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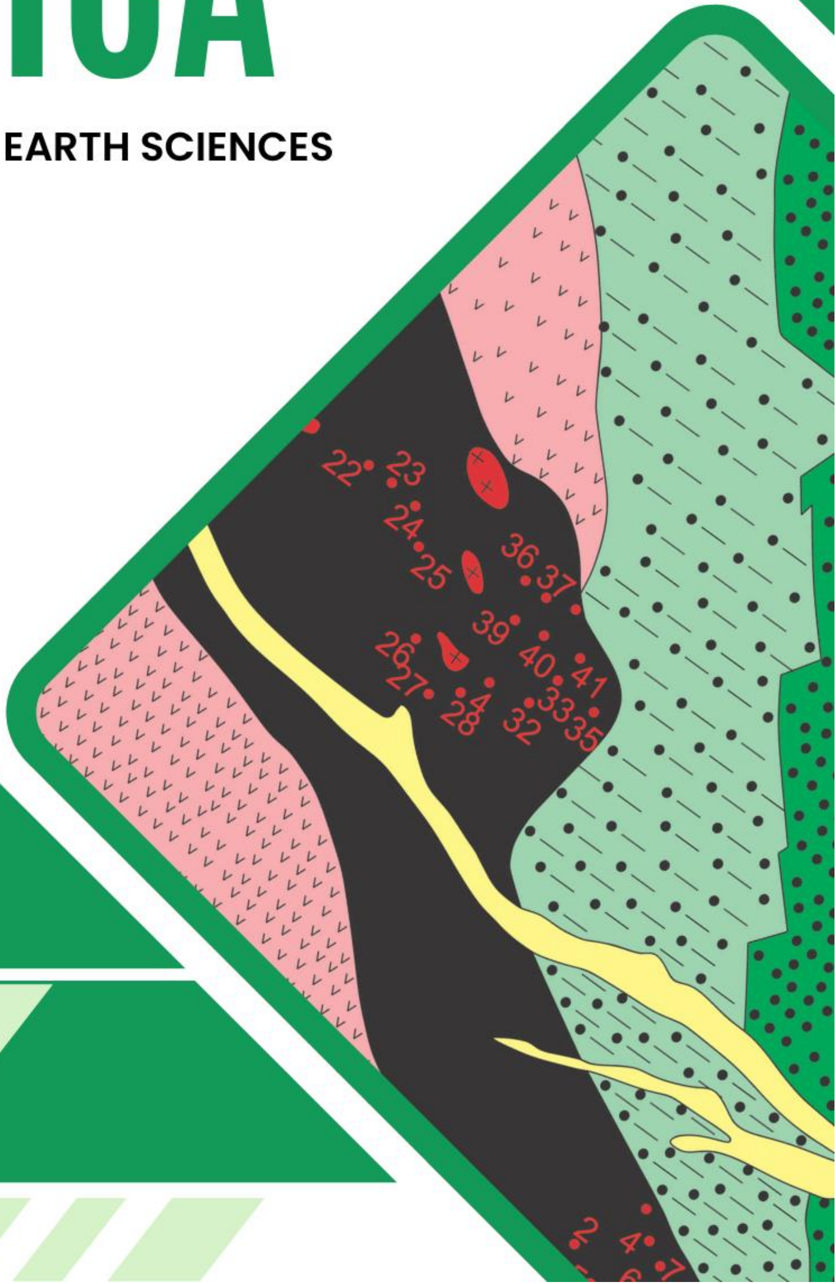

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Continuous Use of Arsenic Contaminated Irrigation Water On Rice Paddies: A Future Threat To Sustainable Agriculture

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Abstract

Research work was conducted in Wazirabad District to investigate the concentrations of arsenic in irrigation water, soil and plant parts and to assess the future threat to the economy. The level of arsenic in irrigation water was very much above the WHO permissible limit (0.01 mg/l) for drinking water but within the FAO permissible limit of 0.10 mg/l for irrigation water. Results indicate that the arsenic is transferred from irrigation water and paddy soil to various parts of rice plants. In none of the studied samples the concentration of arsenic in soil exceeds the permissible limit of 25 mg/Kg, thus the exposure of the people living in the study area to arsenic is not a matter of an immediate concern at a moment but future projections of soil arsenic (52.5 mg/Kg in 2035 and 147 mg/Kg in 2045) cannot be ignored as it possess a serious future threats to human health and economy of the country. To avoid the future disturbance due to arsenic contamination, there is a need to take appropriate steps such as development of new irrigation strategies and continuous monitoring of irrigation water, soil and grain.

