



Health risk assessment consequent to wastewater irrigation in Pakistan

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[Received: February 07, 2019 Accepted: December 19, 2019 Published Online: February 17, 2020]

Abstract

The aim of this study was to investigate the uptake and accumulation of heavy metals including cadmium, chromium, iron, nickel, and lead in 8 food crops irrigated with industrial wastewater and correlate the levels with potential human health risks. The concentrations of these metals in the food crops irrigated with wastewater decreased in the order iron (Fe) > chromium (Cr) > cadmium (Cd) > nickel (Ni) > lead (Pb). In all cases, the metallic contents in the vegetables exceeded the safe limits recommended by FAO/WHO. The highest bioaccumulation factor was recorded for cadmium. Further, the health risk indices of cadmium and lead were greater than 1 in seven food crops. For nickel, health risk was exceeded only for *Pisum sativum* and *Solanum tuberosum*. The indices for various food crops were in decreasing order: Cd > Pb > Ni > Cr > Fe. Out of 8 food crops, only *Cantiquorum antiquorum* showed risk indices lower than 1 for all 5 metals. Data demonstrated that food crops irrigated with wastewater in this study area are highly metal contaminated and may constitute a serious health risk to the local human and animal populations.

Keywords: Food crops, wastewater, health risk index, metals, health risk assessment

Introduction

Lack of freshwater has compelled farmers for extensive use of wastewater for food production in various countries including Pakistan. Both freshwater and wastewaters may contain organic and inorganic compounds that benefit plant growth, but the presence of hazardous metals which exceed recommended EPA (2008)/FAO (2016) standard permissible limits might occur and consequently may induce adverse effects in animals and humans (Jaishankar *et al.*, 2014; Iqbal *et al.*, 2016; Chen *et al.*, 2018b) Global concerns have been raised with respect to public health implications due to metal accumulation in crops and entrance into the food chain (Otitoju *et al.*, 2012).

Wastewater release from industry containing pollutants including metals when used for irrigation purposes may adversely affect food quality (Singh *et al.*, 2010; Qureshi *et al.*, 2016). Several investigators reported adverse health risks associated with the ingestion of contaminated food containing excessive

amounts of metals (Zhao *et al.*, 2014; Kaur and Dua 2015; Liao *et al.*, 2015; Silva *et al.*, 2016; Chen *et al.*, 2018a). Metal toxicity depends upon the concentration, oxidation state, exposure route, age, genetics, consumption level, and gender (Aelion *et al.*, 2008; Barbosa 2017; Branco *et al.*, 2017; Resende *et al.*, 2018). Excessive concentrations of metals may result in various deleterious effects. Similarly, Ni produces allergic contact dermatitis, eczema and respiratory problems (Christensen *et al.*, 1999; Yeganeh *et al.*, 2013; Ahlström *et al.*, 2018). Higher accumulation of Cd in humans might damage kidneys, bone and lungs (Casalino *et al.*, 2002; Liu *et al.*, 2014; Zhang *et al.*, 2014; Issermann *et al.*, 2017), exposure to Pb may produce CNS effects, renal dysfunction and blood disorders (Liu *et al.*, 2014; Serrazina *et al.*, 2018; O'Connor *et al.*, 2018; Shvachiy *et al.*, 2018).

From human health risk assessment perspective of metals through food consumption, daily intake and metal accumulation in foods are important factors. However, the metal accumulation in food crops and associated risk

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has thus far not been comprehensively explored in areas proximal to industrial sites in Pakistan. The Environmental Protection Agency (EPA) regulations in Pakistan strictly direct the use of safe irrigation water (EPA, 2008). Although various researchers reported the risk associated with wastewater irrigation and metal accumulation in food crops globally (Arora *et al.*, 2008; Jan *et al.*, 2010; Gupta *et al.*, 2013; Zhang *et al.*, 2018; León-Cañedo *et al.*, 2019), the consequences of ingestion of contaminated food crops to human health after long-term wastewater irrigation in Hattar Region of Pakistan still need to be addressed. Therefore, the aim of the present study was to determine the levels of heavy metals in water, soil and selected crops grown in the area irrigated with wastewater.

