattle down, and a hydrometer was inserted to take reading after 40 s (R_{sc}) and 4 h (R_c) respectively for silt and clay determination. Upon taking reading, the soil suspension was passed through a 50 μm sieve and thoroughly washed until the water passing through is cleared. The liquid was collected in a beaker. The sands in the sieve were then oven-dried and weighed in an electrical balance (W_{sand}). The liquid collected in the beaker was kept undisturbed overnight, the water was discarded and the soil the bottom of the beaker was oven-dried and weighted with the sand (W_{soil}). Hydrometer reading for a blank sample (60 mL dispersing solution in 1 L hydrometer jar) were also recorded (R_b). Sand, silt and clay percentage was then extracted from the following formulas:

$$\text{Sand (\%)} = W_{sand} \times (100 / W_{soil})$$

$$\text{Clay (\%)} = (R_c - R_b) \times (100 / W_{soil})$$

Fig. 1. Study area map showing experimental sites. Site-I: Ichabpur; Site-II: Laharpur; Site-III: Majidpur. Satellite image was created by using Google Earth (https://earth.google.com/web/).

Table 1
Significance level in the two-way ANOVA for trace metal concentrations in water, soil and potato tubers at three different sites over three consecutive years.

Trace metals	Water			Topsoil			Potato tuber		
	Site (S)	Year (Y)	S × Y	Site (S)	Year (Y)	S × Y	Site (S)	Year (Y)	S × Y
As	**	**	**	**	**	**	**	**	**
Cd	**	**	**	**	ns	ns	**	**	**
Cr	**	*	ns	**	*	ns	**	**	**
Cu	**	**	**	**	**	**	**	ns	ns
Fe	**	**	**	**	**	**	**	ns	ns
Mn	**	**	*	**	**	ns	**	**	**
Ni	**	**	**	**	**	ns	**	**	**
Pb	**	**	**	**	**	**	**	*	ns
Zn	**	**	ns	**	ns	**	**	**	**

**, *, and ns indicate $P \leq 0.01$, $P \leq 0.05$, and not significant, respectively.

$$[\text{Silt} + \text{Clay}]\ (\%) = (R_{sc} - R_b) \times (100 / W_{soil})$$

$$\text{Silt}\ (\%) = [\text{Silt} + \text{Clay}]\ (\%) - \text{Clay}\ (\%)$$

The soil pH was recorded using a pH meter with combined electrode probe. Fifty grams soil was suspended in a 50 mL de-ionized water, stirred for 3 min using an electric stirrer, and allowed to settle down for 10 min. The procedure was continued three times. The pH meter was then calibrated with standard solution and the electrode was dipped in the soil suspension under stirring for recording the soil pH. For determination of electrical conductivity, 50 g soil was suspended in a 50 mL de-ionized water similarly as done for pH. The suspension was then filtered with Whatman No. 42 filter paper. An electrical conductivity meter was then calibrated and inserted in the filtered solution to record the soil electrical conductivity.

2.3. Sample collection

Four replicates of each sample (water, soil and potato tubers) were collected following standard sampling procedures as described by Estefan et al. [62] with the help of global positioning system (GPS) coordinates using a portable GPS tracker. For collecting soil and potato tuber samples, each of the experimental sites was divided into same number of equal-sized blocks and 16 sub-samples were drawn (single sub-sample per block) for each replicate randomly. Soil was collected up to 15 cm of depth using a manually operated screw auger of 2.5 cm diameter [63]. Tuber samples were taken by manually uprooting the plant and collecting a medium sized healthy tuber. Immediately after sampling, samples were stored in airtight zip-lock bags and transported to the laboratory. Four replicates (16 sub-samples per replicate) of water samples were drawn from the head of the irrigation water outlet while the experimental sites were being irrigated from their respective irrigation source [64]. After collecting the water samples, they were kept in an ice box and transported to the laboratory.

2.4. Sample preparation

Sampled soils were initially air-dried and then oven-dried at 72 °C for a week to obtain a constant weight. Dried samples were then grinded to powder using a mechanical grinder, sieved with a 2 mm mesh, and stored in airtight dry plastic bottles [65]. Potato tubers were initially washed with tap water to remove excess dirt, air dried properly, cut into small pieces, and kept in a drying oven at 72 °C until a constant weight was achieved. Later, the samples were grinded to powder using a mechanical grinder and stored in airtight dry plastic bottles [59]. Water samples were treated with 1 ml concentrated nitric acid (HNO_3) per 100 ml of sample to avoid microbial growth and stored at 4 °C in airtight tubes [57]. In every case, an equal amount of 16 sub-samples per replicate were thoroughly mixed to obtain a composite sample and working samples were drawn from the composite samples for further analysis.

Both the soil and plant samples were digested following the procedures of Sarwar et al. [21]. Powdered working samples (1 g each) were digested using a di-acid mixture [1:2 mixture of perchloric acid ($HClO_4$): nitric acid (HNO_3)] in a hot block digestion chamber. Upon completion of digestion the samples were then allowed to cool down at room temperature and volumed to 100 ml using de-ionized water. Samples were then filtered through a 0.45 μm PTFE (polytetrafluoroethylene) membrane filter and stored at 4 °C in airtight vials. Water samples were also digested in the same manner as soil and plant samples, using only HNO_3 instead of a di-acid mixture [57].

2.5. Trace metal detection

Total nine trace metals (As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) content from the digested samples were determined via an inductively coupled plasma-optical emission spectroscopy (PerkinElmer, USA) following the procedures of Phukunkamkaew et al. [66]. For quality control, prior to measurement of every element, system calibration was performed using standard solutions of that respective elements. The system ionized argon gas under a high frequency electric current to create plasma which provided enough energy (from the high-density electron and temperature of 10000 K) for atomization of the samples [67]. The entire system was warmed up for 15 min before actual quantification. The samples were loaded into the designated test tubes and the test tubes were placed into an auto sampler equipped with a sampling probe and a peristaltic pump for automatic operation of trace metals detection by the system itself. The detection wavelengths used for trace metals quantification were selected based on Goncalves et al. [68], and Oral et al. [69] which were 228.812, 226.506, 206.157, 327.395, 238.204, 257.6, 231.604, 220.350, and 213.857 nm respectively for As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn. Standard curve for all trace metals was constructed from prepared standard series and the trace metal contents were expressed as mg L^{-1} for water and mg kg^{-1} for both soil and plant.