Keywords: Arsenic, water, paddy soil, rice, regression, correlation.

Introduction

Arsenic (As), is a Human toxin found in ground water of more than 70 countries and it is affecting more than 1.5 billion people worldwide. Its elevated levels in environment device an imperative public health hazards (Ravenscroft, Brammer, and Richards 2009). Arsenic rich groundwater is used not only use for domestic purpose but also for crop irrigation, particularly for the paddy rice (Meharg and Rahman (2003) Rice is especially a fascinating case as it is greatly more effective in collection of arsenic as compared to different grains, for example, wheat grain, with its arsenic translocation consider frequently approaching solidarity (Bhattacharya et al. 2009). Addition of arsenic from one trophic level to different depends on the aggregate As fixation as well as to a great extent relies upon its bioavailability. Irrigation water with elevated amounts of As may result in area corruption regarding yield crop production (loss of yield) and food safety (food chain contamination). Long-term utilization of As-contaminated irrigation water could bring about As accumulation in the soil. Arsenic contaminated crops causes health risk for human(Brammer and Ravenscroft 2009)

Anaerobic conditions favor arsenic in the form of arsenite, in flooded conditions which is a normal practice in rice producing countries (Brammer 2008). Arsenite in groundwater and sediment is the most readily available form of arsenical to plant roots, where bioaccumulation goes from roots to stem, leaf, and lastly, grain.

The safe limit for a sixty-kilogram adult who consumes about 450 grams of rice with a concentration of 11µg/L arsenic/day and who drinks 4 liters of water with a concentration of 10 µg/L of arsenic/day ingests 130 µg of arsenic daily. This suggests that the provisional tolerable weekly intake is 15 µg of As a week/kilogram of body weight(Agusa et al. 2009). This number significantly multiplies when the allowed arsenic concentration of safe drinking water in Pakistan is 50 µg/L, compared to 10 µg/L, which is WHO limit (Organization 2005)

In South East Asia, arsenic concentrations in raw rice grains were measured up to 1.8 µg/L where the local groundwater concentrations reached 4,700 µg/L (Agusa et al. 2009). Rice, one of the most arsenic absorbent crops, is among the principle irrigated crop in Pakistan, and serves as the staple diet, alongside wheat and vegetables. Few years before, Pakistan superseded USA after Vietnam as the world's third largest rice exporter by volume(Dawe and Slayton 2010). Its export 30% of the world paddy rice. According to report 2.9 million tons of Basmati rice is produces annually in Pakistan Geochemistry of the provinces of Pakistan suggest that the recent deltaic and alluvial sediments of the aquifer of the Indus plain (Punjab and Sindh) are more susceptible to Arsenic (PCRWR, 2006). Pakistan Council for Research in Water Resources (PCRWR) has reported Arsenic contamination (10-200µg/L) in groundwater of different areas of Punjab i.e Lahore, Gujranwalan, Kasur, Shikhopora, Bahawalpur (Hagras 2013). Different studies suggest that in terms of drinking water (10–200 µg/L), most areas of Pakistan are exposed to high level of arsenic. In Pakistan, Khairpur and Nagarparkar study reveals the arsenic contamination in irrigation water (Brahman et al. 2013)

From an export and GDP standpoint, Pakistan's economy heavily relies on rice. Rice is currently accumulating extreme amounts of arsenic from ground water. The rice cultivation is mostly depended on underground water in Pakistan particularly in dry season, since the sources of surface water like river, dam, pond etc. of these regions becomes dry throughout the season. Long term use of arsenic contaminated underground water in irrigation may results in the increase of its concentration in agricultural soil and eventually in crop plants(Ullah, Malik, and Qadir 2009);(Rahman, Hasegawa, Rahman, Rahman, et al. 2007; Rahman, Hasegawa, Rahman, Islam, et al. 2007)