Materials and Methods

Sample collection

In total eight commonly used food crops *viz.*: *Colocasia esculenta* L (taro), *Colocasia antiquorum* L (eddoe), *Pisum sativum* L (pea), *Raphanus sativus* L (radish), *Rumex obtusifolius* L (broad-leaved dock), *Solanum tuberosum* L (potato), *Spinacia oleracea* L (spinach) and *Triticum aestivum* L (wheat) were collected randomly in pre-cleaned high-density 5 L polyethylene bottles from fields irrigated with wastewater in May-July, 2015 from selected wastewater irrigated areas of Hattar Industrial locations involved in steel rerolling, paper mills, food industries and pharmaceutical industrial.

Briefly, soil samples were collected from three different sampling sites (33°53'59" N 72°50'49" E, 33°53'30" N 72°51'14" E and 33.91°E 72.85°E, 33°54'08" N 72°51'10"E) that have been irrigated for long-term with industrial wastewater. Surface soil (0-20 cm) samples were collected in triplicate in a radius of 1.2 m as composite sample after thoroughly mixing in plastic bags (Wu *et al.*, 2015). These samples were air-dried and sieved via a 2-mm sieve. Thereafter, these samples were digested using mixture of H₂SO₄, HClO₄ and HNO₃ ((1:1:5 ratio) for the determination of total contents of heavy metals. Grab wastewater samples were collected in bottles from the points where fields were being irrigated with wastewater on sampling day. The samples were acidified with few drops of nitric acid to stop microbial activity.

The soil properties of the study area have been investigated by Haroon *et al.* (2019). According to this study, the soil pH was 7.6~8.7, EC was 554~728 μS cm⁻¹, total C was in range of 22 ~ 42 g kg⁻¹, CEC ranged 192~302 mg kg⁻¹ while its texture was loamy sand.

Plant sample analysis

Food crop samples were collected from three locations in triplicate. Briefly, edible parts of the crop were taken, washed with distilled water, oven dried for 24 h at 70-80 °C, ground, and subsequently sieved through 2 mm mesh (Jamali *et al.* 2009). Further, 1 g crop samples were digested with 15 mL mixture of H₂SO₄, HClO₄ and HNO₃ (1:1:5 ratio) (Sigma Aldrich-St. Louis, MO 63178, USA) at 100 °C until the solution became transparent. Subsequently, transparent solution was filtered through Whatman No. 42 filter paper and the filtrate were made up to 50 mL volume with distilled water and stored at 4 °C for analysis. Three water samples were filtered through Whatman No. 42 filter paper, and the final volume made up with distilled water to 50 mL.

Determination of heavy metals

Heavy metal concentrations were estimated using a graphite furnace atomic absorption spectroscopy (Model Analyst 700, PerkinElmer, Massachusetts, 02451, USA) in the Instrumental Lab, COMSATS Abbottabad, Pakistan. Standard metal solutions (1000 mg L⁻¹) (Perkin Elmer) were diluted to make concentration ranges of 1~100 mg L⁻¹. Drift and blank samples were run after each batch of four samples and all samples were analyzed in triplicate. Soil and plant standard reference materials (NIST, 2709 San Joaquin; NIST, 1547 peach leaves) of the National Institute of Standards and Technology (Gaithersburg, MD, USA) were used. Recoveries were: Cd 96%, Cr 96%, Fe 101%, Ni 98%, and Pb 97%.

