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Risks of heavy metals accumulation in soil and crop irrigated with municipal wastewater in Mymensingh

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ABSTRACT

Two field experiments were conducted at Bangladesh Agricultural University campus and Gonsar Mor, Mashkanda, Mymensingh during January to May 2018 to evaluate the nutrients and heavy metal contents in soil and crop. Freshwater and wastewater irrigation and four varieties of rice: V1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58) were used with three replications. Analyses of wastewater for pH, EC, DO and heavy metals (Pb, Ni Cd and Cr), soil (before land preparation and after harvest) for pH, EC, OM, N, P, K and heavy metals (Pb, Ni, Cd, Cr), straw and grain for N, P, K and heavy metals were done using standard methods. The pH, EC, Pb, Ni, Cd and Cr content of wastewater was 7.4, 0.612 mS cm^{-1}, 4.3 ppm, 0.09 ppm, 0.40 ppm and 1.80 ppm, respectively. The values of pH, EC, Pb and Ni were within the permissible limit whereas Cd and Cr content in wastewater exceeded the permissible limit for irrigation. OM, N, P and K contents were higher in wastewater irrigated soil and average Cd (0.014 ppm) and Cr (44.81 ppm) content of soil under wastewater irrigation was within the permissible limit. Nutrients and heavy metals (Cd: 0.001 to 0.006 ppm; Cr: 0.49 to 0.59 ppm) contents of straw and grain were also higher under wastewater irrigation. Bioaccumulation and translocation factors for Cd ranged from 0.103 to 0.431 and 0.097 to 0.865 and for Cr ranged from 0.011 to 0.013 and 0.010 to 0.012, respectively. Significant variations were found among four varieties regarding nutrient contents of straw as well as bioaccumulation and translocation of Cd in grain. Bioaccumulation factor was <1, indicating absorption only, not accumulation of Cd and Cr in straw or grain, and municipal wastewater can safely be used as an alternate water source for irrigation.

Introduction

Wastewater is a byproduct of domestic, industrial, commercial or agricultural activities that is mainly composed of water (>95%) and solid portion (contains 40-50% organics, 30-40% inert materials, 10-15% bio-resistant organics and 5-8% miscellaneous substances on oven dry weight basis) (Antil *et al.,* 2008). Although not legally permitted in most countries, the use of untreated wastewater for crop irrigation has been practiced in many countries all over the world due to the shortage of good quality water (Zhang and Shen, 2017; Jaramillo and Restrepo, 2017; Khalid *et al*., 2017). Demand for water, especially in agriculture is continuously increasing worldwide and many countries are currently facing water shortages or forecasting its future scarcity. The proportion of water available for agriculture is projected to decline to 62% worldwide and 73% in developing countries by 2020 (Barker, 2000). Wise use of supplemental water resources (e.g., wastewater) is possible solutions to the global problems of water shortage. Approximately 20 million ha land was irrigated with wastewater in fifty countries worldwide (Abaidoo *et al*., 2010; Jiménez, 2006). The use of wastewater for crop irrigation has further been increased in recent years. Farmers in developing countries are using raw sewage wastewater to irrigate nearly 49 million acres (20 million hectares) of cropland.

In Bangladesh, there is 8.3 Mha (million hectare) of cultivable land of which 7.0 Mha are potential area for irrigation (Mojid *et al*., 2010). However, only 4.48 Mha is covered by irrigation systems and 2.52 Mha remains out of irrigation due to shortage of water (BBS, 2004). Shortage of surface and underground water could be partially overcame by the use of wastewater. The Economic and Social Commission for Asia and the Pacific reported an annual production of 725 Mm^3 of wastewater from the urban areas of Bangladesh. In Mymensingh municipality area, 63% wastewater is produced from domestic sources, 10% from markets, 10% from small factories and industries, 9% from offices and 8% from other sources (Parvez *et al*., 2007). At the advent of increased scarcity of freshwater in Bangladesh, the wastewater reuse for the purpose of irrigation may

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have a significant contribution towards water conservation as well as food production. However, wastewater carries appreciable amounts of trace toxic metals and elevates the levels of heavy metals in the receiving soils (Yadav *et al*., 2002). Toxic metal accumulation in organic matter in soils leads to degradation of soil health and contamination of food chain mainly through the crops grown on such soils (Rattan *et al*., 2002) and may pose serious risks to the human and environmental health (Khan *et al*., 2008).