2.6. Calculation of bio-concentration factors and pollution load index

Bio-concentration factor (BCF) is the major pathway for human exposure to trace elements and is used to quantify the transfer of trace metals from soil to edible plant parts [70]. The degree of trace metal translocation from soil to potato tubers was determined via BCF estimation following Marrugo-Negrete et al. [71] using the formula: [$BCF = C_{plant}/C_{soil}$; where, C_{plant} and C_{soil} represent the trace metal concentrations of the plant and soil samples, respectively].

Soil pollution load index (PLI) not only indicates the status of trace metals in soil but also insights regarding the extent of their saturation [57]. Soil PLI was measured following the method described by Khan et al. [70] using the following formula: [$PLI = C_{soil}/C_{reference}$; where, C_{soil} and $C_{reference}$ represent the trace metal concentrations of the soil samples and reference soils, respectively]. During calculation of PLI, maximum permissible limit of trace metals in soil was considered as its $C_{reference}$ value.

2.7. Estimation of human health risk index

Health risk index (HRI) measures the risks associated with consumption of food products (potato tubers in this case) grown on soil irrigated with wastewater [57]. Human HRI was assessed according to Khan et al. [70] using the formula: [$HRI = DIM/RfD$; where, DIM is the daily intake of metals through consumption of contaminated potato and RfD is the reference oral dose for each metal according to USEPA [72] (Table 2S)]. Daily intake of metals (DIM) was determined by the following equation: [$DIM = (CM \times DI)/BW$; where, CM, DI and BW represents trace metal concentrations in plants (mg kg^{-1}), daily intake of potato tuber and average human body weight, respectively] [73]. The average daily potato intake was considered 64.8 g $person^{-1}$ day^{-1} according to the report of Bangladesh Bureau of Statistics [46] and the average human body weight were considered 52.5 kg according to the report of World Health Organization [74].

Table 2
Trace metal concentration (mg L^{-1}) in irrigation water (groundwater, surface water, and wastewater) samples collected from the three experimental sites.

Trace metal	Experimental site	Cropping Year			Maximum Permissible limit
		2019	2020	2021	
As	Site-I (GWI)	0.0103 ± 0.0003 cA	0.0106 ± 0.0005 bA	0.0106 ± 0.0007 bA	0.1[c,d]
	Site-II (SWI)	0.0109 ± 0.0003 bA	0.0112 ± 0.0003 bA	0.0113 ± 0.0002 bA	
	Site-III (WWI)	0.0211 ± 0.0002 aC	0.0225 ± 0.0002 aB	0.0241 ± 0.0003 aA	
Cd	Site-I (GWI)	0.0098 ± 0.0002 cA	0.0099 ± 0.0002 bA	0.0099 ± 0.0003 cA	0.01[a,b]
	Site-II (SWI)	0.0128 ± 0.0001 bA	0.0132 ± 0.0004 bA	0.0134 ± 0.0002 bA	
	Site-III (WWI)	0.0536 ± 0.0004 aC	0.0592 ± 0.0004 aB	0.0654 ± 0.0005 aA	
Cr	Site-I (GWI)	0.0782 ± 0.0003 bA	0.0796 ± 0.0009 bA	0.0798 ± 0.0006 cA	0.1[b]
	Site-II (SWI)	0.0797 ± 0.0004 bB	0.0836 ± 0.0003 bB	0.0897 ± 0.0005 bA	
	Site-III (WWI)	0.2150 ± 0.0035 aA	0.2534 ± 0.0059 aA	0.2768 ± 0.0021 aA	
Cu	Site-I (GWI)	0.0604 ± 0.0002 cB	0.0626 ± 0.0003 cA	0.0639 ± 0.0008 cA	0.2[b]
	Site-II (SWI)	0.0904 ± 0.0014 bC	0.0929 ± 0.0013 bB	0.0968 ± 0.0011 bA	
	Site-III (WWI)	0.1480 ± 0.0009 aC	0.1740 ± 0.0033 aB	0.2061 ± 0.0044 aA	
Fe	Site-I (GWI)	0.2725 ± 0.0024 cB	0.2806 ± 0.0018 bA	0.2826 ± 0.0061 cA	5[a,b]
	Site-II (SWI)	0.2865 ± 0.0012 bB	0.2936 ± 0.0031 bAB	0.3027 ± 0.0030 bA	
	Site-III (WWI)	0.4530 ± 0.0028 aC	0.4816 ± 0.0075 aB	0.5327 ± 0.0055 aA	
Mn	Site-I (GWI)	0.0323 ± 0.0005 cA	0.0347 ± 0.0004 bA	0.0390 ± 0.0005 cA	0.2[a,b]
	Site-II (SWI)	0.0523 ± 0.0011 bA	0.0571 ± 0.0009 bA	0.0595 ± 0.0004 bA	
	Site-III (WWI)	0.1441 ± 0.0013 aC	0.1657 ± 0.0016 aB	0.1903 ± 0.0015 aA	
Ni	Site-I (GWI)	0.1332 ± 0.0010 cC	0.1362 ± 0.0016 cB	0.1419 ± 0.0012 cA	0.2[a]
	Site-II (SWI)	0.1684 ± 0.0008 bC	0.1771 ± 0.0009 bB	0.1835 ± 0.0020 bA	
	Site-III (WWI)	0.2157 ± 0.0048 aB	0.2306 ± 0.0022 aB	0.2865 ± 0.0057 aA	
Pb	Site-I (GWI)	0.0439 ± 0.0003 cA	0.0433 ± 0.0005 cA	0.0438 ± 0.0002 cA	0.1[a]
	Site-II (SWI)	0.0535 ± 0.0002 bC	0.0546 ± 0.0003 bB	0.0577 ± 0.0003 bA	
	Site-III (WWI)	0.0965 ± 0.0026 aB	0.1012 ± 0.0015 aB	0.1358 ± 0.0045 aA	
Zn	Site-I (GWI)	0.2117 ± 0.0046 cC	0.2198 ± 0.0015 cB	0.2291 ± 0.0022 cA	2[b]
	Site-II (SWI)	0.2590 ± 0.0081 bA	0.2727 ± 0.0041 bA	0.2988 ± 0.0029 bA	
	Site-III (WWI)	0.4511 ± 0.0058 aC	0.4913 ± 0.0088 aB	0.5281 ± 0.0024 aA	

Data are means of four replications ± standard errors. For every trace metal, means within a column followed by the different lowercase letters and means within a row followed by different uppercase letters are statistically different based on Tukey's honest significant difference test at $P \leq 0.05$. GWI−groundwater irrigation, SWI−surface water irrigation, WWI−wastewater irrigation.
Source: [a]Ahmad et al. [10], [b]Ayers and Westcot [75], [c]Chiroma et al. [76], [d]Khan et al. [57].