Data analysis

Statistical analyses were conducted using open-source application RStudio version 1.1.383 for Windows (R Core Team, 2017; RStudio, 2016). Metal Bio-concentration factors (BCFs) were calculated as described by Khan *et al.* (2010) and Li *et al.* (2012) using the following equation.

$$BAF = \text{metal concentration in edible part} / \text{metal concentration in soil} \quad (1)$$

The metal pollution index (MPI) was calculated taking the average of all metals in the crops by using the equation (Usero *et al.*, 1997):

$$MPI(\mu\text{g g}^{-1}) = C_{f_1} \times C_{f_2} \times \dots \times C_{f_n}^{1/n} \quad (2)$$

The contamination was considered as zero, when the PI value was ≤ 1, slight contamination when PI was ≤ 3, moderate contamination was assumed if the PI values were in the range of 3 to 5 and the contamination was ranked as severe when the PI values > 5.



Daily intake of heavy metals (DIM) for individuals in the area (average weight 55 kg and average age 49 years) by consuming food crops was estimated according to the equation by Jan *et al.* (2010). Children (0-6 years old) usually consume 1/3 that of adults.

$$\text{"DIM}=(\text{metal concentration} \times \text{food intake})/(\text{average body weight})"$$

(3)

these sites, the highest water concentration of Pb was recorded at site 2. Whereas, the highest water concentrations of remaining metals Ni, Fe, Cd and Cr were detected at site 1. The difference in metal levels may be attributed to differing sources of wastewater such as “ghee”, soap, steel manufacturing, batteries, and paint industries.

Table 1: Concentrations of Cd, Cr, Pb, Ni and Fe (mg L⁻¹) in wastewater used for the irrigation of vegetable crops at different study sites and respective soils

Sampling site	Cd	Cr	Pb	Ni	Fe
Water samples (mg L⁻¹)					
SITE -1	2.96 (2.95)	0.21 (0.21)	3.70 (2.60)	8.73 (5.20)	2.96 (2.62)
SITE-2	0	0	6.72 (3.37)	0.01 (0.001)	0.03 (0.03)
SITE-3	1.48 (0.92)	0.10 (0.001)	5.22 (1.19)	4.36 (0.91)	1.49 (0.28)
Soil samples (mg kg⁻¹)					
SITE -1	22.2 (20.9)	16.8 (7)	98.1 (92.1)	21.0 (16.32)	501 (6.78)
SITE-2	5.4 (4.13)	33.5 (11.6)	17.6 (12.7)	5.3 (2.64)	501 (7.28)
SITE-3	0.9 (0.18)	30.5 (7.1)	15.2 (11.39)	5.9 (2.41)	499.1 (9.65)
Permissible limits*	0.5	n/a**	50	25	n/a

Each value was the mean of three replicates and standard error was given in parentheses. *Limits described by Rowell (1994) in mg kg⁻¹, **n/ a, not available.

To further assess the risk of exposure to heavy metals by consuming food crops produced in study area, Health risk indices (HRI) were estimated according to the equation described earlier (Singh *et al.*, 2010; Pandey *et al.*, 2012):

$$\text{HRI} = \frac{\sum_n (C_n \times \text{DIM})}{\text{RfD} \times B_w}$$

(4)

Where C_n is the average metal concentration in plants (mg kg⁻¹), DIM represents the average daily intake, RfD the reference oral dose, and B_w the average body weight.

One-way ANOVA was applied to determine significant variations within wastewater, soil and vegetable samples while two-way ANOVA was applied to evaluate the significant differences between heavy metal concentrations in food crops grown in wastewater and ground water.

Results and Discussion

Concentrations of heavy metals in irrigation water and soil

Wastewater collected from three water channels in Hattar was used to irrigate different food crops cultivated on both sides of these channels. The metal contents in collected water samples are presented in Table 1. Among

Soil samples obtained from three farmlands irrigated with industrial wastewater showed higher concentrations of selected heavy metal (Table 1). Highest soil contamination in terms of Cd, Pb, Ni and Fe were noted at site 1 (Table 1). Whereas the Cr concentration was highest at site 2, and Fe contamination was similar at all sites. On the basis of overall metal contamination, site 1 was highly contaminated, whereas site 2 was ranked as moderately contaminated.