In Bangladesh, although a few peri-urban farmers have started irrigating their crops with this water, it is not yet well documented. A number of studies have reported the deposition of metals in soil, crops and vegetables grown in the vicinity of industrial areas around the world as well as in Bangladesh (Balkhair and Ashraf, 2016; Mottalib *et al*., 2016; Goni *et al*., 2014). However, no work has been done to study the rate of metal accumulation in rice cultivated on municipal wastewater irrigated field. Therefore, it is important to assess heavy metal concentrations in soil and crop to ensure safe food production and prevent environmental and public health risks. The objectives of this study were: to examine the chemical properties of municipal wastewater used in this study, to assess the accumulation of nutrient and heavy metals in soil and above ground plant parts and to calculate the translocation of heavy metals from soil to plant parts to determine the relative uptake of heavy metals by the plants with respect to soil.

Materials and Methods

Site selection

The wastewater of Mymensingh Municipal area is released at Noyapara outlet and passing through channel from Mashkanda to Chorar Beel area. From January to March in every year, the farmers of these peri-urban areas have no other alternative to use municipal wastewater as irrigation for rice cultivation due to shortage of water. Gonsar Mor, Mashkanda, the nearest wastewater releasing point was therefore selected for this study. However, in Gonsar Mor area, there is no availability of fresh water for irrigation purpose due to absence of deep tube well. Therefore, Field Laboratory of Department of Environmental Science, BAU, Mymensingh was selected for setting an experiment under fresh water irrigation to compare the results with that of wastewater irrigation.

Environmental Science Field Laboratory is located at 24.75°N latitude and 90.50°E longitude and the rice field of Gonsar Mor, Mashkanda is located at 24.45°N latitude and 90.22°E longitude. The both sites belong to the Sonatola soil series of non-calcareous dark grey flood plain soil under the Old Brahmaputra Alluvial Tract and are characterized by moderately high temperature and heavy rainfall during the Kharif season (April-September) and scanty rainfall with moderately low temperature during Rabi season (October- March).

Experimental design and layout

The study was carried out during the period from 16 January to 15 May 2018. The experiment was laid out in a Randomized Complete Block Design (RCBD) with three replications. Irrigation water was used as main treatment in this study. Treatments were: T_1: Fresh water irrigation and T_2: Municipal wastewater irrigation. Varietal effects under irrigation were also investigated in this study. Varieties were: BINA dhan11 (V_1); BRRI dhan28 (V_2); BRRI dhan29 (V_3) and BRRI dhan58 (V_4). The total numbers of unit plots were 24. The area of each plots was 20 square meter (5m × 4 m). The treatment combinations were randomly distributed to unit plots.

Crop establishment

The land was first opened on 16 January 2018. In municipal wastewater irrigated area, farmers do not use the recommended doses of fertilizer due to high nutrient content of soil. Only 300 g MOP was applied per plot during final land preparation and after 60 days of transplanting, 200g urea was applied in each plot. In fresh water irrigated land, fertilizer was applied following the recommended doses. Seedlings were transplanted in well prepared land maintaining three seedlings per hill with 15 cm × 20 cm row and hill spacing on 21 January 2018. Necessary intercultural operations were done for maintaining the normal growth and development of the crop.

Collection and analysis of wastewater

Wastewater was collected from the drain of Gonsar Mor rice field for the analysis of pH, EC, DO and heavy metals (Pb, Ni, Cd and Cr). The pH, DO and EC were measured by pH meter (model: pHep, HANNA Instruments), DO meter (model:YK-22DO) and EC meter (model:3251, COND/TEMP/METER), respectively at the Laboratory of Department of Environmental Science, BAU, Mymensingh. Heavy metals (Pb, Ni, Cd, Cr) were analyzed at Bangladesh Agricultural Research Institute (BARI), Gazipur by Atomic Absorption Spectrophotometer (AAS) (Varian spectra AA55B, Australia).