Survey on paddy soil throughout Bangladesh showed that arsenic concentrations were higher in agricultural soils of those areas where shallow tube wells (STWs) have been in operation for longer period of time and arsenic contaminated underground water from those STWs have been irrigated to the crop fields (Meharg and Rahman 2003).(Onken and Hossner 1995) reported that plants grown in soil treated with arsenic had higher rate of arsenic uptake compared to those grown in untreated soil. (Abedin, Feldmann, and Meharg 2002) and Rahman, Hasegawa et al. 2007) also reported elevated content of arsenic in tissues of rice when the plant was grown in soils contaminated with higher concentrations of arsenic. Since the irrigation water for rice paddies creates a large bioaccumulation of arsenic, any rice exported from Pakistan would have unsafe levels of arsenic for ingestion. PCRWR reported that Punjab and Sindh are more suitable for arsenic contamination of groundwater and these two provinces are the largest exporter of rice. Studies have been reported on drinking ground water arsenic contamination in Punjab (PCRWR, 2006) but to date no study conducted to know the concentrations of arsenic in irrigation water and its impacts on soils and rice.

Therefore, the main objective of this study was to investigate the concentrations of As in irrigation water, distribution, and mobility of As in the soil of a rice paddy and in order to evaluate the influence of irrigation with As-rich groundwater on soil and plant and to assess the threat to our economy in coming years using the data of present study. This study would help to evaluate the transfer of arsenic from irrigation water and paddy soil to the rice plant.

Materials and Methods

Study Area

Wazirabad is well-known for its high-quality export rice and ground water is the source of irrigation in the area. USDA has reported presence of arsenic in export quality Basmati rice of Pakistan (Zavala and Duxbury 2008). No previous literature has been reported from the present study site regarding arsenic contamination in irrigation water. Wazirabad is situated on the bank of the Chenab River nearly 100 kilometers north of Lahore on the Grand Trunk Road. Samples of groundwater, soil, and rice were taken from agricultural land of five different villages' i.e, Dohnkal (Site A), Begowali (site B), Mansoorwali (Site C), Sohdra (Site D) and Ojlankalan (Site E) of Wazirabad (Fig.1).

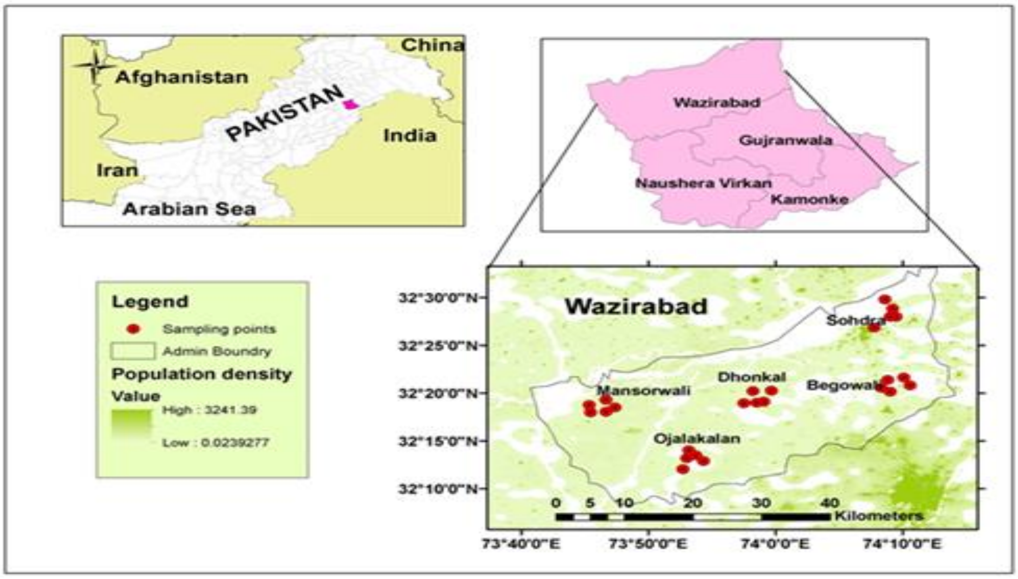

Figure 1 Map showing study Area.

Sampling

Five sampling locations were selected covering area as they were directed along East, West, North and South sites of study area. From each site five shallow tube wells (depth ranged from 60 to 150 feet) were selected as sampling points. 25 irrigation water samples, 5 composite soil samples (0-25 cm) and 5 rice samples (composite) simultaneously were collected from each tube well and its surrounding cultivated land during the month of March 2014. Water samples were collected in pre-washed polyethylene bottles with concentrated nitric acid (1:1. Samples were preserved with nitric acid (0.1% v/v) and were collected after 10-20 minutes after pumping and were filtered through 0.45 107 Millipore filter paper. About 100 ml of water was collected from each TW and preserved in the refrigerator adding 10 ml 2M hydrochloric acid in them.