Plant metal content

The edible parts of food crops presented high levels of metal content (in terms of mean concentrations) in a decreasing order: Fe > Cr > Cd > Ni > Pb. Metal uptake in food crops ranged from: Fe (180.1-506.6 mg/ kg, Figure 1a), Ni (1.86-17.7 mg kg⁻¹, Figure 1b), Pb (0.81-15.43 mg kg⁻¹, Figure 1c), Cr (1.28-355.8 mg kg⁻¹, Figure 1d), and Cd (0.55-59.17 mg kg⁻¹, Figure 1e). The concentrations of heavy metals varied among different food crops as illustrated in Figure 1. *Pisum sativum* and *Triticum aestivum* contained less Fe content compared to other food crops, but highest levels of Pb were found in *Triticum aestivum*. The highest concentration of Ni was detected in *Rumex obtusifolius*. The highest concentrations of Cr and Cd were noted in *Colocasia esculenta*.



Bio-accumulation factor (BAF) and metal pollution index (MPI)

The BAFs of various metals from soil to food crops are shown in Figure 2. BAF values > 1 suggested that metal was

effectively translocated from soil to plants. The range of BAF values varied for different metals. The trend of BAF (mean BAF) of selected heavy metals in differing food crops was in decreasing order of Cd > Cr > Ni > Fe > Pb.