Collection and analysis of soil samples

The soil sample of fresh water irrigated land and municipal wastewater irrigated land was collected from 0-15 cm depth before final land preparation and after harvest for the analysis of total nitrogen (N), available phosphorus (P), exchangeable potassium (K) and heavy metals (Pb, Ni, Cd and Cr) content. The N content was estimated by micro-Kjeldahl method, total P was extracted from the soil following the method of Olsen *et al.* (1954), total K was determined using flame photometer (Black, 1965) at the Laboratory of Department of Soil Science, BAU, Mymensingh. Pb, Ni, Cd and Cr were analyzed at BARI, Gazipur.

Collection and analysis of plant sample

Fully mature rice was harvested on 15 May 2018. Above ground plant parts (straw and rice) were used for the analysis of total N, available P and exchangeable K, Cd and Cr.

Assessment of bioaccumulation and translocation of heavy metals

Bio-accumulation factor (BAF)

The BAF of heavy metals in the plant samples was calculated as:

$BAF = C_{plant}/ C_{soil}$

Where, C_{plant} is the concentration of the element in the plant, and C_{soil} is the concentration of same element in the soil on dry weight basis.

Translocation factor

Translocation of heavy metals from the soil to the edible parts of the crop was determined by the accumulation factor (AF) (Li *et al*., 2012).

All the experimental data were statistically analyzed using MS Excel 2013 and MiniTAB 17.0 data processing software.

Results and Discussion

Chemical properties of wastewater

The pH of wastewater used in experimental site was 7.4 (Table 1). The standard pH of irrigation water is 6.0-8.5 (Ayers and Wescot, 1985). Therefore, pH value of studied wastewater was within the acceptable limit and not harmful for crops. The EC value of wastewater was 0.612 mS cm^{-1} (Table 1). The standard EC value of irrigation water is 1.2 mS cm^{-1} (Ayers and Wescot, 1985). Higher EC value reflected the higher amount of salt concentration which affected irrigation water quality related to salanity hazard (Agarwal *et al*., 1982). The Dissolved Oxygen (DO) of wastewater was 3.00 mgL^{-1} (Table 1). The maximum limit of DO value for irrigation water is 10 mgL^{-1}. Lower values of DO cause root injury as well as change plant metabolism, both of which are needed for a healthy plant (Drew, 1997).

Table 1. Chemical properties of wastewater

Parameter	Values
pH	7.4
EC (mS cm^{-1})	0.612
DO (mgL^{-1})	3.00
Pb (ppm)	4.3
Ni (ppm)	0.09
Cd (ppm)	0.40
Cr (ppm)	1.80

The Pb, Ni, Cd and Cr content of wastewater was 4.3, 0.09, 0.40 and 1.80 ppm, respectively (Table 1). According to WHO (2007), the values of Pb and Ni for irrigation purpose were within the permissible limit and it is 5.00 and 0.20 ppm, respectively. On the other hand, the values of Cd and Cr exceeded the permissible limit and it is 0.01 and 0.1 ppm, respectively (WHO, 2007).

Effects of wastewater irrigation on nutrient and heavy metal content of soil

Organic matter content (OM)

The average values of soil OM content in freshwater irrigated field were 1.41%, 1.10%, 1.41%, and 1.35% in V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 1a). On the other hand, OM content in wastewater irrigated field were 2.34%, 2.26%, 2.40%, and 2.07% V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 1a). OM content was higher in wastewater irrigated field and the lower in freshwater irrigated field. It might be happened due to the presence of various organic substances in wastewater compared to fresh water**.** Parvez (2000) found that irrigations with raw effluent increased the soil organic matter by 1.8%. There was no varietal effect on organic matter content of soil.