Soil Samples (20-25g) were collected with the help of 37.5 mm diameter PVC pipe sampler (about 750 mm in height), sun dried immediately after collection, tagged properly, kept air tied in polyethylene bag and transported to the laboratory for analysis. In fume cupboard, on plastic trays, soil samples were spread and air dried for 8 days at room temperature. The dried soil was manually grinded and then sieved through nylon sieve < 100µm mesh (Chatterjee, Das, and Chakraborti 1993).

The rice samples (each sample consisted of five sub samples with a distance of approximately 300 cm between each other). The samples from all the villages (25 sites agricultural lands) were sampled by hands covering with vinyl gloves, carefully placed them in individual polyethylene bags, stored in a cold box at 4 ºC and transported to laboratory and kept them on 4 ºC until further treatment. The root, stem, leaf and grain parts of each sample were separated with quartz knife (Queirolo et al. 2000). All parts were dried in open air under diffused sun light for 24 hours, followed by drying in an oven at 50 ºC for complete dryness. Manually ground them to a fine powder with a micro hammer Mill No 61171 and passed through a 30-mesh sieve. Samples for As analysis were prepared by acid digestion. Hydride generator AAS was used for the analysis of total arsenic in all of the water and soil samples. The detection limit of arsenic was is 05 µg/L. (Roychowdhury 2008).

Total arsenic in rice plants were analyzed by Ion hydride generator atomic absorption spectroscopy. Plants samples were prepared by digestion method. 5ml HNO_3 was added in 0.5-gram plant sample and then kept overnight. Samples were heated at 60 ºC and then cooled. 2ml $HClO_4$ was added and then heated again at 60 ºC till white dense fumes were made.

Quality Control

In each analytical batch, at least two reagent blanks, one spike and three duplicate samples were included in the acid digestion to assess the accuracy of the chemical analysis. The recovery of spike was 89.6 % (n = 6). The precision of the analysis was also checked by certified standard Geological reference materials Japan such as JSD1 and JSD2 were used to verify the results for As in soil.

Statistical Analysis

Statistical analysis was carried out from the acquired datasets using SPSS software version 20. Correlation between water, soil and different parts of rice and Linear Regression for future simulation was conducted using XLSTAT-Pro v7.5.2-2014

Results And Discussion

Arsenic Concentration In Water, Soil And Plant

Table 1 shows the mean arsenic concentration in water, soil, and different parts of plant. The results reveals that the mean arsenic concentration in the irrigation water in Site A, Site B, Site C and Site E is 52.8±6.45, 48.03±2.03, 38.31±10.35, 35.27± 6.8, 35.96± 10.1 (µg/l), respectively. This clearly indicates that all water samples are well within the Food and Agricultural Organization (FAO) limits of 100 (µg/l) for irrigation water, however, the mean concentration of all water samples exceeds WHO limits of 10 (µg/l) for drinking water and in two sites (A and B) it exceeds Pakistan Standards and Quality Control Authority (PSQCA) limit of 50 (µg/l). Overall range of arsenic concentration was found in a range of 22.43 to 60.32 ppb in irrigation water of Tehsil Wazirabad. The result of As concentrations in irrigation water were low with those reported in, Pakistan (0.40-918µg/l), West Bengal, India (110-760µg/l), Bangladesh (79-436µg/l)(60-720µg/l), Nepal (5-1014 µg/l)) and in India (0.5-250 µg/l)) (PCRWR, 2006),(Bhattacharya et al. 2009; Saha and Ali 2007; Dahal et al. 2008; Das et al. 2004; Norra et al. 2005)

Analysis of Irrigated rice paddy soil revealed that arsenic concentration in soil was found to be in the range of 2.0-7.29 mg/Kg in study area, which was below the maximum acceptable limit for agricultural soil of 25.mg/kg (Xiong et al. 1987). The concentration of arsenic in agricultural soil of current study were in accordance with previous researches conducted in West Bangal (1.38-12.27mg/Kg) (Bhattacharya et al., 2009), in Banladesh (1.5 - 3.0 mg/kg (Saha and Ali, 2007) whereas were low reported in Nepal (5.8- 12.5 mg/kg) (Dahal et al., 2008) and West Bangel,india (38 mg/kg)(Norra et al. 2005).