2.8. Statistical analysis

Data obtained were subjected to two-way analysis of variance (ANOVA) for trace metal contents of water, soil and potato tubers using Statistix10 (Analytical Software, Tallahassee, FL, USA) software program. Means for significant treatment effects were compared by Tukey's honest significant difference test at $P \leq 0.05$. Data for significant treatment effect are presented based on the highest order of factorial combination that was significant in the ANOVA.

3. Results

The two-way ANOVA of the tested trace metal contents of irrigation water, topsoil and potato tubers from the three selected experimental sites (S) over three consecutive years (Y) revealed significant variations (Table 1). Both the individual and interactive effects were considerably significant; however, the interactive effect (S × Y) was found non-significant for Cr, and Zn in water; Cd, Cr, Mn, and Ni in topsoil; and Cu, Fe, and Pb in potato tuber. The individual effect of the experimental site (S) was highly significant ($P \leq 0.01$) for both water, topsoil and potato tubers. Likewise, the individual effect of cropping year (Y) was also significant except for Cd, and Zn in soil; and Cu, and Fe in potato tuber (Table 1).

3.1. Trace metal status of irrigation water

The trace metal concentration of irrigation water of the three experimental sites varied considerably. Regardless of the year, all the trace metal contents in the coalmine wastewater used for irrigating site-III were significantly and substantially higher than both the site-I and site-II irrigation water. The surface water of site-II had statistically higher trace metal contents compared to the groundwater of site-I for most of the tested metals regardless of the cropping year; however, irrigation water from both sites showed statistically similar As content during 2020, and 2021; Cd content during 2020; and Cr content during 2019, and 2020 (Table 2). On the other hand, while comparing the trace metal contents of the irrigation water sampled from the three experimental sites over three consecutive years, we found a gradually increasing trend for all the nine tested trace metals in the irrigation water. Although most of the trace metal contents of water from the three sites remained below the maximum permissible limit within the tested time period, Cd, Cr and Ni content of site-III wastewater was found above the maximum permissible limit. Cadmium (Cd) content of groundwater was found to be almost equal to its maximum permissible limit, whereas in surface water it was a little higher (Table 2). Moreover, in 2020,

Table 3
Trace metal concentration (mg kg^{-1}) in topsoil samples collected from the three experimental sites.

Trace metal	Experimental site	Cropping Year			Maximum Permissible limit
		2019	2020	2021	
As	Site-I (GWI)	8.116 ± 0.292 bA	8.231 ± 0.167 bA	8.342 ± 0.202 bA	20[b,e]
	Site-II (SWI)	8.228 ± 0.150 bA	8.369 ± 0.146 bA	8.419 ± 0.244 bA	
	Site-III (WWI)	13.525 ± 0.236 aB	14.556 ± 0.177 aA	15.260 ± 0.196 aA	
Cd	Site-I (GWI)	0.230 ± 0.008 bA	0.237 ± 0.006 bA	0.224 ± 0.007 cA	3[g]
	Site-II (SWI)	0.259 ± 0.003 bA	0.264 ± 0.001 bA	0.288 ± 0.003 bA	
	Site-III (WWI)	0.373 ± 0.004 aC	0.410 ± 0.009 aB	0.445 ± 0.008 aA	
Cr	Site-I (GWI)	15.533 ± 0.352 cA	15.573 ± 0.858 cA	15.641 ± 0.223 cA	50[d]
	Site-II (SWI)	19.869 ± 0.556 bA	20.182 ± 0.944 bA	20.870 ± 0.391 bA	
	Site-III (WWI)	30.774 ± 0.483 aC	31.984 ± 0.485 aB	34.742 ± 0.790 aA	
Cu	Site-I (GWI)	19.170 ± 0.201 cB	20.465 ± 0.270 cA	20.618 ± 0.168 cA	50[g]
	Site-II (SWI)	25.433 ± 0.397 bB	26.053 ± 0.373 bB	27.960 ± 0.296 bA	
	Site-III (WWI)	28.863 ± 0.273 aC	30.880 ± 0.235 aB	33.043 ± 0.449 aA	
Fe	Site-I (GWI)	25755.8 ± 113.1 cA	26170.0 ± 168.3 cA	26342.6 ± 103.2 cA	21000[f]
	Site-II (SWI)	27218.2 ± 130.6 bB	27902.2 ± 139.5 bB	29770.9 ± 120.1 bA	
	Site-III (WWI)	32090.5 ± 170.4 aB	34998.9 ± 154.1 aA	35488.0 ± 166.9 aA	
Mn	Site-I (GWI)	306.484 ± 5.098 cB	326.211 ± 3.039 cA	339.179 ± 4.720 cA	2000[a,c]
	Site-II (SWI)	383.076 ± 3.071 bB	396.621 ± 4.057 bAB	417.188 ± 5.834 bA	
	Site-III (WWI)	516.768 ± 3.765 aC	529.733 ± 5.314 aB	554.961 ± 4.461 aA	
Ni	Site-I (GWI)	9.046 ± 0.360 cA	9.253 ± 0.046 cA	9.719 ± 0.122 cA	50[b,c,e]
	Site-II (SWI)	10.469 ± 0.206 bB	11.285 ± 0.233 bAB	11.950 ± 0.233 bA	
	Site-III (WWI)	13.150 ± 0.309 aA	14.135 ± 0.259 aA	14.951 ± 0.313 aA	
Pb	Site-I (GWI)	10.470 ± 0.143 cA	10.553 ± 0.215 cA	10.941 ± 0.213 cA	100[b,e,g]
	Site-II (SWI)	14.185 ± 0.321 bB	14.743 ± 0.268 bAB	15.562 ± 0.151 bA	
	Site-III (WWI)	21.753 ± 0.158 aC	23.370 ± 0.148 aB	26.476 ± 0.197 aA	
Zn	Site-I (GWI)	45.921 ± 0.422 bB	45.914 ± 0.322 cB	58.099 ± 0.407 cA	200[b,e,g]
	Site-II (SWI)	63.210 ± 0.778 bB	64.860 ± 0.411 bAB	66.316 ± 0.315 bA	
	Site-III (WWI)	77.212 ± 0.416 aA	80.291 ± 0.864 aA	82.076 ± 0.652 aA	