Total nitrogen (N) content

The average values of total nitrogen content in freshwater irrigated field were 0.136%, 0.120%, 0.136% and 0.125%, in V_1 (BINA dhan11), V_2 (BRRI dhan28),V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 1b). On the other hand, the values of total nitrogen content in wastewater irrigated field were 0.223%, 0.210%, 0.232% and 0.226% in V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 1b). The results indicate that irrigation by wastewater increase the N content of soil compared to the normal water irrigation. N content increased by 63.97%, 75%, 69.34% and 80.8% in V_1, V_2, V_3 and V_4 plot by wastewater irrigation. Kiziloglu *et al*. (2008) found an increase in N content in soil by the application of wastewater irrigation.

Available phosphorous (P) content

The average values of P content in freshwater irrigated field were 7.51 ppm, 7.31 ppm, 7.46 ppm and 7.8 ppm in V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 1c). On the other hand, the values of P content in wastewater irrigated field were 40.85 ppm, 47.26 ppm, 43.95 ppm and 44.48 ppm in V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 1c). The P content of wastewater irrigated land was more than 5 times higher compared to fresh water irrigated land. It might be happened due to higher P content in wastewater. Taberi *et al*. (2009) assessed the impact of municipal wastewater irrigation on soil properties and elder pine trees and reported that P content was increased due to use of wastewater irrigation and it has no harmful effect.

Exchangeable potassium (K) content

The average values of exchangeable K ranged from 37.53 to 38.96 ppm in the freshwater irrigated field and it was 38.96, 37.53, 37.66 and 40.27 ppm, respectively in variety V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58) planted plot (Fig. 1d). On the other hand, the values of exchangeable K of wastewater irrigated land were 84.45, 92.92, 84.21 and 99.94 ppm in V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58) containing plot, respectively (Fig. 1d). It was found that by using wastewater irrigation, average K content was increased by 116.76%, 147.58%, 123.80% and 148.17% in V_1, V_2, V_3 and V_4 plot, respectively. Galavi *et al.* (2010) assessed the effect of treated wastewater on soil properties and found exchangeable K content ranged from 181-189 ppm.