Recently, debates exist; either the accumulation of arsenic in paddy soil is escalating by arsenic rich water is positive or negative. Mostly, scholars argue that increased arsenic concentrations in irrigated results in increased arsenic concentrations in soil. The studies of (Das, Sur, and Das 2008) and (Sarkar et al. 2012) demonstrate direct correlation between irrigating with arsenic-contaminated water and elevated soil arsenic levels. The present study also indicates that the agricultural soil of the study area accumulates arsenic due to excessive use of arsenic contaminated irrigation water; this is reflected by high arsenic concentration in irrigation water and soil in both Site A and Site B as compared to other sites (Table 1). Various factors show their influence in arsenic retention in paddy soil such as soil texture, relatively high phosphorous (P) and low iron (Fe) concentrations in irrigation water, distance from the irrigation inlet field to field, water distribution method and arsenic leaching out during monsoon flooding (Senanayake and Mukherji 2014)

Arsenic Accumulation In Different Parts Of Rice

Excessive and long-term use of arsenic contaminated irrigation water may leads accumulation of arsenic in soil paddy and eventually in the edible part of crop. Paddy rice is considered to be one of major source of arsenic for humans (Meharg and Rahman 2003;(Mondal and Polya 2008) because of its deposition in the topsoil from irrigation water and subsequent uptake in rice grain (Dittmar et al. 2007). Various studies on rice grain reported that grain is prone to arsenic contamination due to continuous use of arsenic contaminated water on irrigated rice paddies Berner et al. 2005 ; (Garnier et al. 2010);(Roberts et al. 2007). In present study, the contents of As in the grain of rice were generally low as compared to root and shoots i.e Root > Stem > Leaf > Grain. Graphical representation of the distribution of arsenic in different parts of rice plant cultivated in the District Wazirabadis is shown in Figure 2. The results showed that the arsenic accumulated mostly in the root of rice plant (Table 1 and Fig. 2).

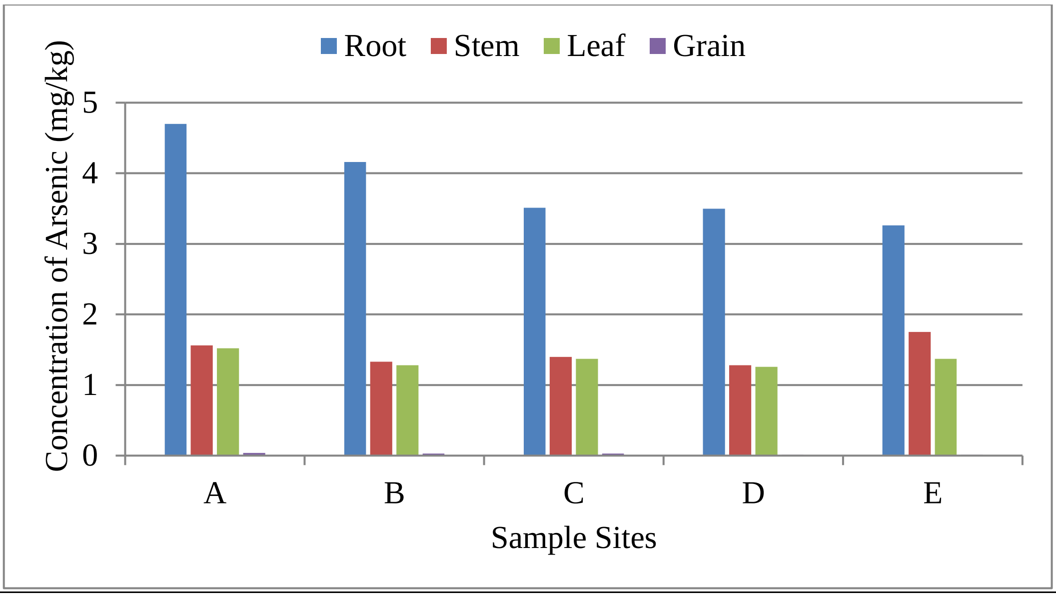

Figure 2 Distribution of arsenic in different parts of the rice plant in the study area

Table 1 Arsenic concentration in water, soil and rice parts of study area.