Data are means of four replications ± standard errors. For every trace metal, means within a column followed by the different lowercase letters and means within a row followed by different uppercase letters are statistically different based on Tukey's honest significant difference test at $P \leq 0.05$.
GWI−groundwater irrigation, SWI−surface water irrigation, WWI−wastewater irrigation.
Source: aAhmad et al. [10]; bChiroma et al. [76]; cEuropian Union [77]; dFAO/WHO [78]; eKhan et al. [57]; fUSEPA [79]; gUSEPA [80].

the Pb content of site-III wastewater and in 2021, the Cu and Fe contents of the wastewater from the same site rose above their respective maximum permissible limits. The level of trace metals in irrigation water undoubtedly raised during 2019–2021 at the three experimental sites, but the increase in site-I groundwater as well as site-II surface water did not show much statistical significance for most of the tested trace metals. Contrariwise, all tested trace metals content at the site-III wastewater significantly increased during this time (Table 2).

3.2. Trace metal status of cultivating soil

Considerable variations in the trace metal contents of the soil samples were evident across the three experimental sites over the tested time and vice versa. All the nine trace metals of site-III soil samples were found significantly and substantially higher compared to both site-I and site-II soils regardless of the cropping year. Similarly, trace metal levels of site-II soil were also significantly higher than site-I soil except for As. Cadmium (Cd) content of site-II soil was statistically similar to site-I till 2020; however, in 2021 it was statistically higher (Table 3). We also recorded a gradual increase in the tested trace metal contents over the selected three consecutive years regardless of the experimental site. Despite the increasing trend there was not much considerable statistical significance. For instance, most of the trace metal levels remained statistically the same in the soil samples of site-I and site-II except Cu, Mn, and Zn. In the case of site-III soil, As, Cd, Cr, Cu, Fe, Mn, and Pb content increased significantly over the years; whereas, the other trace metal levels remained statistically similar (Table 3). Apart from these variations, it was found that all tested trace metal contents scored below their maximum permissible limit regardless of the cropping year as well as the experimental site; however, Fe level was the only exception (Table 3).

3.3. Trace metal status of potato tubers

In the potato samples, all the nine tested trace metal contents showed significant variations across the three experimental sites as well as over the three-years. We recorded that all trace metal contents were significantly higher in potato tubers sampled from site-III compared to those sampled from site-I and site-II regardless of the cropping year. Similarly, potato tubers sampled from site-II also showed significantly higher trace metal contents compared to site-I with one and only exception in case of Cr content (Table 4). On the other hand, it was observed that an increasing trend for all the tested trace metals accumulated in potato tubers across the time period from 2019 to 2021 regardless of the experimental site (Table 4). The increasing trace metal accumulation over these three consecutive

Table 4
Trace metal concentration (mg kg^{-1}) in potato tuber samples collected from the three experimental sites.

Trace metal	Experimental site	Cropping Year			Maximum Permissible limit
		2019	2020	2021	
As	Site-I (GWI)	0.366 ± 0.010 cB	0.371 ± 0.011 cA	0.372 ± 0.004 cA	7[a,c]
	Site-II (SWI)	0.383 ± 0.008 bB	0.391 ± 0.013 bAB	0.399 ± 0.003 bA	
	Site-III (WWI)	0.688 ± 0.012 aB	0.704 ± 0.009 aB	0.738 ± 0.022 aA	
Cd	Site-I (GWI)	0.0325 ± 0.002 cC	0.0334 ± 0.001 cB	0.0342 ± 0.003 cA	0.2[b]
	Site-II (SWI)	0.0347 ± 0.002 bB	0.0359 ± 0.002 bB	0.0413 ± 0.001 bA	
	Site-III (WWI)	0.0828 ± 0.005 aC	0.0887 ± 0.004 aB	0.0993 ± 0.004 aA	
Cr	Site-I (GWI)	0.359 ± 0.009 bA	0.361 ± 0.007 bA	0.366 ± 0.008 bA	2.3[b]
	Site-II (SWI)	0.368 ± 0.009 bA	0.375 ± 0.011 bA	0.380 ± 0.005 bA	
	Site-III (WWI)	0.539 ± 0.010 aB	0.638 ± 0.012 aB	0.773 ± 0.012 aA	
Cu	Site-I (GWI)	6.491 ± 0.124 cA	6.531 ± 0.169 cA	6.541 ± 0.089 bA	73[b,c]
	Site-II (SWI)	8.289 ± 0.197 bA	8.547 ± 0.091 bA	8.788 ± 0.145 bA	
	Site-III (WWI)	12.918 ± 0.220 aA	13.656 ± 0.310 aA	14.739 ± 0.132 aA	
Fe	Site-I (GWI)	56.604 ± 1.741 cA	57.194 ± 1.352 cA	57.540 ± 1.485 cA	425[b,c]
	Site-II (SWI)	66.092 ± 1.410 bC	67.599 ± 1.629 bB	69.300 ± 1.615 bA	
	Site-III (WWI)	86.294 ± 1.082 aA	88.509 ± 1.928 aA	89.205 ± 1.516 aA	
Mn	Site-I (GWI)	12.147 ± 0.156 bA	12.237 ± 0.553 cA	12.560 ± 0.279 cA	500[b]
	Site-II (SWI)	13.389 ± 0.229 bC	13.723 ± 0.363 bB	14.396 ± 0.117 bA	
	Site-III (WWI)	20.037 ± 0.358 aC	22.379 ± 0.313 aB	25.891 ± 0.188 aA	
Ni	Site-I (GWI)	0.322 ± 0.002 cA	0.325 ± 0.006 cA	0.332 ± 0.004 cA	67[b]
	Site-II (SWI)	0.454 ± 0.008 bC	0.516 ± 0.020 bB	0.564 ± 0.015 bA	
	Site-III (WWI)	1.199 ± 0.047 aB	1.436 ± 0.042 aA	1.523 ± 0.012 aA	
Pb	Site-I (GWI)	0.042 ± 0.001 cA	0.044 ± 0.001 cA	0.044 ± 0.001 bA	0.3[b,c]
	Site-II (SWI)	0.088 ± 0.003 bB	0.092 ± 0.003 bB	0.097 ± 0.002 bA	
	Site-III (WWI)	0.198 ± 0.007 aA	0.212 ± 0.008 aA	0.244 ± 0.005 aA	
Zn	Site-I (GWI)	5.643 ± 0.171 bA	5.914 ± 0.084 cA	6.163 ± 0.138 cA	99[b,c]
	Site-II (SWI)	6.064 ± 0.114 bB	6.370 ± 0.209 bAB	6.645 ± 0.078 bA	
	Site-III (WWI)	9.315 ± 0.377 aC	10.729 ± 0.338 aB	12.095 ± 0.241 aA	