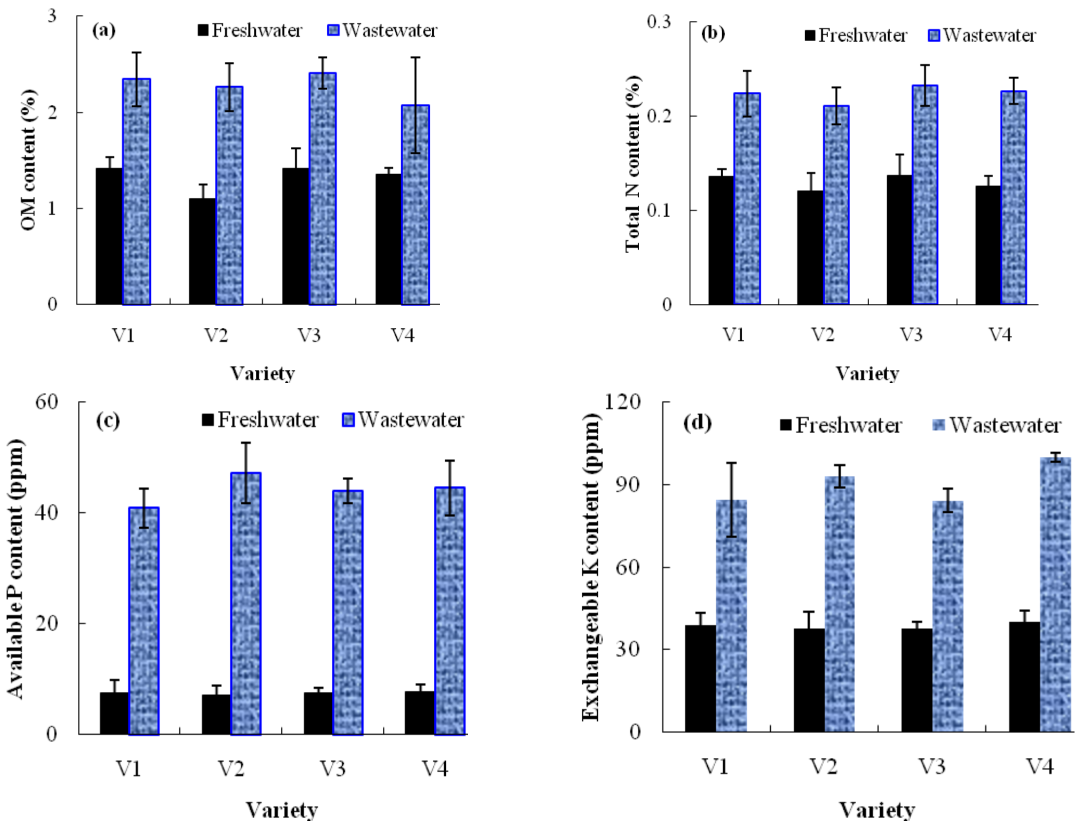

Fig. 1. Effects of wastewater irrigation on: (a) Organic matter (OM) content; (b) Nitrogen (N) content; (c) Phosphorous (P) content and (d) Potassium (K) content of soil

Heavy metal content of soil

The values of Cd and Cr in wastewater exceeded the permissible limit (Table 1). Therefore, only Cd and Cr contents were investigated in soil and plant (straw and grain) samples.

Cadmium (Cd)

The average Cd content in soil were 0.01267, 0.01267, 0.01367 and 0.01333 ppm in variety V_1 (BINAdhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 2a). The highest value of Cd (0.0136 ppm) was found in variety V_3 and the lowest value (0.0126) was found in variety V_1 and V_2. According to WHO (1996), the maximum permissible limit of Cd in soil is 0.8 ppm. All the values were below the permissible limit indicating that wastewater irrigation has no negative effect on Cd content in soil. Yadav *et al.* (2013) assessed the level of different heavy metals like Cd, Ni, Pb in vegetables irrigated with wastewater and found the value of Cd in soil (0.028 to 0.036) was within the permissible limit.

Chromium (Cr)

The average Cr content of soil were 44.63, 45.63, 45.2 and 43.8 ppm in variety V_1 (BINAdhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 2b). The highest value of Cd was found in variety V_2 (45.63 ppm) and the lowest value was found in variety V_4 (43.8 ppm). Acording to WHO (1996), the permissible limit of Cr in soil was 100 ppm. The Cr content of soil under wastewater irrigation was within permissible limit. Satpathy *et al.* (2014) assessed the amount of Cr in soil irrigated with untreated water and found Cr ranged from 1.3 to 7.8 ppm.

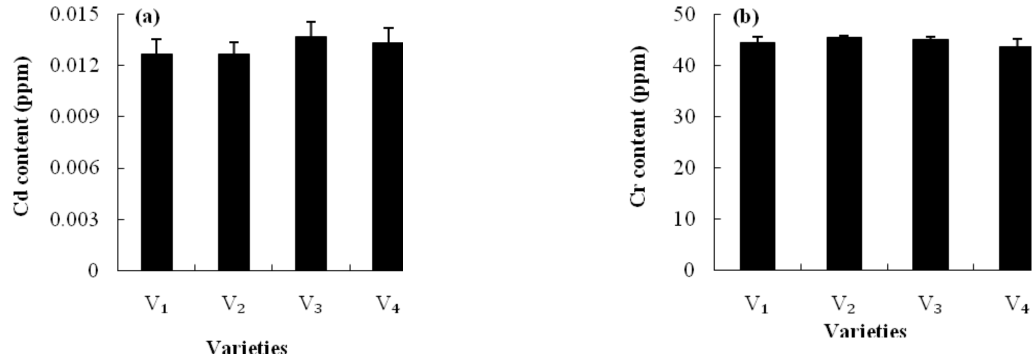

Fig. 2. Effects of wastewater irrigation on (a) Cadmium (Cd) content and (b) Chromium (Cr) content of soil

Effects of wastewater on nutrient and heavy metal content of straw and grain

Nitrogen content

The total nitrogen content of straw in freshwater irrigated field were 0.308% in V_1 (BINA dhan11), 0.352% in V_2 (BRRI dhan28), 0.291% in V_3 (BRRI dhan29) and 0.302% in V_4 (BRRI dhan58) (Fig. 3a). On the other hand, the values of total N content of straw in wastewater irrigated field were 0.464% in V_1 (BINA dhan11), 0.280% in V_2 (BRRI dhan28), 0.470% in V_3 (BRRI dhan29) and 0.4368% in V_4 (BRRI dhan58) (Fig. 3a). The highest total N content was found in V_3 (BRRI dhan29) variety and the lowest was found in V_1 (BRRI dhan28) variety under wastewater irrigation. Day *et al*. (1979) reported that N concentration in shoots were higher when grown with wastewater and found that N recovery of plants with wastewater irrigation was higher than the N recovery of plant grown with freshwater.