Matrices	**Site A**	**Site B**	**Site C**	**Site D**	**Site E**
	(n[a] = 5)	**(n[a] = 5)**	**(n[a] = 5)**	**(n[a] = 5)**	**(n[a] = 5)**
	Mean± SD	**Mean ±SD**	**Mean ±SD**	**Mean± SD**	**Mean± SD**
Water (µg/l)	52.8±6.45	48.03±2.03	38.31±10.35	35.27± 6.8	35.96± 10.1
	47.3-60.32	45.5-50.32	28.30-49.43	24.09-40.21	22.43-46.43
Soil (mg/Kg)	6.27± 0.56	5.31± 0.71	4.55± 1.8	3.78± 1.02	4.62± 1.98
	5.48-6.9	4.5-6.32	2.92-7.29	2.6-4.69	2-6.49
Root (mg/Kg)	4.7±0.63	4.16±0.23	3.51±0.82	3.50±0.92	3.26±0.96
	4.10-5.60	3.90-4.50	2.68-4.40	2.01-4.18	2.10-4.19
Stem (mg/Kg)	1.56±0.11	1.33±0.145	1.4±0.27	1.28±0.09	1.75±0.162
	1.39-1.7	1.23-1.59	1.10-1.81	1.13-1.35	1-1.68
Leaf (mg/Kg)	1.52±0.145	1.28±0.11	1.37±0.33	1.26±0.14	1.37±0.088
	1.32-1.69	1.19-1.47	1.09-1.91	1.07-1.41	1-1.48
Grain(mg/Kg)	0.047±0.008	0.036±0.0089	0.033±0.017	0.0123±0.0142	0.0019±0.0238
	0.039-.005	0.03-0.052	0.017-0.06	0.001-0.035	0.001-0.05

Previously Hasegawa et al. 2007 have reported that regardless rice varieties, accumulation of arsenic was 28 and 75 folds higher in root than that of shoot and raw rice grain, respectively. Although the actual mechanism of higher accumulation of arsenic in the rice root is still not well understood, but its uptake is reported. (Bae et al. 2002). Iron oxide generally present in rice roots decreases the uptake of arsenic towards plant tissue (Bae et al. 2002). The higher accumulation of arsenic in root (3.8364 mg/Kg) of the rice plant is followed by Stem (1.4076 mg/Kg), Leaf (1.3668 mg/Kg) and Grain (0.029712 mg/Kg). These results about arsenic in root, stem, leaf and grain were in good accordance with previous studies conducted in Nepal (Dahal et al. 2008) and Bangladesh

(Meharg and Rahman 2003), Itlay (Baroni et al. 2004)

The lowest concentration was find in edible part 'grain' with range of 0.001-0.06 mg/Kg which is well below permissible limit of FAO for rice grain (0.2 mg/Kg). Results had lower value than reported in Bangladesh (8-19.8 mg/Kg), West Bengal, India (0.25-0.73 mg/kg) arsenic in different parts of rice when irrigated with arsenic contaminated water. (Bhattacharya et al., 2009). As arsenic is mostly present in its reduced form in flooded paddy soil, resultantly it dissolved in the soil-pore water from where it easily accumulates in crop roots. (Senanayake and Mukherji 2014).

Arsenic Translocation from Water to Soil And Rice

The correlation coefficients (r) among arsenic concentrations in irrigation water, soil and in different parts (root, stem, leaf and grain) of the rice plant is shown in Table 2. It is interesting to note the fact that the arsenic content of the soil is significantly correlated with the arsenic content of irrigation water (r=0.582). This clearly indicates that the usage of arsenic rich irrigation water in the study area have a potential to contaminate the agricultural soil with arsenic. High significant correlation is obtained between arsenic concentrations in irrigation water and root (r=0.601). Rice is generally grown in water flooded field, due to which it is affected by arsenic. In flooded field topsoil is anaerobic and converts the arsenic in Free State which becomes available to plant roots. Significant correlation of the arsenic content in root with the arsenic contents in stem (r=0.738), with leaf (r=0.684) and with grain (r=0.720). Thus, from the results it can be concluded that the arsenic contaminated irrigation water and the agricultural field soil are highly responsible for the transfer and uptake of arsenic in rice plants. Similar trend was also reported by (Islam and Shamsad 2009) and (Roychowdhury 2008)in Bangladesh who showed that the arsenic contents in agricultural plants are related to the degree of arsenic contamination of water and soil .