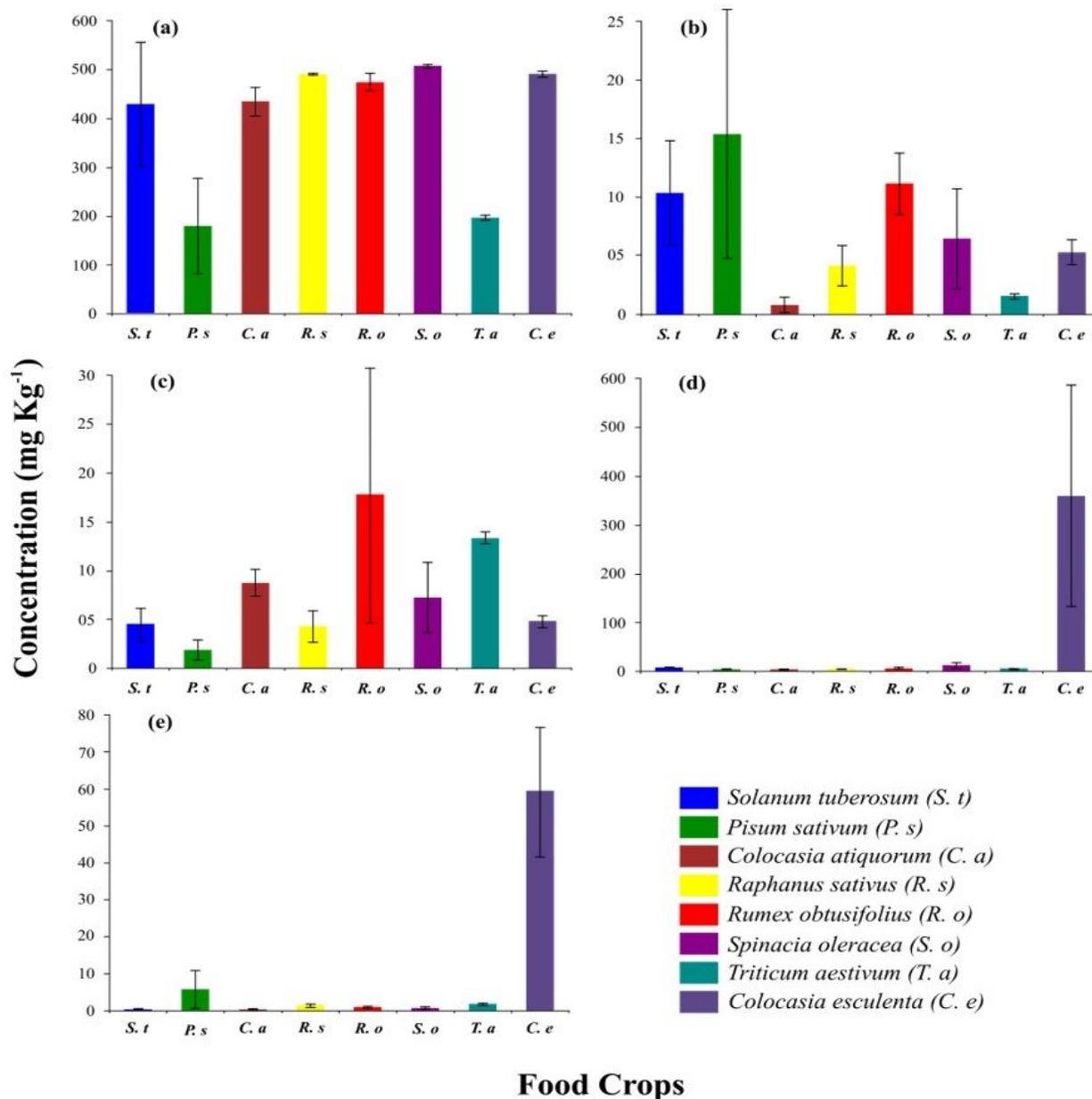


Figure 1: Accumulation of Fe (a), Pb (b), Ni (c), Cr (d), and Cd (e) in food crops with long-term wastewater irrigation. Investigated food crops are represented by different abbreviations and colours: *Solanum tuberosum* (S. t; blue), *Pisum sativum* (P. s, green), *Colocasia antiquorum* (C. a, brown), *Raphanus sativus* (R. s, yellow), *Rumex obtusifolius* (R. o, red), *Spinacia oleracea* (S. o, magenta), *Triticum aestivum* (T. a, cyan) and *Colocasia esculenta* (C. e, dark purple)