Phosphorous (P) content

The average values of P content of straw in freshwater irrigated field were 0.134%, 0.152%, 0.118% and 0.163% in V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 3b). The values of P content of straw in wastewater irrigated field were 0.089% in V_1 (BINA dhan11), 0.133% in V_2 (BRRI dhan28), 0.108% in V_3 (BRRI dhan29) and 0.184% in V_4 (BRRI dhan58) (Fig. 3b). The highest and lowest value was found in V_4 and V_1, respectively under wastewater irrigation. P recovery efficiency was higher in freshwater irrigated plot compared to wastewater irrigated plot. Rusan *et al*. (2006) reported that P concentration in barley shoot increased significantly as years of wastewater irrigation increased. It increased from 0.18% in the control treatment to 0.28% in the 10 years irrigated with wastewater.

Potassium (K) content

The K content of straw in freshwater irrigated field were 2.20% in V_1 (BINA dhan11), 2.45% in V_2 (BRRI dhan28), 1.8% in V_3 (BRRI dhan29) and 2.52% in V_4 (BRRI dhan58) (Fig. 3c). The K content of straw in wastewater irrigated field were 2.22%, 2.75%, 2.03% and 2.37% in V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 3c). The highest value was found in wastewater irrigated field (V_2: BRRI dhan28) and the lowest value was found in freshwater irrigated field (V_3: BRRI dhan29). Mohammad and Mazahreh (2003) reported an increase in K uptake by the plants irrigated with treated wastewater. However, there was no considerable variation in K content between freshwater and wastewater irrigated plot in this study.

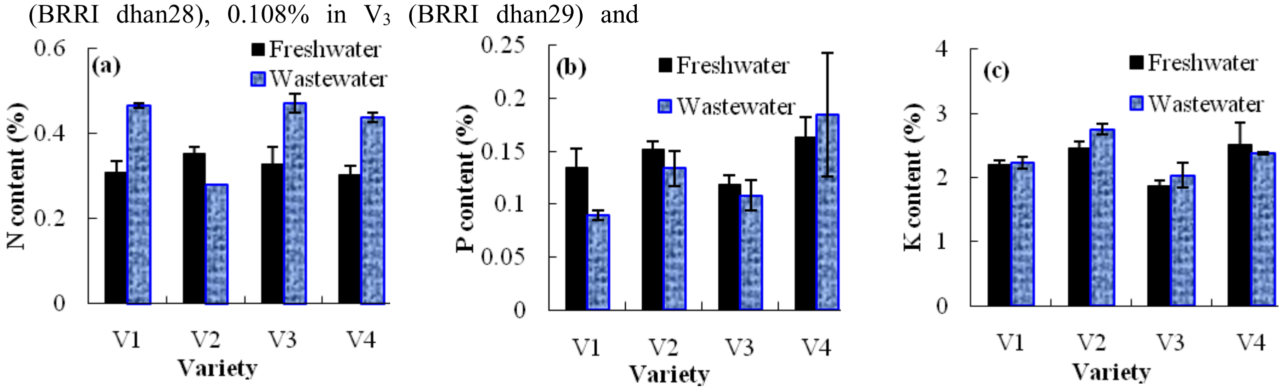

Fig. 3. Effects of wastewater irrigation on a) Nitrogen (N) content; (b) Phosphorous (P) content and (c) Potassium (K) content of straw

Heavy metal content in straw and grain

Cadmium (Cd)