Table 2 Correlation coefficients (r) among arsenic concentrations in irrigation water, soil and in different parts (root, stem, leaf and grain) of the rice plant cultivated in the Wazirabad.

Variables	Water	Soil mg/Kg	Root	Stem	Leaf	Grain
Water	**1**					
Soil	**0.582***	**1**				
Root	**0.601***	**0.842***	**1**			
Stem	0.210	**0.686**	**0.738**	**1**		
Leaf	0.192	**0.676**	**0.684**	**0.921**	**1**	
Grain	0.329	**0.728**	**0.720**	**0.622**	**0.577**	**1**

Values in bold are different from 0 with a significance level alpha=0.05
*Correlation is significant at the 0.05 level (1-Tailed)
**Correlation is significant at the 0.01 level (1-Tailed)

Future Threats to Sustainable Agriculture

As might accumulate in soil over time at different concentrations in irrigation water, assuming an annual water application of 1000 mm. A soil irrigated with 1000 mm of water containing 50 ppb As receives 0.5 kg/ha As per annum. The limited evidence at present suggests that the safe limit of soil As for rice lies somewhere in the range 25–50 mg/kg (Saha and Ali 2007; Duxbury et al. 2003). On basis of results of arsenic in irrigation water in present study we have assessed the potential affect of arsenic concentrations in irrigation water on soils for the coming years upto 2045. Linear regression model was used to predict the arsenic contamination of soil with the irrigation water contains Arsenic concentration of 50 ug/L. A correlation factor of arsenic addition to soil per 10 years was calculated from the study of (Brammer and Ravenscroft 2009)

According to Figure 3, arsenic concentration in paddy shows increasing trend with continuous irrigation with Arsenic contaminated water of 50 ug/L. The important point to note here is that in 2035, the arsenic concentration in soil will cross the maximum allowable limit of 25 mg/Kg and in 2045 it will be 147 mg/Kg, if irrigated with same quality of water. Although, actual soil loading rates will vary with the amount of irrigation water applied, As concentrations in the water and losses due to volatilization, leaching and crop removal. Because of the short period since As contamination of soils by groundwater irrigation was recognized, little information is available at present on As accumulation and removal rates. This model indicates As contamination of soil with irrigation water assuming the concentration of As in irrigation water constant (50 ug/L) for upcoming years but keeping in view the continues increase of As in ground water due to the use of fertilizer and pesticides the As content in soil would increase dramatically, which would possibly affects plant growth. Different studies have showed the reduction in plant growth due to arsenic contamination in soil which is irrigated with arsenic contaminated water. In the study of US, 80-90% reduction in rice yield has reported with high arsenic in soil (Yan et al. 2005) . In Faridpur, Bangladesh study of (Duxbury et al. 2003) on single rice variety (BR.29) reported rice yield reduction from 8.9 tones/ha at 26.3 ppm soil arsenic to 3 tones/ha at 57.5 ppm arsenic. In Bangladesh, glass house experiment showed decrease in content of chlorophyll a and b in rice leaf significantly ($p < 0.05$) with 60 to 90 ppm As, no one single rice plant survived up to its maturity stage. Significant correlation found between growth and yield which shows that effect on photosynthesis ultimately leads to reduction of rice yield Rahman, et al. 2007. In Pakistan, Rice meets more than two million tons of country food requirements. More than 45% of milled rice is consumed while rest is exported. It's export earns about US $ 933 annually from foreign exchange and contributes 1.3–1.6% of GDP and more than two million families' livelihood is associated with the crop economy (Manzoor et al. 2006).Therefore, this future simulation of As in soil (52.5 mg/Kg in 2035 and 147 mg/Kg in 2045) not only poses a future health risks to the population but also a matter of concern for the economy of the country. The distribution of Arsenic in soil in the investigated sites does not seems to be risk at a moment, however, to avoid the future up-heaval due to arsenic contamination in top soil as predicted by model (Fig.3), health effects on the local and international communities and fluctuation in rice exports in international markets, there is a need to take appropriate steps such as development of new irrigation strategies, use of high efficiency irrigation systems, continuous monitoring of irrigation water, soil and grain.