Data are means of four replications ± standard errors. For every trace metal, means within a column followed by the different lowercase letters and means within a row followed by different uppercase letters are statistically different based on Tukey's honest significant difference test at $P \leq 0.05$. GWI−groundwater irrigation, SWI−surface water irrigation, WWI−wastewater irrigation.
Source: aChiroma et al. [76]; bFAO/WHO [78]; cKhan et al. [57].

years in potato tubers revealed variable significance depending on the site. For example, in case of potato tubers sampled from site-I, most of the tested trace metals did not increase much in amount but rather remained statistically similar. In case of potato tubers sampled from site-II, all the trace metals except Cr and Cu showed a significant increase in accumulation level. Similarly, potato tubers sampled from site-III also showed a significant increase in all tested trace metals accumulation levels except Cu and Fe within the designated time (Table 4). Moreover, it was also observed that all nine trace metal contents of potato tubers remained below the maximum permissible limit regardless of the experimental site as well as the cropping year (Table 4).

3.4. The extent of soil contamination with trace metals

The extent of trace metal contamination in soil was determined by pollution load index (PLI) that showed a wide variation across the three experimental sites. The highest PLI values for all nine trace metals were recorded in soil of experimental site-III regardless of the cropping year, which were also found significantly higher compared to the PLI values obtained for both site-I and site-II (Table 5). Pollution load index for As contamination did not show any statistical difference between soils of site-I and site-II regardless of the cropping year. Similar to that, PLI values for Cd (during 2019 and 2020), and Zn (during 2019) were also found statistically similar in soils between site-I and site-II. The rest of the trace metals associated PLI values for site-II were significantly higher than site-I (Table 5). It was also observed an increasing trend of PLI values during the three consecutive cropping years regardless of the experimental site; however, there was not much statistical significance recorded. In case of site-I, all of the trace metals showed minute and non-significant rises in PLI values from 2019 to 2021 except Cu, Mn, and Zn. Likewise, in site-II only Cu, Mn, Ni, Pb and Zn showed significant increase in PLI values during the above-mentioned time period. Contrariwise, in case of the site-III, all of the trace metals showed a significant increase in PLI values from 2019 to 2021 except Zn (Table 5). We also noticed that all of the trace metals secured PLI values < 1 with Fe as the only exception scoring PLI values > 1 (Table 5).

3.5. Translocation of trace metals from soils to potato tubers

Trace metal transfer from the soil of the three experimental sites to the potato tubers cultivated at those sites was determined by calculation of bio-concentration factor (BCF). Among the three experimental sites, we found that the BCF values were comparatively higher in site-III than in both site-I and site-II which was statistically significant for most of the tested trace metals (Table 6). However, there was not much significant difference in BCF values in between site-I and site-II. Similarly, we also did not record much statistical variation in trace metals BCF value while comparing among the cropping years regardless of the experimental sites (Table 6). Overall,

Table 5
Pollution load index (PLI) for the trace metals in soils of the three experimental sites.

Trace metal	Experimental site	Cropping Year		
		2019	2020	2021
As	Site-I (GWI)	0.406 bA	0.412 bA	0.417 bA
	Site-II (SWI)	0.411 bA	0.418 bA	0.421 bA
	Site-III (WWI)	0.676 aB	0.728 aA	0.763 aA
Cd	Site-I (GWI)	0.077 bA	0.079 bA	0.075 cA
	Site-II (SWI)	0.086 bA	0.088 bA	0.096 bA
	Site-III (WWI)	0.124 aC	0.137 aB	0.148 aA
Cr	Site-I (GWI)	0.311 cA	0.311 cA	0.313 cA
	Site-II (SWI)	0.397 bA	0.404 bA	0.417 bA
	Site-III (WWI)	0.615 aC	0.640 aB	0.695 aA
Cu	Site-I (GWI)	0.383 cB	0.409 cA	0.412 cA
	Site-II (SWI)	0.509 bB	0.521 bB	0.559 bA
	Site-III (WWI)	0.577 aC	0.618 aB	0.661 aA
Fe	Site-I (GWI)	1.226 cA	1.246 cA	1.254 cA
	Site-II (SWI)	1.296 bB	1.329 bB	1.418 bA
	Site-III (WWI)	1.528 aB	1.667 aA	1.690 aA
Mn	Site-I (GWI)	0.153 cB	0.163 cA	0.170 cA
	Site-II (SWI)	0.192 bB	0.198 bAB	0.209 bA
	Site-III (WWI)	0.258 aC	0.265 aB	0.277 aA
Ni	Site-I (GWI)	0.181 cA	0.185 cA	0.194 cA
	Site-II (SWI)	0.209 bB	0.226 bAB	0.239 bA
	Site-III (WWI)	0.263 aA	0.283 aA	0.299 aA
Pb	Site-I (GWI)	0.105 cA	0.106 cA	0.109 cA
	Site-II (SWI)	0.142 bB	0.147 bAB	0.156 bA
	Site-III (WWI)	0.218 aC	0.234 aB	0.265 aA
Zn	Site-I (GWI)	0.230 bB	0.230 cB	0.290 cA
	Site-II (SWI)	0.316 bB	0.324 bAB	0.332 bA
	Site-III (WWI)	0.386 aA	0.401 aA	0.410 aA

For every trace metal, means within a column followed by the different lowercase letters and means within a row followed by different uppercase letters are statistically different based on Tukey's honest significant difference test at $P \leq 0.05$. GWI−groundwater irrigation, SWI−surface water irrigation, WWI−wastewater irrigation.

we observed that the BCF trend for trace metals were in order of Cu > Cd > Zn > Ni > As > Mn > Cr > Pb > Fe (Table 6).