The Cd contents of straw were 0.004, 0.002, 0.003 and 0.001 ppm in variety V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 4a). The figure represents that the highest value of Cd was found in variety V_1 and the lowest value was found in variety V_4. According to WHO (1996), the permissible limit of Cd for plants is 0.02 ppm. All the values of Cd in straw were within the permissible limit. The average Cd contents of grain were 0.0013, 0.0036, 0.0030 and 0.0057 ppm in variety V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 4a). The highest value was found in variety V_4 and the lowest value was found in variety V_1. The results revealed that Cd uptake ability of BRRI dhan58 was higher than other variety. According to WHO (1996), the permissible limit of Cd for plants is 0.02 ppm. All the results were within the permissible limit. Naser *et al.* (2009) studied the levels of heavy metals like Cd, Ni in tomato grown in wastewater polluted areas and found that Cd and Ni content was increased and ranged from 0.630-1.303 ppm and 2.031-4.957 ppm, respectively.

Chromium (Cr)

The average Cr contents of straw were 0.543, 0.520, 0.490 and 0.536 ppm in variety V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 4b). The permissible limit of Cr in straw was 1.30 ppm (WHO, 1996). The Cr content of straw in all varieties under wastewater irrigation was within permissible limit.

The average Cr contents of grain were 0.593, 0.540, 0.553 and 0.500 ppm in variety V_1 (BINA dhan11), V_2 (BRRI dhan28), V_3 (BRRI dhan29) and V_4 (BRRI dhan58), respectively (Fig. 4b). The highest value of Cr was found in variety V_1 and the lowest value was found in variety V_4. Cr content of grain was higher in V_1, V_2 and V_3 compared to Cr content of straw. However, all values were within permissible limit (WHO, 1996). Hamid (2012) assessed vegetables irrigated with wastewater and found that Cr content was 13.2 ppm which was higher than permissible limit (2.4 ppm).

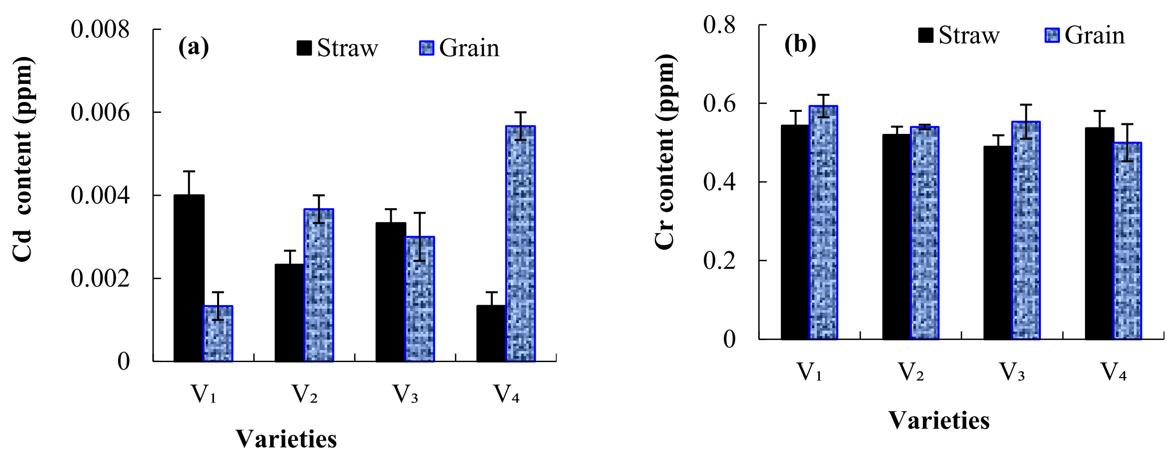

Fig. 4. Effects of wastewater irrigation on (a) Cadmium (Cd) content and (b) Chromium (Cr) content of above ground plant parts

Bioaccumulation of heavy metals

Bioaccumulation factors (BAFs) for the heavy metals (transfer from soils to rice) are shown in Fig. 5(a). The BAF values of Cd ranged from 0.1 to 0.4 and that of for Cr was 0.011 to 0.0113. BAF values were higher for Cd whereas relatively low BAF values were found for Cr. Soil-to-plant transfer is one of the key processes of human exposure to toxic heavy metals through the food chain. When BAF < 1 or BAF = 1, it denotes that the plant only absorbs the heavy metal but does not accumulate. When BAF >1, this indicates that plant accumulates the heavy metals. BAF values of Cd and Cr were less than 1 in the rice grain, which indicates that plants only absorb the heavy metals. Among the variety, V_4 had the higher absorption ability of Cd compared to other varieties.