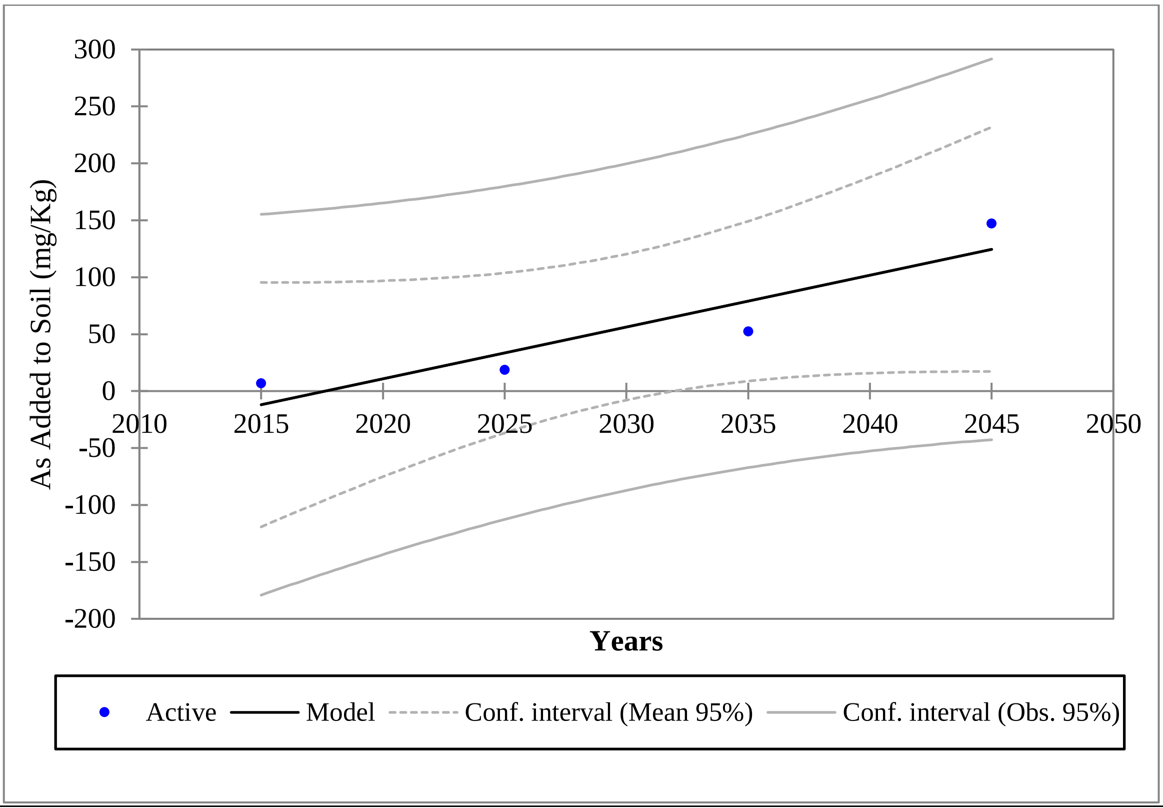

Figure 3 Regression of As Added to Soil (mg/Kg) by Years (R^2=0.8534).

Conclusions and recommendations

The level of Arsenic in irrigation water in the study area is very much above the WHO permissible limit of 0.01 mg/L for drinking water but within the FAO permissible limit of 0.10 mg/L for irrigation water. Soil gets contaminated with Arsenic due to irrigation with Arsenic contaminated water. Although, it is quite clear that the arsenic is transferred from irrigation water and paddy soil to various parts of rice plants (Roots, Stem, Leaf and Grain) with different pattern of distribution. In none of the studied samples the concentration of arsenic in soil exceeds the permissible limit of 25 mg/Kg, thus the exposure of the people living in the study area to arsenic is not a matter of an immediate concern at a moment but future projections of soil arsenic (52.5 mg/Kg in 2035 and 147 mg/Kg in 2045) cannot be ignored as it possess a serious future threats to human health and economy of the country. To avoid the future up-heaval due to arsenic contamination in soil and eventually in rice grain, health effects on the local and international communities and fluctuation in rice exports in international markets, there is a need to take appropriate steps such as development of new irrigation strategies, use of high efficiency irrigation systems and continuous monitoring of irrigation water, soil and grain.

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