3.6. *Intake rate of trace metals and associated health risks*

Accumulating high amounts of trace metals in food products potentiates a great health risk which has been expressed as health risk index (HRI) relating directly to the daily trace metals intake through oral consumption. In our study, we recorded HRI values ranging between 0.015 and 0.954 and there were significant variations among them across the three experimental sites over the three consecutive years and vice versa. We also found that the HRI trend for trace metals in potato tubers was in the order of Cr > Cu > Mn > Fe > Cd > Ni > As > Pb > Zn (Table 7). Potato tubers harvested from site-III showed significantly and substantially higher HRI values for all the nine trace metals compared to the tubers harvested from site-I and site-II regardless of the cropping year. Likewise, tubers from site-II showed significantly higher HRI values compared to the tubers from site-I for all the nine trace metals except Mn during 2019 (Table 7). Moreover, during the period from 2019 to 2021, we observed an increasing trend of HRI values regardless of the experimental site, and the increase over time did reveal quite significant differences. For example, in case of site-III, HRI values associated with As, Cd, Cr, Mn, Ni and Zn increased significantly in 2021 compared to previous years. A similar phenomenon was observed for As, Cd, Mn, Ni, Pb and Zn in the case of site-II. Contrariwise, HRI values associated with trace metals of site-I did not show such significant variations except for As and Cd (Table 7).

4. Discussion

Trace metal contents in irrigation water are a very crucial water quality parameter for ensuring soil health as well as food crop safety. Irrigation with contaminated water can actively pollute soil with trace metals which may get transported into the human body through the food chain [40,42,43,45,57]. Complete exclusion of trace metals from the food system is both impossible and unnecessary because some of the trace metals like Cu, Fe, Zn, Mn, and Ni are essential for living beings [81]. Thus, a maximum permissible limit has been set for trace metals in water, soil, and plant products to avoid unwanted pollution, toxicity and health disorders (Tables 2–4). In the present study, three possible crop irrigation water sources, namely groundwater (site-I), surface water (site-II) and coalmine wastewater (site-III) were tested for nine trace metal status (As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) both in irrigation water, cultivation soil, and potato tubers grown in the soil over three consecutive years compared with their respective maximum permissible limits. Coal is a highly valuable natural resource and coal mines are a very important as they help in industrial energy supply [82,83]. The target location of the study is one of the biggest coal mining sites located at the heart of a vast cropping lands of northern Bangladesh.

Table 6
Bio-concentration factor of trace metals in potato tubers from the three experimental sites.

Trace metal	Experimental site	Cropping Year		
		2019	2020	2021
As	Site-I (GWI)	0.045 aA	0.045 aA	0.045 aA
	Site-II (SWI)	0.047 aA	0.047 aA	0.048 aA
	Site-III (WWI)	0.051 aA	0.048 aA	0.048 aA
Cd	Site-I (GWI)	0.144 bA	0.141 bA	0.154 bA
	Site-II (SWI)	0.134 bA	0.147 abA	0.144 bA
	Site-III (WWI)	0.222 aA	0.217 aA	0.224 aA
Cr	Site-I (GWI)	0.019 bA	0.019 aA	0.018 aA
	Site-II (SWI)	0.018 bB	0.020 aAB	0.022 aA
	Site-III (WWI)	0.023 aA	0.023 aA	0.024 aA
Cu	Site-I (GWI)	0.339 bA	0.319 bA	0.318 bA
	Site-II (SWI)	0.326 bA	0.328 bA	0.314 bA
	Site-III (WWI)	0.448 aA	0.442 aA	0.446 aA
Fe	Site-I (GWI)	0.0022 cA	0.0022 cA	0.0022 bA
	Site-II (SWI)	0.0024 bA	0.0024 bA	0.0023 abB
	Site-III (WWI)	0.0027 aA	0.0025 aA	0.0025 aA
Mn	Site-I (GWI)	0.040 aA	0.038 bA	0.037 bA
	Site-II (SWI)	0.035 aA	0.035 cA	0.035 cA
	Site-III (WWI)	0.039 aC	0.042 aB	0.047 aA
Ni	Site-I (GWI)	0.036 cA	0.035 cA	0.034 cA
	Site-II (SWI)	0.043 bA	0.046 bA	0.047 bA
	Site-III (WWI)	0.091 aA	0.102 aA	0.103 aA
Pb	Site-I (GWI)	0.0041 cA	0.0041 cA	0.0040 cA
	Site-II (SWI)	0.0062 bA	0.0062 bA	0.0062 bA
	Site-III (WWI)	0.0091 aA	0.0091 aA	0.0092 aA
Zn	Site-I (GWI)	0.098 bB	0.129 aA	0.134 aA
	Site-II (SWI)	0.096 bA	0.098 bA	0.100 bA
	Site-III (WWI)	0.121 aA	0.134 aA	0.150 aA

For every trace metal, means within a column followed by the different lowercase letters and means within a row followed by different uppercase letters are statistically different based on Tukey's honest significant difference test at $P \leq 0.05$. GWI−groundwater irrigation, SWI−surface water irrigation, WWI−wastewater irrigation.

Table 7
Health risk index associated with trace metals intake through oral consumption of potato tubers from the three experimental sites.