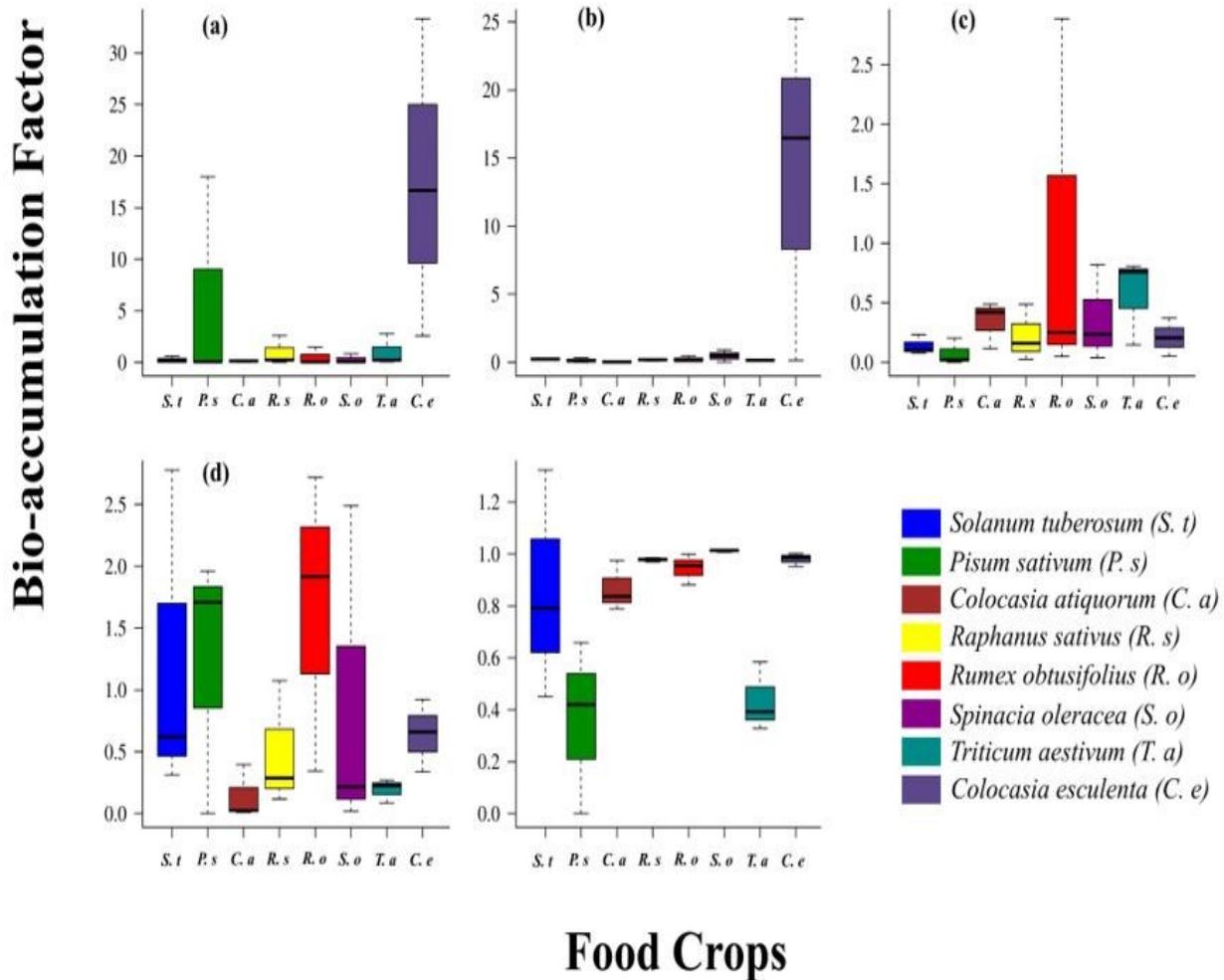


Figure 2: Comparison of the transfer factors of Cd (a), Cr (b), Pb (c), Ni (d) and Fe (e) for various vegetable crops collected from agricultural sites irrigated with industrial wastewater. Investigated food crops are represented by different abbreviations and colours: *Solanum tuberosum* (*S. t*; blue), *Pisum sativum* (*P. s*, green), *Colocasia antiquorum* (*C. a*, brown), *Raphanus sativus* (*R. s*, yellow), *Rumex obtusifolius* (*R. o*, red), *Spinacia oleracea* (*S. o*, magenta), *Triticum aestivum* (*T. a*, cyan) and *Colocasia esculenta* (*C. e*, dark purple). The boundaries of the central box are the 25th and 75th percentiles; the horizontal line is the median and whiskers of the boxplot represent the minimum and maximum values

The highest accumulation of Cd was found in *C. esculenta* (33) > followed by *P. sativum* (16), *T. aestivum* (2), *R. estivum* (2) and *R. obstifolius* (1.5); whereas all other food crops exhibited a value <1 which was below the permissible limit. In case of Cr, the highest BAF quantity was noted for *C. esculenta* (25), but all other crops showed BAF < 1. BAF values for Ni were within the safe limit < 1 except *R. obtusifolius* (2.8); whereas the BAF amounts for

Pb were >1 in *S. oleracea* (2), *S. tubersome* (2.7) and *P. sativum* (1.7-1.9). However, bioaccumulation of Fe was below the acceptable limit except for *S. tubersome* (1.3). The plants metal pollution index is depicted in (Figure 3). Spinach accumulated high levels of metals (13) while radish was the lowest (5). The order of metal pollution index was *S. oleracea* > *T. aestivum* > *P. sativum* > *R. obestosifolious* > *C. aesculenta* > *C. antiquorum* > *R. sativus*.