Translocation of heavy metals

The average values of translocation factor (TF) of metals from soil to plant were found to be in order of Cd (0.097 to 0.865)> Cr (0.0108 to 0.0122) (Fig. 5b). The result indicated the bioavailability of heavy metals in investigated soils. The higher the TF values, the more mobile/available the metals (Khan *et al.,* 2008). Transfer of Cd was more from soil to above ground plant parts which is known to be relatively mobile in plants than Cr. The mobility of Cd was higher in V_1 variety compared to others. Satpathy *et al.* (2014) assessed the translocation of heavy metals irrigated with untreated water and found the heavy metals in the order of Zn (0.4 to 0.9) >Mn (0.3 to 0.7) > Cd (0.3 to 0.6) >Pb (0.2 to 0.4) >Cr (0.2 to 0.3) > Cu (0.09 to 0.2).

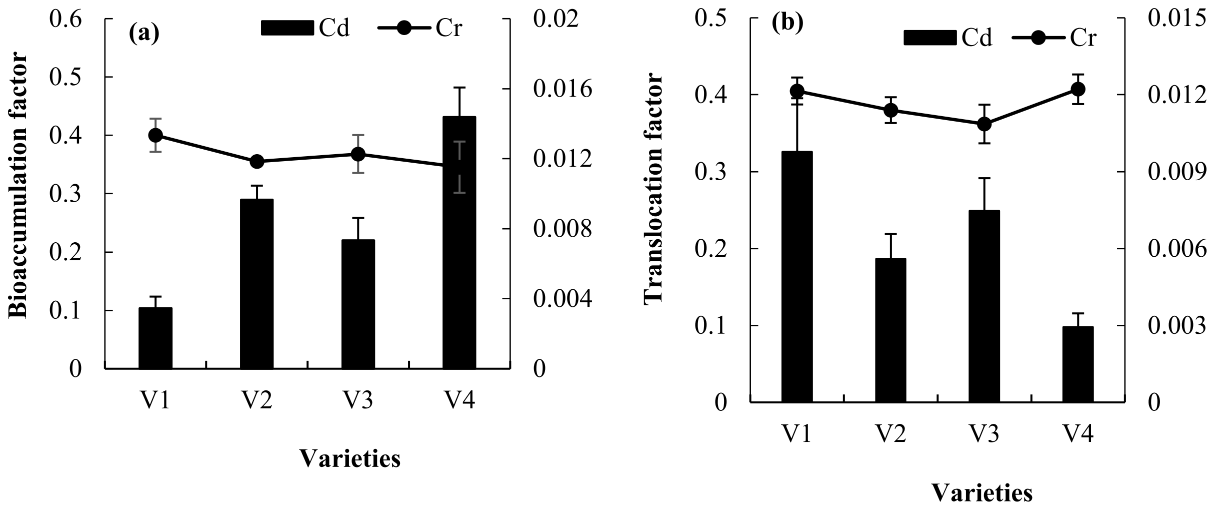

Fig. 5. Effects of wastewater irrigation on (a) Bioaccumulation and (b) Translocation of heavy metals

Conclusion

Nutrient content of soil and above ground plant parts was higher in wastewater irrigated field compared to freshwater irrigated field. Heavy metals (Cd and Cr) were also higher in straw and grain grown under wastewater irrigation. However, the values of Cd and Cr were within the permissible limit. From this study, it can be concluded that municipal wastewater can be safely used as an alternative water source for the irrigation of rice field, although monitoring will be required to determine the effects with regard to soil and crop contamination by other heavy metals and harmful microorganisms (especially *E. coli*) as well as other potential health concerns.

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