Trace metal	Experimental site	Cropping Year		
		2019	2020	2021
As	Site-I (GWI)	0.032 cB	0.033 cA	0.033 cA
	Site-II (SWI)	0.034 bB	0.034 bAB	0.035 bA
	Site-III (WWI)	0.061 aB	0.062 aB	0.065 aA
Cd	Site-I (GWI)	0.040 cC	0.041 cB	0.042 cA
	Site-II (SWI)	0.043 bB	0.044 bB	0.051 bA
	Site-III (WWI)	0.102 aC	0.110 aB	0.123 aA
Cr	Site-I (GWI)	0.443 bA	0.445 bA	0.452 bA
	Site-II (SWI)	0.454 bA	0.463 bA	0.469 bA
	Site-III (WWI)	0.665 aB	0.788 aB	0.954 aA
Cu	Site-I (GWI)	0.200 cA	0.202 cA	0.202 bA
	Site-II (SWI)	0.256 bA	0.264 bA	0.271 bA
	Site-III (WWI)	0.399 aA	0.421 aA	0.455 aA
Fe	Site-I (GWI)	0.100 cA	0.101 cA	0.101 cA
	Site-II (SWI)	0.117 bC	0.119 bB	0.122 bA
	Site-III (WWI)	0.152 aA	0.156 aA	0.157 aA
Mn	Site-I (GWI)	0.107 bA	0.108 cA	0.111 cA
	Site-II (SWI)	0.118 bC	0.121 bB	0.127 bA
	Site-III (WWI)	0.177 aC	0.197 aB	0.228 aA
Ni	Site-I (GWI)	0.020 cA	0.020 cA	0.021 cA
	Site-II (SWI)	0.028 bC	0.032 bB	0.035 bA
	Site-III (WWI)	0.074 aB	0.089 aA	0.094 aA
Pb	Site-I (GWI)	0.015 cA	0.015 cA	0.015 bA
	Site-II (SWI)	0.030 bB	0.031 bB	0.033 bA
	Site-III (WWI)	0.068 aA	0.073 aA	0.084 aA
Zn	Site-I (GWI)	0.023 bA	0.024 cA	0.025 cA
	Site-II (SWI)	0.025 bB	0.026 bAB	0.027 bA
	Site-III (WWI)	0.038 aC	0.044 aB	0.050 aA

For every trace metal, means within a column followed by the different lowercase letters and means within a row followed by different uppercase letters are statistically different based on Tukey's honest significant difference test at $P \leq 0.05$. GWI−groundwater irrigation, SWI−surface water irrigation, WWI−wastewater irrigation.

Adjacent to the mining factory, there exists three big surface water reservoirs (a canal and two lakes) which supply a substantial amount of water for irrigation activity of the area. The mine yields about 0.6 million tonnes of coal annually [84] and during its operation effluents can easily find their way to contaminate these surface water resources both directly and indirectly [83,85]. Studies have been conducted in the past in the same target location that reported alarming accumulation of trace metals particularly Ba, Cr, Ni, Cu, Ti, Mn, Zn, Pb, As, Fe, Rb, Sr, Nb and Zr in the soil of the mine drainage lines; as well as toxic levels of Mn, Fe and Ni in the mine discharged wastewater [82,83,85–87]. In another study, Hossain et al. [88] also suggested that coalmine effluents not only deteriorated the surrounding water and soil quality but also showed significant changes in pH values, heavy metal, organic carbon, and exchangeable cations of nearby farmland soils. Such intriguing results reflected the risk of trace elements contamination in soil and water resources due to coalmine wastewater-irrigation and insighted potential health hazards in the long run [83,89] that has been addressed in the current study. Our results revealed that, irrigation with groundwater remained safe as all the trace metal contents were below their respective maximum permissible limits; however, this was not the case with surface water and wastewater. In the case of surface water, we only found Cd exceeding its maximum permissible limit but in case of wastewater total six trace metals (Cd, Cr, Cu, Fe, Pb and Ni) were available higher than their respective maximum permissible limits (Table 2). This effectively reflected the unsuitability of the coalmine wastewater for irrigation purposes. Several previous studies also have outlined the high flux of trace metals in the discharged liquid from the Barapukuria coalmine [82,83,85,87] which was the most likely reason for the contamination of the adjacent waterbodies. On the other hand, use of groundwater may be safe for irrigation but due to the increasing trend of Cd content in groundwater over time, it might become risky in years to come. The reason behind the high level of Cd content in ground water is the most likely result of aquifer contamination by leaching [23,26,83,87]; however, a lowered water table might be another possible reason as site-I irrigation water comes from a deep tube-well. Similar to groundwater, Cd content of surface water was relatively high and it was a bit higher than its maximum permissible limit (Table 2). Such a phenomenon can only occur due to surface water pollution either by runoff or seepage [90], which strongly suggests that exposure to heavy metal containing effluents from non-anthropogenic source are directly or indirectly responsible. Khan et al. [57] and Tiwari et al. [59] also reported similarly while examining the trace metal contamination of irrigation water from different sources.

Trace metal levels in soil raises over time if long-term wastewater irrigation is practiced [23,83,85]. The result of our present study also agrees with the statement. It was noted that, site-III soil (receiving long-term coalmine wastewater irrigation) accumulated substantially higher trace metals compared to soils of both site-I and site-II. Moreover, the trace metals level in site-III soil from 2019 to 2021 was rising at comparatively higher rate than the rest of the two (Table 3). Such trends are in agreement with previous reports regarding adverse effects on soil health due to irrigation with contaminated wastewater [10,21,44,45,56–59,83,85,91]. On the contrary, irrigation with groundwater and surface water at site-I and site-II soils respectively did not raise most trace metals content significantly as the irrigation water quality of both sites was more or less within their maximum permissible limit (Tables 2 and 3). We also noticed that all tested trace metals level in soils regardless of the site remained below their respective MPLs except Fe which was above its MPL in all three sites (Table 3). Iron remains the fourth most abundant element in soil that is less prone to leaching and runoff loss due to its strong bonding capacity with soil particles (sesquioxide) and deposition as insoluble ferric (Fe^{3+}) form both of which are non-toxic in nature [92,93]. This gradual building-up nature of Fe in soil could be the reason for high Fe levels observed during our investigation. We also evaluated the soil pollution severity by determining soil pollution load index (PLI) during our study. The index is the ratio of every tested metal in the soil to its respective baseline or maximum permissible limits. So, PLI value > 1 indicates soil pollution [94,95]. In the present study, only Fe showed PLI value above 1 whereas all the other eight trace metals had PLI values below one (Table 5) which means that the soil from the three experimental sites is still in good condition. However, PLI values of trace metals of site-III soil kept increasing significantly between 2019 and 2021 and some of the trace metals notably As, Cr and Cu had PLI values of 0.763, 0.695 and 0.661 respectively (Table 5) indicating potential pollution hazards in near future as suggested by numerous previous investigations on similar topic [10,21,57,96].