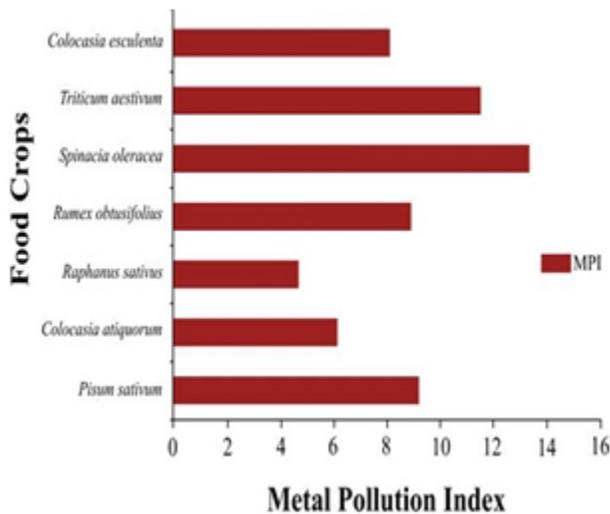


Figure 3: Metal pollution index of various crops in long-term wastewater irrigation

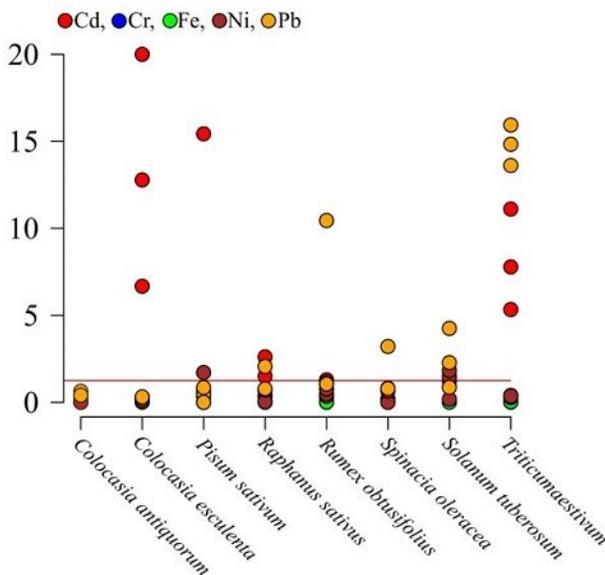


Figure 4: Health Risk Index calculated from the consumption of food crops by the population of the study area. The horizontal red line is the threshold limit for HRI (1). Points with different colors represent various heavy metals (Red; Cd, Blue; Cr, Green; Fe, Brown; Ni, and Orange; Pb)

Health risk index

In order to assess the health risk of heavy metal exposure through consumption of food crops irrigated with wastewater in the study region, HRI was determined (Figure 4). High HRI values of metals were observed in decreasing

order: Cd > Pb > Ni > Cr > Fe. Cd displayed the highest HRI level (20) followed by the second highest Pb (15.9). Out of 8 food crops, only *C. antiquorum* and *S. oleracea* exhibited HRI of Cd < 1. Whereas, lower HRI of Pb (HRI < 1) were found for only three food crops *C. antiquorum*, *C. esculenta* and *P. sativum*. In case of Ni, only *P. sativum* and *S. tuberosum* showed high risk (HRI > 1). HRIs of Cr and Fe were less than 1 for all investigated food crops. There was only one food crop, *C. antiquorum* that demonstrated lower risk for all heavy metals.

Discussion

Data indicated an absence of a correlation between soil metal and water contamination. In some cases, metals were not detected in water samples, but these elements were present in soil. It is of interest that Cd and Cr were not detected in wastewater samples at site 2, but high concentrations were observed in the soil (Table 1). These findings may be attributed to erratic changes in effluent types and metal concentrations at that location. In most cases, Cd concentration in soil and water exceeded the permissible limit (MEF, 2007; Tóth *et al.*, 2016; Vodyanitskii, 2016). Lead levels were also higher than the standard value at one site (Site 1). Although the concentrations of other heavy metals under study were below the permissible limits for soil, it should be noted that prolonged wastewater irrigation may generate metal accumulation in soils and subsequent bioaccumulation in edible parts of food crops. In comparison to other studies, most of the heavy metal concentrations in soil detected in the present study were higher than those reported in other countries (Khan *et al.*, 2008; Liu *et al.*, 2013; Alghobar and Suresha, 2017; Zeng *et al.*, 2018).