Plants can uptake ionic elements from the soil solution because of the concentration gradient [97]. Therefore, excess trace metals could accumulate inside plants if they are available in soil in higher quantities [38,54,92]. In the present study we observed similar results, where trace metal contents in potato tubers harvested from wastewater irrigated site-III showed substantially higher trace metals compared to potato tubers harvested from groundwater irrigated site-I as well as surface water irrigated site-II (Table 4). The only possible explanation could be higher trace metal availability in site-III soil leading to higher uptake and accumulation in potato tubers [38,54,97]. Numerous previous studies investigating the trace metals accumulation in different plants as a result of wastewater irrigation also concluded the same way that, wastewater carried a substantial amount of trace metals that in turn increased soil trace metal levels and ultimately elevated trace metal accumulation in plant parts [5,10,21,57,58,96]. Our results on the soil trace metal contents of the three experimental sites also support the reasoning of high trace metal levels of potato tubers harvested from site-III (Table 3). Higher trace metals accumulation due to wastewater irrigation has also been reported in other vegetables like tomato and okra that also supported the results observed in our study [98,99]. However, trace metal accumulation rate in a particular soil could be variable depending on the plant species [100]. Thus, the translocation efficiency of trace metals from soil to plant could be an important criterion for assessing the cultivation suitability of a crop in contaminated soil which was determined as bio-concentration factor (BCF) in this study. Higher BCF value (>1) means higher trace metal accumulation in plant parts than soil [101]. During our investigation we did not observe BCF value exceeding 1 for all the trace metals; however, higher BCF values were recorded for site-III potato tubers compared to both site-I and site-II (Table 6). This again explains that plants tend to accumulate more trace metals if available in the soil. Several other reports are also in line with our results [10,57,59,70,96]. Moreover, over the tested three years period (2019–2021), trace metal accumulation increased quite significantly in potato tubers harvested from both site-II and site-III although trace metals accumulation remained below their respective MPLs regardless of the site or cropping year (Table 4). Based on this data it could be insighted that, potato produced from the three tested sites are safe for human consumption but still there could be a potent health risk from some of the trace metals because trace metal toxicity to human is exclusively related to the amount of exposer either by oral or dermal means. Potato is the most produced and consumed vegetable for Bangladeshi people and the consumption rate is quite high compared to many other countries of the world [102,103]. There is a good chance that people consuming potatoes grown on wastewater-irrigated soil might intake high quantities of some trace metals even though the trace metal contents of potato tubers remained within the safe zone. Thus, we determined the health risk index (HRI) of the nine trace metals through daily intake of potatoes in comparison with their respective reference oral dose outlined by USEPA [80]. Values of HRI$>$1 pose a greater risk to human health [58,104]. In our observations, we found substantially higher HRI values for potato tubers of site-III compared to both site-I and site-II (Table 7) meaning greater health risk potential for potatoes grown under coalmine wastewater irrigation. Moreover, we also observed a gradual increase in HRI values especially for site-III over three consecutive years (2019–2021) (Table 7). Many previous studies conducted in different parts of the world reported acute health risks for various heavy metals while assessing the health risks of food consumption grown under industrial and/or municipal wastewater irrigation [5,29,57,58,70,105,106] but we did not observe any HRI value > 1 for potato tubers grown under coalmine wastewater irrigation (Table 7). This confirms that consumption of potato tubers grown at site-III remained safe. However, we did notice the rapidly and significantly increasing trend of HRI values for all the trace metals at site-III during 2019–2021 which could potentially give rise to health risks in near future. In particular, HRI value of Cr for potato grown in site-III raised from 0.665 to 0.954 within three years and it already is very close to creating health issues like skin, lung and gastrointestinal disorders [31,32]. Although it is not the same case for other eight-trace metals but the trend seems to be similar, which is an alarming fact and could potentially become hazardous in upcoming future.

5. Conclusion

Irrigation of potato crops over three years using coalmine wastewater substantially increased trace metal concentrations in the topsoil and raised trace metal accumulation of potato tubers. Direct or indirect coalmine effluent discharge gradually contaminated nearby surface water resources with toxic levels of trace metals notably Cd, Cr, Cu, Fe, Ni and Pb rendering the water unsuitable for crop irrigation. Unmindful cropping with regular wastewater-irrigation alarmingly boosted trace metal accumulation rate both in soil and potato tubers. Although the study came to the conclusion that, soil of the wastewater irrigated site remained within the safe zone; however, wastewater irrigation should be checked to avoid any potential topsoil toxicity. Trace metal levels of potato tubers grown on wastewater-irrigated soil remained safe for consumption but the increasing rate of health risk index was quite alarming. Thus, coalmine wastewater use for irrigation should be immediately stopped to avoid any potential ecological and human health hazard in years to come. The results of this study should prove useful for strategizing waste management policies to avoid environmental intoxication as well as ensuring food safety.

Ethics approval

Not applicable.

Consent to participate

Not applicable.

Consent for publication

Not applicable.

CRediT authorship contribution statement

Jannatul Ferdoushi Asha: Writing – original draft, Methodology, Investigation, Conceptualization. **Sheikh Faruk Ahmed:** Writing – review & editing, Visualization, Formal analysis. **Arindam Biswas:** Writing – review & editing, Visualization. **Zannatul Ferdaous Bony:** Writing – review & editing, Methodology. **Md. Rizvi Chowdhury:** Writing – review & editing, Methodology, Investigation. **Bikash Chandra Sarker:** Writing – review & editing, Supervision, Methodology, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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