In all cases, the metal content in vegetable parts exceeded the safe limits described by FAO (2016). The major source for the higher than recommended heavy metals which exceed recommended EPA (2008)/FAO (2016) standard permissible limits metal content might be related to continuous wastewater irrigation (Li *et al.*, 2017; Chen *et al.*, 2018a). High concentrations of Fe present in both soil and edible parts of the crops may be attributed to the soil geology of the study area, which contained an Fe-rich rock bed. The second reason of metal bioaccumulation in the vegetable crops may also be related to the original seed used in cultivation where farmers in general obtain seed from their own farmland which was previously irrigated with wastewaters. Further, plant physiology may be responsible for differences in heavy metal concentrations in food crops from identical sites due to variability in uptake, absorption exclusion, translocation within plants and accumulation (Voutsas *et al.* 1996; Ashok *et al.*, 2009; Ahmad and Goni, 2010). Some specific metals may not originate exclusively



from wastewater. It is well-established that higher levels of Pb in plants may be due to metallic emissions from vehicles, from the agrochemical practices in the field area or release from industry (Ndiokwere, 1984).

Transfer of metal from soil to plant is a key mechanism for human exposure through the food chain. Higher transfer coefficients are characterized by greater ability of plants for metallic uptake while lower values exhibit stronger sorption of elements with soil colloids (Coutate, 1992). The BAF is a function of both the food crops and soil properties and indicative of transfer of soil nutrients and metals from soil to plant parts (Kachenko and Singh, 2006).

According to the standard concentrations reported by Wu *et al.* (2015), the crops under investigation in this investigation were highly contaminated with metals (Figure 3). Accumulation of heavy metals in food crops is influenced by a wide range of physicochemical and environmental factors including contaminants in soil, soil properties, crop type, climate and harvesting time (Bhargava *et al.*, 2012; Ali *et al.*, 2013). In the present investigation, natural occurrence of heavy metals (Alam *et al.*, 2003), lower retention in soil (Zurera *et al.* 1987) and continuous wastewater irrigation may constitute factors for the observed higher bioaccumulation in food crops. Data showed higher bioaccumulation of heavy metals in comparison to several other investigations (Sharma *et al.*, 2009; Qureshi *et al.*, 2016; Zeng *et al.*, 2018); clearly indicating that these sites in Pakistan appear to present as a higher health risk for humans. Indeed, higher BAF values for all metals, especially for Cd and Pb are a concern for human population in this study area. It should be noted that Cd and Pb are highly toxic metals and ingestion through food chain produced adverse health issues in human population (El Fadeli *et al.*, 2014).

The results showed that the health risk to the regional population is low with respect to exposure to Cr and Fe while consuming food crops, but the higher HRI for Cd and Pb suggest that the food crops cultivated in this region might not be safe for use with consequent serious health issues. Higher health risk of Cd and Pb effects were previously reported (Mahmood and Malik, 2014; Iqbal *et al.*, 2016; Chaoua *et al.*, 2018; Zeng *et al.*, 2018; Mehmood *et al.*, 2019). However, it is worthwhile noting that the risk level observed in the present study is several folds greater than the previous studies.

Conclusions

Concentrations of heavy metals including Cd, Cr, Pb and Fe that exceeded the permissible levels were observed in seven food crops collected from the study area. Cd displayed

the highest HRI level (20) followed by the second highest Pb (15.9). Out of 8 food crops, only *C. antiquorum* and *S. oleracea* exhibited HRI of Cd <1. Hence, it is suggested that farmers should not use wastewater for irrigation purposes without treatment. It is recommended that wastewater should be treated before it is used for irrigation to avoid health risk through wastewater irrigated crops.

Acknowledgements

We greatly acknowledge the contributions of Prof. Dr. Mark S. Coyne of University of Kentucky, USA to improve language of this manuscript.

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